NUREG/CR-2139 PNL-3786

Aerosols Generated by Free Fall Spills of Powders and Solutions in Static Air

Prepared by S. L. Sutter, J. W. Johnston, J. Mishima

Pacific Northwest Laboratory Operated by Battelle Memorial Institute

Prepared for U.S. Nuclear Regulatory Commission

> 8201070332 811231 PDR NUREG CR-2139 R PDR

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and

National Technical Information Service Springfield, Virginia 22161

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Manuscript Completed: December 1981 Date Published: December 1981

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Prepared for Division of Risk Analysis Office of Nuclear Regulatory Research U.S. Nuclear Regulatory Commission Washington, D.C. 20555 NRC FIN B2287

ABSTRACT

Safety assessments and environmental impact statements for nuclear fuel cycle facilities require an estimation of potential airborne releases. Aerosols generated by accidents are being investigated to develop the source terms for these releases. The lower boundary accidental release event would be a free fall spill of powders or liquids in static air. Experiments measured the mass airborne and particle size distribution of these aerosols for various source sizes and spill heights. Two powder and liquid sources were used: TiO_2 and UO_2 ; and aqueous uranine (sodium fluorescein) and uranyl nitrate solutions. Spill height and source size were significant in releases of both powders and liquids. For the source powders used (1 µm UO_2 and 1.7 µm TiO_2 , quantities from 25 g to 1000 g, and fall heights of 1 m and 3 m), the maximum source airborne was 0.12%. The maximum source quantities ranging from 125 to 1000 cc at the same fall heights).

The median aerodynamic equivalent diameters for collected airborne powder ranged from 6 to 26.5 μ m; liquids ranged from 4.1 to 34 μ m. All of the spills produced a significant fraction of respirable particles 10 μ m and less.

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INTRODUCTION

Aerosol generation characteristics of accidental particle release in nuclear fuel cycle facilities are being investigated at Pacific Northwest Laboratory (PNL). The work is sponsored by the U.S. Nuclear Regulatory Commission, Office of Research, Division of Risk Analysis. Safety analysis reports and environmental impact statements must evaluate the consequence of postulated accidents in or involving the facility in question. The dominant pathway to man is usually airborne release, so it is necessary to determine an aerosol source term, i.e., the quantity of material initially airborne from an accident.

Assessments of potential consequences are based on accident scenarios defining the sequence of largely inadvertent conditions that result in the airborne releases. Typical events range from spilling the materials in free fall through air to releases due to fire and explosion. Now it is difficult to make reasonable assessments, largely because information permitting such analysis is lacking or scattered in the literature. Our research has several directions: to define a range of accident conditions, to review available published information in order to assess the aerosol generation and behavior under these conditions, and to perform experimental studies to provide new data.

Setting upper and lower boundaries for accidental airborne releases is desirable, and it can be reasoned that a free fall spill in static air would be a lower boundary release event. Data to calculate the release from free falling powders and liquids were not found in a literature review. While the rate of fall of individual particles can be calculated, Bagnold (1941) noted that even a collection of similarly sized particles or grains (as in a spill) will not have the same rate of fall. Calculational techniques for these collections have not been developed. Therefore, experiments were performed investigating the characteristics of aerosols generated in free fall spills.

The objective of the experiments performed was to measure the mass and size distribution of particulates that become airborne as a result of free fall spills. The data will be used in ongoing work developing release models based on physical parameters.

As the problem of sampling aerosols generated by accident stresses is considered, many variables are identified. Some of the variables can be controlled: sampling methods, sampler location (both horizontal and vertical), sampling time, source size, and spill height. The first experiments were exploratory and identified the best experimental parameters for these variables. These experiments used uranine (sodium fluorescein) solutions and TiO₂ powder traced with uranine. Using these materials permitted a rapid analysis of the results. Radioactive materials, depleted uranium dioxide (DUO) powder and uranium in nitric acid solution (UNH) were used in a statistical matrix of experiments investigating the effect of source size and spill height on the airborne releases. These materials are surrogates for plutonium compounds, and are also found in nuclear fuel cycle facilities.

These experiments, therefore, measured releases of powders with two theoretical densities, 4.26 g/cc (TiO_2) and 10.76 g/cc (DUO), with similar mass median diameter particle sizes, 1.7 μ m and 1 μ m. Solution densities were about 1 g/cc (uranine) and 1.7 g/cc (UNH).

Using statistical procedures, we determined the experimental matrix for the source quantity and spill height, which are both significant in determining the airborne release.

General experimental procedures and results are discussed in the main portion of the report.

For ease of following the narrative describing the work, details of the experiments have primarily been confined to the appendices. Airborne mass measurements used in the analysis are in Appendices A and B. Appendix C describes the source materials and analytical methods; Appendix D, sampling. Probable sampling error caused by particle settling is discussed in Appendix E, and spill velocity, in Appendix F.

SUMMARY AND CONCLUSIONS

To provide data for use in developing models predicting accidental airborne releases, experiments were performed measuring aerosols generated by free fall spills of powders and solutions. Uranine simulants were used in preliminary experiments investigating sampling methods, sampler location, sampling time, spill distribution, and spill height. Evaluation of these experimental results led to selection of the most suitable parameters for investigations with radioactive materials, DUO powder and natural uranium solutions. Information on all of the work has been included in this report.

The mass airborne and particle size distribution of the aerosol generated by free fall spills was measured as a function of:

- source size
- spill height.

Material was placed in a container, then spilled, the total airborne portion measured, and the particle size distribution of the aerosol determined.

High-volume sampling was the optimum sampling method and was used in statistical matrices of runs using depleted uranium dioxide (DUO) powder and uranyl nitrate hexahydrate (UNH) solution. The abbreviations are used throughout this report to avoid confusion with uranine.

Three DUO powder masses: 100, 500, and 1000 g; and three UNH volumes: 125, 500, and 1000 cc; were spilled from 3-m and 1-m heights. Spills were made in the Radioactive Aerosol Release Tank (RART) with a volume of 20 m^3 --about the size of a small room. The total airborne material generated was collected by high-volume samplers, with the airborne particle size distribution measured using a cascade impactor.

Spill height and source size were both significant variables, with no interaction.

Weight percent of the source airborne can be used to estimate a release: 0.12% is the suggested maximum for powder, and liquid an order of magnitude lower.

Other observations were:

- Trends from the liquid spills were similar to the powder but at a lower level.
- Uranine solutions produced the finest and most homogeneous aerosol.
- UNH generated the aerosol with largest particles.
- All of the spills produced a significant fraction of particles 10 µm and less (considered respirable).
- Aerosol levels were higher in the lower portion of the RART.

Some suggestions for future work that would expand our understanding of aerosols generated by spills are included in this report.

EXPERIMENTAL

Experiments were performed to investigate the amount of airborne material and aerodynamic particle size of the aerosols generated by free fall spills of powders and liquids in static air. We were interested in determining the airborne mass generated during the spill event, rather than time dependent behavior. In these events we identified bounding conditions, i.e., the lower and upper limit of the airborne release. Midpoint values were measured, but a study of a wide range of conditions was not within the scope of this work. The experiments were limited by the equipment available and planning decisions. Since indoor releases are of interest, spills were performed in a room-size enclosure.

Exploratory experiments using uranine tracers and a matrix of runs with radioactive uranium materials were completed. Uranium was used because it is a material found in nuclear fuel cycle facilities, and depleted uranium dioxide is a standard PuO₂ simulant. The matrix was used to investigate source quantity and spill height effects with no air flowing. Although the spill took place in still air, the spill itself and the required sampling disturbed the static air.

EXPLORATORY EXPERIMENTS

When the problem of accidental releases is considered, even the simplest event will have many variables. A list of variables associated with free fall experiments was compiled. The variables were so numerous that some screening was desirable. Exploratory experiments were performed using uranine (sodium fluorescein) tracers: uranine is readily soluble in water and measureable by fluorometric means, so simple, rapid in-house analyses of the samples were performed, as ally on the same day an experiment was performed.

The expectments are described briefly in this section, but a more complete description of the sampling is included in the appendix section.

Experimental Procedure

The general procedure for conducting an experiment was to spill the source material--powder or liquid--from a beaker and sample the airborne material for a designated time. Once the beaker was overturned, $\binom{(a)}{a}$ all of the source material dropped from the beaker, falling and splashing as it hit the floor of the room-size enclosure.

RART

Spill experiments were conducted in the Radioactive Aerosol Release Tank (RART), a room-size enclosure. It is a stainless steel tank approximately 3 m high and 2.9 m in diameter, with a volume of about 20 m^3 . The interior of the RART is shown in Figure 1. An impactor and high volume samplers are visible in the foreground, and a one-liter beaker containing the aerosol source material can be seen near the ceiling of the RART.

Spill Description

Each run consisted of a single spill. The center of the RART was selected for the spill in order to minimize aerosol loss by deposition to the walls, hopefully giving a more uniform aerosol and better interpretation of results. The height of the RART limited the maximum spill height to 3 m. One meter was selected as a lower spill height.

Experimental Materials

An appropriate source requires sufficient material to generate a measurable release and to simulate a real situation. Safety considerations, however, limited the maximum amount of material suitable for use.

⁽a) The overturning of a beaker insured nearly instantaneous release of the powder. Other schemes considered, such as an instantaneous guillotineopened bottom of a bin, can have problems. The sticky powder types such as DUO have difficulty in flowing immediately and the guillotine blade shear forces disturb the powder itself.



Liquid solutions of uranine in water, 1 g/l and 10 g/l, in volumes from 50 to 1000 cc, were used for these exploratory experiments. Titanium dioxide was traced with uranine and used as a powder source in weights ranging from 25 to 1000 g. The materials are fully described in Appendix C.

Samplers

Filtration, pulling air through a porous material to separate suspended particles from a gas, is the most widely used method for collecting particulate matter, and we considered it suitable for our anticipated range of experiments. Open-face 47-mm filter samplers, fitted with polycarbonate membrane filters with 0.6 μ m pore size, were used. Cascade[®] impactors (28 ℓ pm) collected samples for calculation of the particle size distribution.

Sample Collection Location

Two vectors were available for sampler locations: vertical distance from the floor and horizontal distance from the center of the vessel which was the point of spill impact. Several locations were used, and experiments investigating uniformity of aerosol distribution were included.

Length of Sampling Time

Since particles released in the "accident" will be airborne for a finite time, our sampling method extracted nearly all the airborne material.^(a) Several different sampling times were investigated to accomplish this and to give some information on aerosol depletion during sampling.

MATRIX OF EXPERIMENTS

An appropriate sampling system was arrived at through exploratory experiments while varying the source quantity and spill height. In addition to TiO_2 and uranine solutions, low-level radioactive materials in the form of a 1 μ m, mass median diameter-depleted uranium dioxide (DUO) powder, and uranyl nitrate hexahydrate (UNH), a nitric acid solution, density 1.7 g/cc.

[®] Andersen Samplers, Inc., Atlanta, Georgia.

⁽a) See Appendix E for apparent sampling errors caused by particle settling.

Samples were collected for 30-minutes. After this sampling time, the samplers were turned off and a second set of two samplers were uncovered and run for an additional 30 minutes. This method confirmed that 99% of the mate-rial was collected in the first half hour.

Sampling

The sampling system, shown in Figure 1, used four high-volume samplers and one high-volume cascade impactor, each equipped with appropriate glass fiber filters. The 8-in. x 10-in. glass fiber filters are 99.9% efficient for 0.3 μ m particles. The sample rate was 1.4 m³/min for 30 minutes. As shown in the spill schematic in Figure 2, particle-laden air is drawn into the sampler; the particles are removed by the filter; and clean air exits through the back of the filter. High-volume filter samplers were placed at the 1-m and 2-m level, an impactor and background samplers at 1.5 m.





We were concerned that the large airflow exiting from the samplers into the RART would cause resuspension of deposited material. However, uraninetraced TiO₂ powder placed on the RART floor, followed by a sampling run, indicated this was not a problem. The airflow effects are mitigated by the jets at the rear of the sampler striking the RART wall.

Spills of 1000 g Ti0_2 powder and 1000 cc water indicated contamination would cause a less than 1% error.

Experimental Design

The experimental matrix was designed for Analysis of Variance (AOV) techniques. AOV divides the total variation of the Jependent variables (mass airborne here) into the variation from:

- factors under investigation
- interaction between factors under investigation
- experimental error.

The variation of the factors under investigation is compared to the experimental error to reach a conclusion regarding uncertainty in the results.

Wall Deposition

Foil squares were placed on portions of the RART walls to collect aerosol deposition samples. They were analyzed only for the uranine experiments because of the cost of uranium chemical analysis. Our work focussed on measuring airborne material. Wall deposited material, while of interest, would not become airborne and would not contribute significantly to the final results. The presence of the walls can have two effects. First, the wall shortens the trajectory of the initial impact, possibly eliminating the added shear stresses on these particles that could have caused more airborne fines. Second, the impact itself probably created fines that might not be otherwise generated in the unrestricted path. These two effects are counterbalancing and their net contribution is likely to be small. However, future experiments for minimizing impact stress are discussed later.

RESULTS AND DISCUSSION

The exploratory experiments were used to help define experiments using DUO and UNH. They provided valuable information on spill releases and are included in the following discussion as are results of the high-volume sampling of both uranine tracers and uranium experiments.

EXPLORATORY EXPERIMENTS

Height and Source Volume Effects

Initial release measurements from liquid spills were made using particle collection on 47-mm filters at three levels in the RART. Release calculations required three assumptions: 1) uniform radial aerosol distribution in the RART, 2) a sample representative of all aerosol distribution at that height, and 3) straight line variation with height.

The average calculated weight percent airborne showed higher release levels in the lower portion of the RART, as shown in Table 1.

Spill height and source size were significant variables, and the mass collected decreased with sampler height. The weight percent airborne from the one-meter spills of 500 cc were about an order of magnitude less than 3-m spills. As might be anticipated, less of the aerosol generated reached the

		3-	m Spill	
Source Volume, cc	0-61 cm	61-152 	152-244 cm	244-Ceiling cm
500	0.0027	0.0019	0.006	0.0004
50	0.0014	0.0012	0.0056	0,00005
			1-m Spill	
Source Volume, cc	0-61 cm	61-152 cm	152-244 cm	244-Ceiling cm
500	0.0001	0.00009	0.00002	0.000005
50	0.0007	0.0005	0.0001	0.000004

TABLE 1.	Average Calcul	lated Weight	Percent Airborne
	as a Function	of Sample Ha	ight

upper levels of the RART. Fractional releases from the 50-cc spill reaching this level could be as much as two orders of magnitude less than in the lower portion.

Overall weight percent airborne values from the replicate runs are shown in Table 2. This allows comparison with the high-volume samplin used in later experiments.

These results were not considered conclusive because the 50 cc, 1 g/l, spills did not provide sufficient uranine for satisfactory results for either spill height. Some of the filter samples at upper levels were in the back-ground range; the lower detection limit is about 10^{-9} g/cc. This would be about 10^{-4} wt% airborne. This series of experiments seemed to cover the lowest measurable release levels for uranine in this sampling matrix.

Foil was placed on the RART wall to collect deposited aerosol, but samples did not reach measurable levels. They therefore did not contribute further to our analysis.

Time Effects

Solutions of 10 g/2 uranine were used for the initial time effects experiments, and all but one of the five samplers were started simultaneously, then shut off at successive time intervals. These experiments indicated required sampling times. The one with no airflow was a background sample. Although data from these runs were inconclusive, they did not contradict the conjecture that aerosol concentration became depleted with time.

The experiment was redesigned with the following sampling scheme: all samplers ran for 10 minutes at different time increments. Samples collected

TABLE 2. Overall Average Weight Percent Airborne from Spilis of 1 g/& Uranine Solution

Source Volume,	Weight Percent	Airborne	
CC	3 m	1 m	
500	0,0056	0.0003	
50	0.0030	0.001	

represented the airborne mass at 0- to 10-, 10- to 20-, 20- to 30-, and 30- to 40-minute segments of two runs. One run was sampled at 90 to 100 minutes and another at 180 to 190 minutes.

As might be anticipated, the airborne concentration decreased with time. The majority of the mass, about 70%, was collected in the first 20 minutes of a run. At 90 minutes, 4% of the material sampled in the first 10 minutes was still airborne. A collection of 3.13×10^{-7} µg contrasted to an earlier 7.49 x 10^{-6} µg uranine. At 180 minutes, the aerosol was depleted, as indicated by background levels (readings the same as water) of fluorescence on the filter. This value could represent less than 1% of the material collected.

The TiO₂ powder traced with uranine also showed a rapid depletion of the airborne concentration, which was less pronounced at the higher levels in the RART. At the lowest level, 98% of the material was collected in the first 20 minutes, at the upper levels, 96%. Much less aerosol reached this level; the total collection at 244 cm above the RART floor was one-third that collected at 61 cm.

Time effects are not the primary thrust of this portion of the accident aerosol release study, but aerosol depletion with time must be considered when planning sampling. Our experiments were not continued beyond confirmation of aerosol depletion.

Location Effects

An experiment evaluating the location effect of five side-by-side filter samplers at three levels led to these conclusions:

- Sampling level has a significant effect: the lowest level (0.6 m) had greater uranine collection than the other two levels (1.5 and 2.4 m).
- There is no clear separation among the radial position averages.
- The standard deviation decreases with level, indicating a more homogeneous aerosol at the upper levels.

The experiments defined the distribution in all sectors of the RART. The average collection from the eight samples in the lower and upper portion of the RART for replicate spills of two liquid (500 cc) and two powder (450 g) is shown in Table 3.

The liquid aerosol results indicate uniform aerosol distribution in all positions of the RART. The powder showed a pattern with a tendency for more of the material to become airborne in one quadrant. (Note the large standard deviation.) Any powder sampling, therefore, required collecting all airborne material to remove this bias.

The calculated total weight percent airborne from this preliminary work was: powder, 0.003; liquid, 0.0003.

Particle Size

Particles generated by the solution spills were fairly small with the average airborne mass median diameter 2.8 μ m for this preliminary work. Collection height was not identified as an important variable for particle size distribution with the impactor sampling.

TABLE 3.	Average Sample and Upper RART	Collection in Lowe		
	Run 1, µg	Run 2, µg		
Liquid				
Lower Upper	0.474 ± 0,19 0.420 ± 0.18	0.8175 ± 0.14 0.773 ± 0.12		
Powder				
Lower Upper	8.598 ± 8.24 2.943 ± 1.41	6.109 * 5.13 2.850 * 2.30		

MATRIX OF EXPERIMENTS

Results of uranine runs using high-volume sampling and the DUO/UNH statistical matrices are both included in this section to enable comparison between different powders and solutions. All of the samples had adequate collection for analysis, and sample contamination was not a problem.

Powder Spills

While pouring the initial spill might appear asymmetrical, the powder impacted in the center of the RART, as Figure 3 shows, after a 1000-g DUO spill from 3 m. Powder radiates from the center and at some points reached the edge of the RART. With center impact, minimal wall deposition would be generated. This photo illustrates the randomness of an impact event, so variable results were anticipated. Figure 4 shows the technician removing a filter, with the particle collection visible.

The average powder mass airborne from the TiO₂ and DUO spills is listed in Table 4 and plotted in Figure 5. The plot shows the amount of airborne powder increasing with increased source mass.

More powder was collected on samplers in the lower portion of the RART, and, as in the exploratory experiments, the aerosol was more homogeneous in the upper portion.

	3	m	1 m		
Source, g	Ti02, 9	DUO, g	Ti0 ₂ , g	DUO, g	
1000	1.2230	0.7129	0.3363	0.0665	
500		0.3598		0.0250	
450	0.3835		0.03525		
100	0.1092	0.0399		0.0052	
25	0.0203		0.0035		

TABLE 4. Average Powder Airborne from Spills of TiO₂ and DUO







FIGURE 4. Sample Collection after a 1000-g DUO Spill from 3 m



FIGURE 5. Powder Mass Airborne Resulting from a Free Fall Spill in Static Air, as a Function of Source Size

The height of the spill, 1 m or 3 m, had a more pronounced effect than the quantity of DUO in the spill.

There was a time effect: later runs had higher levels of sampled DUO than would be expected. Eighty-five percent of the residual error in a run was indicated by time of occurrence. This indicates the possible influence of some uncontrolled parameter(s). Gravimetric analysis revealed this trend, later confirmed by chemical analysis. One possible explanation is suggested by the measurements of initial air temperature made with each run. Relative humidity and temperature both fluctuated, with no pattern for relative humidity and temperature showing a trend to lower values for the later runs. Replicate runs were made at temperatures 2° to 6°C lower than the first set, with the last experiments showing the largest variation.

We can reasonably assume that, consistent with known particle behavior, the denser air at lower temperatures would support more fine particles airborne. This cannot be quantified on the basis of these experiments since the results involve other considerations. A few of these include: increasing air temperature in he RART during sampling, effects of air displacement by the powder front, particle breakup and subsequent orientation during fall.

The statistical behavior of the four high-volume samplers was essentially the same based on individual analysis of variance for each sampler. The impactor sampler, with less airflow, was less efficient than the high-volume samplers.

Source Volume Correlation

When making a safety assessment, source mass is usually used for estimating releases, so our experiments were designed to measure releases as a function of mass. It can be speculated that the aerosol generated from a powder spill should be related to source volume rather than mass. Therefore, to help understand spill mechanisms, the volume relationship will be considered.

The powder volume (V) is related to mass (M) and density (D): V = M/D. Several densities would be available to consider: theoretical, bulk (or pour), or tap density. Since the experiments used a mass of powder poured loosely into a beaker, this "pour" density was used. The bulk powder density was determined by repeated weighings of powder poured into a known volume. The experimentally determined pour density was 0.63 g/cc for TiO₂ and 1.5 g/cc for DUO. These densities were used to estimate the powder volume, mass/density.^(a)

(a) We do note that both theoretical and pour-density ratios are nearly identical $\left(\frac{P_{DUO}}{P_{TIO_2}}\right)_{th} = \frac{10.76}{4.26} = 2.53$ and $\left(\frac{P_{DUO}}{P_{TIO_2}}\right)_p = \frac{1.5}{0.63} = 2.38$.

A logarithmic transformation of the results was made and plotted in Figure 6. There appears there could be a correlation with source volume, but this was not investigated further.

Blank Runs

Titanium dioxide spills indicated that contamination between runs would not contribute significantly to release measurements. The material collected was in the range of TiO₂ interferences in the DUO chemical analysis (blank runs). Background aerosol samples after the first half-hour showed a maximum of about 1% error could be contributed from this source.

Liquid Spills

Aerosols generated by liquid spills increased with increasing source volume as shown in Figure 7, and the total mass airborne is listed in Table 5. Uranine solutions, except at the lowest levels, produced greater mass airborne release than comparable volumes of UNH. The more dense, more viscous UNH solution apparently requires greater stresses for breakup than the less viscous uranine solutions.

In the majority of spills, a beaker was tipped and required 0.64 sec for spilling. The liquid was released in two uranine solution spills by pulling a plug from the container bottom, requiring 1.69 sec for the spill. Both releases had the same amount of material airborne.

The aerosol was more uniform than the powder spills and, as with powder, tended to collect more in the lower portion of the RART.

There was no time effect for liquid spills as there was for the DUO spills. Temperatures varied less ($1^{\circ}C$ to $3^{\circ}C$) than the powder runs. The response pattern of the liquid spills is very similar to that of the powder spills, but is at a lower level.



FIGURE 6. Powder Airborne from a Free Fall Spill in Static Air, Mass/Pour (Bulk) Density





TABLE 5. Average Total Calculated Mass Airborne from Uranine Solution and UNH Spills

	3	m	1 m			
Source,	Uranine, g	UNH, g	Uranine, g	UNH, g		
1000	0.1036	1.895×10^{-2}	3.0374×10^{-2}	1.945×10^{-3}		
500	0.020	1.13×10^{-2}	2.913×10^{-3}	1.22×10^{-3}		
125	4.147×10^{-3}	3.465×10^{-3}	7.768 x 10 ⁻⁴	7.5×10^{-4}		

WEIGHT PERCENT AIRBORNE

These experiments measured the mass and size of particulates airborne as a result of stresses imposed by a free fall spill. As such, they could be used to estimate accidental releases as required for safety assessments. The weight percent of a source airborne is a direct method used to define releases, so our data for powder is presented in this manner in Table 6 and plotted in Figures 8 and 9.

Air can only support a finite mass of airborne particles--a "saturated" condition--so it might be assumed that the weight percent of the source airborne in an enclosure would be larger for a small source than for a larger one. We did not find this, possibly because of source amount and enclosure volumr limitations. Also, the stresses were probably too small to accomplish saturation. Therefore, as expected, weight percent airborne was affected by changes in the source size. For example, the 1000-g TiO₂ source was 10 times larger than the 100-g source and had about 10 times more powder airborne, which calculates to the same weight percent airborne in both cases.

While it appears from the plot that the weight percent airborne from the DUO could be less than TiO_2 , this is inconclusive because of variability in the replicate experiments.

We suggest, with limited information, that the 0.12 wt% airborne be used in estimating powder releases at an unknown height.

	11000 11	onder opr	115		
	3	m	1 m		
Source, g	Ti02	DUO	Ti02	DUO	
1000	0.12	0.07	0.034	0.007	
500		0.07		0.005	
450	0.09		0.008		
100	0.11	0.04		0.005	
25	0.08		0.014		

TABLE 6. Average Weight Percent Airborne from Powder Spills



Liquid releases from the same source mass, expressed as weight percent airborne, were an order of magnitude lower than powder releases. The weight percents airborne are listed in Table 7 and plotted in Figure 9. Volume is used for the plot, but calculations were made on a mass basis. Therefore, 1000 cs of UNH with a density about 1.7 g/cc would weigh 1700 g; 1000 cc of uranine, with a density of 1 g/cc would weigh 1000 g. The uranine results from the 3-m spill compare well with exploratory experimental calculations.



UNH weight percent airborne decreased as a function of source volume, while the uranine increased. Increasing the uranine source volume further might increase the weight percent airborne. The airborne mass concentration from the maximum uranine spill could be estimated as 5 mg/m^3 . Estimation of the maximum airborne for liquids has been made at 10 mg/m³ (Oak Ridge National Laboratory 1970), the concentration of fogs or mists. Increased accident stresses, i.e., larger source, might conceivably increase the amount airborne.

ABLE 7.	Average	Weight	Percent	Airborne	from
	Uranine	and UNH	Soluti	on Spills	

Source, cc	3 1	n	1 m		
	Uranine	UNH	Uranine	UNH	
1000	0.01	0.001	0.003	0.0001	
500	0.004	0.001	0.0005	0.0001	
125	0.003	0.002	0.0006	0.0004	

However, on the basis of the present investigations, a 0.01 wt% airborne is the value suggested for liquid spills (density 1 g/cc), and for UNH spills we suggest a lower value of 0.002 wt%.

Since the weight percent airborne from liquids is an order of magnitude less than powder, they would be of less concern in an accident situation.

PARTICLE SIZE DISTRIBUTION

Powders

Some conclusions can be reached for the powder results, shown in Table 8, even with variation in particle distribution between and within runs. The geometric standard deviation range for TiO_2 was 2.8 to 7.7; DUO, 2.6 to 50. Particles generated by a spill from a larger source mass tended to be larger (seen in Figure 10). When a small source spills, there is more opportunity for air-particle interaction, and shear stress forces act to disperse the powder as it falls.

> TABLE 8. Average of the Median Aerodynamic Equivalent Diameter Particle Size Generated by a Free Fall Spill of Powder in Static Air

Source Mass, g	Particle Size, µm					
	3	m	1	m		
	TiO	DUO	Ti02	DUO		
1000	26.5	20.8	12.8	10.3		
500		11		6.4		
450	15		16			
100	20	6		6.1		
25	9.7		6			





The airborne DUO Particles from the larger sources were about two times larger for the 3-m spill than those generated by the 1-m spill. This could be a function of the amount airborne or an artifact of the sampling. More large particles are entrained above the samplers for the higher spills with greater releases. Hence, more of these are caught before settiing. Airborne particle size is plotted versus powder mass airborne in Figure 11. This plot is not intended to suggest that the former is caused by the latter. They both are dependent variables and agglomeration is minimal in these experiments. The geometric standard deviations (σ_q) of the particle-size distribution within a run were large, indicating a wide range of particle sizes. In some cases there was a wide variation in replicate run distributions. Another consideration was that in reporting only one particle size for a run, we fail to show changes in the particle size during a run. Size distributions will shift to smaller sizes since the larger ones settle faster, as one UNH solution spill illustrated. The spill was inadvertently sampled after the first half hour had elapsed. The AED was 4.2 µm compared with the replicate run AED of 34 µm collected in the first half-hour of a run. The particle sizes plotted in Figures 10 and 11 are probably agglomerates since the source powder AED was DUO, 3.3 µm; TiO2, 3.5 µm.




Liquids

100

UNH solutions generated the largest particles, 34 µm diameter, for either powder or liquid spills as seen in Table 9 and plotted in Figure 12. These solutions are more viscous than water, requiring greater shear stresses to break up the solution. As less mass was airborne, however, the UNH showed the trend to smaller particle size, as shown in Figure 13. This might be a surface tension effect which will be treated later.

Uranine/water solution spills generated primarily small particles 10 μ m and less in diameter. The aerosol size distributions showed a σ_g range of 1.8 to 3.5 indicating a more uniform distribution than other sources. The amount airborne did not show the trend to smaller particle size as less mass was airborne.

TABLE 9. Average of the Median Particle Size Airborne Generated by a Free Fall Spill of Liquid in Static Air

		Particle	Size, µm	
Source,	3 1	n	1 m	
Volume, cc	Uranine	UNH	Uranine	UNH
1000	6.6	33	8.6	24.8
500	10.8	34(a)	6.0	15
125	8.2	25	4.2	4.1

(a) Sample from Run 6 not used, experimental error.





Particles 10 µm and Less

Particle size information from this study was obtained to estimate the fraction which would be respirable and therefore of radiological consequence. Because practically all particles 10 μ m (AED) and larger when inhaled are deposited in the naso-pharyngeal region, particles larger than 10 μ m have much less radiological significance than do smaller particles that are deposited preferentially in the tracheo-bronchial and pulmonary regions. Other factors



FIGURE 13. Median Aerodynamic Equivalent Diameter Particle Size Versus Liquid Mass Airborne

of particle solubility, clearance rates, and breathing mode also determine the significance of inhaled particles. Detailed characterization of airborne particles may still not permit a definitive statement of hazard because of the remaining uncertainties in the biological response. Radiation protection guides for airborne radioactive particles are based on generalized assumptions regarding particle size retention and clearance rates from the lung and body and do not account for all parameters in specific situations. As seen in Table 10 and 11, a large fraction of the mass of airborne particles was associated with the less than 10 µm AED size.

TABLE 10.	Average	Percent of	Particles	10 µm
	and Less	Generated	by Powder	Spills

Source	3	m	1 m		
Mass, g	Ti02	DUO	Ti02	DUO	
1000	33	37	46.5	48	
500		54		67.5	
450	42		41.5		
100	38.5	67.5		72	
25	50		60		

TABLE 11. Average Percent of Particles 10 µm and Less Generated by Liquid Spill

Source,	3 m		1 m	
CC	Uranine	UNH	Uranine	UNH
1000	49	20	53	38
500	66	16	62	58
125	58	31	72	62

WALL DEPOSITION

Wall deposition samples were collected on eight 1-ft squares of aluminum foil and the total calculated. The samples were erratic--undetectable for many releases--while others were high because of visible splashing. Only uranine samples were analyzed. Since we are interested in airborne material, and the chemical analysis of the extra DUO and UNH samples was not cost effective for the results, they were collected but not analyzed.

Results showing the average percent of the total of estimated mass and wal? deposition are listed in Table 12.

The standard deviation between individual foil samples in a run was about the same as the sample collection, indicating a high degree of variability. Trends identified were:

 More wall deposition appeared (or was evident) in the lower half of the RART.

Source	3 m	1 m
Powder, g		
1000	7.9	2.6
450	12.5	
100	14.5	12.3
25	6.9	ND
Liquid, cc		
1000	27.6	ND
500	23.6	ND
125	ND	ND

TABLE 12. Average Percent of Collection Deposited On Walls

- One-meter spills generated little, if any, wall deposition.
- When deposition occurred, a higher percentage of liquid aerosol deposited than powder, although powder deposited on more wall samples.

Optical microscope examination of powder particles deposited on the wall gave a physical diameter of 16.6 μ m and a σ_g of 1.27 as a rough estimate of the geometric diameter by count. Using the formulation (Stockman 1977)

 $\log d_{\rm qm} = \log d_{\rm qc} + 6.908 \log^2 \sigma_{\rm g}$,

where

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 $d_{gm} = geometric median by mass$

 d_{gc} = geometric median by count

 $\sigma_{\rm g}$ = geometric standard deviation,

a mass median diameter of 20 μm was estimated, calculating to an aerodynamic equivalent diameter 41 μm .

FUTURE WORK

The experiments reported here are the initial work in our program investigating accident-generated aerosols. They are of the simplest type: a single spill event in static air. As such, they are the first step in understanding accident-generated aerosols and in suggesting other research areas. These will be discussed in the following section, with options for experimentai investigation.

Spills are considered lower boundary release events; pressurized releases are considered upper boundary. The logical sequence for our study would be to examine pressurized releases experimentally. Work has therefore begun to generate pressurized powder releases as a function of powder source mass and chamber pressure.

After these runs, further study would be useful, but with a caveat: the spill itself may be such a random event that subtle effects may not be distinguishable over the randomness of an event. Some potential areas of research are suggested and proposed in the following section.

Source particle size distribution effects could provide useful information. Quality assurance requirements specified a maximum particle size of 75 μ m for cur DUO. There can be variations of size distribution between batches of powder yet still remain within specifications. These distributions can vary, depending on conditions of manufacture, for example, sintering temperature. Experiments reported here used a very fine, 1- μ m MMD powder. Another DUO powder with a 9- μ m MMD is available and can be used in experiments comparing releases from the same type of powder with a different size distribution.

Other powder materials have been suggested for experimental use in an attempt to identify density effects. While it would be useful to spill other powders, if a significant difference in release occurs the question is: can it be attributed to density rather than humidity, surface effects, morphology, or other powder properties? The appropriate density for correlation must also be considered: theoretical (handbook), apparent (bulk) or tap density. Bulk density of the powder as it is spilled would seem to be the most realistic.

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Variables affecting this loose powder density have been identified (Hausner 1967): material type; m 'erial density; particle porosity, size and shape; particle shape distribution, specific surface, oxide films, additions; and medium surrounding the particles. Powder density can also be related to manufacture, e.g., DUO temperature determines the oxygen content thus varying density. Pour density of experimental powders would be determined, and mass density correlations investigated further. If no difference in releases is observed, this would also be useful information in that we could extrapolate our results to a wide range of powders.

Enclosure size limited the extent of these experiments. It would be desirable to spill powder from 4 m or 5 m, which would represent additional realistic situations. A larger enclosure could also investigate the release as a function of distance from the impact area. It would be assumed that releases generated by a spill would remain close to the impact area.

Source quantities used in these experiments were limited by safety and cleaning considerations associated with radioactive DUO. We concluded that the weight percent of a source powder airborne does not increase with increases in the source for a specific spill height. This could be further confirmed using traced TiO₂ powder. Multiple simultaneous spills could increase the total release. The limiting weight percent of uranine solutions airborne was not defined by these experiments. This should be accomplished by a series of experiments increasing the source volume spilled.

Powders should be emphasized in future work, since liquid releases were an order of magnitude lower for about the same source size. In addition, the UNH solutions were very corrosive to equipment, causing rust in the samplers. This would be a potential contamination source in future experiments, so UNH should be used in restricted amounts only.

Another area of investigation mi," b variations in the spill mechanism considering an immediate spill or one occurring over a finite period of time. In liquids, spill type did not change the release. While flowing powder would appear to be an option, these powders do not flow, as determined in bin flow tests included in Appendix C. In these tests powder did not flow through the 3.2 cm² area because it requires a large opening before it will flow. Studies of nuclear fuel powder flow (Densley 1979) determined that a large diameter opening, 28 cm, was required before gravity flow occurred.

Another type of powder spill could be achieved by equipping a container with a vertical blade that could release powder through a guillotine-type break. This should not change the length of time required for a spill, but would change the powder frontage area exposed to aerodynamic entrainment. It is questionable whether this would increase releases for the masses or change the configuration used in these experiments.

Particles leave the powder stream as it falls, and again on impact with the floor. The relative magnitude of these releases could be studied experimentally by directing the spill into a pool of water. This would minimize impaction releases, and the powder leading edge releases can be compared to earlier work where both release mechanisms operated.

Spills into flowing airstreams is another potential study area. Since additional forces are expected in flowing streams, higher releases might be expected than from similar spills in static air.

There are many options for future investigation of spill-generated accident releases. These are in avaluated within the scope of the project. The project is not designed to be a comprehensive study of all parameters, but rather an empirical look at lower and upper boundary accidental airborne releases within a nuclear fuel cycle manufacturing facility. This means that only a few experiments will be identified to continue this initial work on lower boundary releases.

REFERENCES

- Bagnold, R. A. 1941. The Physics of Blown Sands and Desert Dunes. Methuen and Co., London, England.
- Collins, L. H. and A. C. Zook. 1979. "Application of a Direct Method for the Determination of Trace Uranium in Safeguards Samples by Pulsed Laser Fluorometry." Trans. Am. Nucl. Soc., Suppl., 3, pp. 12–13.
- Davies, W. and W. Gray. 1964. "A Rapid and Specific Titrimetric Method for the Precise Determination of Uranium Using Iron (II) Sulphate as Reductant," Talanta, 11:1203-1211.
- Densley, P. J. and J. W. Carson. 1979. Nuclear Fuel Powder Flow Characterization. HEDL-SA-1694FP. Hanford Engineering Development Laboratory, Richland, Washington.
- Hausner, H. H. 1967. "Friction Conditions in a Mass of Metal Powder." International Journal of Powder Metallurgy, 4:7-13.
- Oak Ridge National Laboratory. 1970. Siting of Fuel Reprocessing and Waste Management Facilities. ORNL-4451, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Robbins, J. C. 1978. Field Technique for the Measurement of Uranium in Natural Waters. C.I.M. Bulletin, 71:61.
- Schwendiman, L. C. 1977. Supporting Information for the Estimation of Plutonium Oxide Leak Rates Through Very Small Apertures. BNWL-2198, Pacific Northwest Laboratory, Richland, Washington.
- Stockham, J. D. and E. G. Fochtman. 1977. Particle Size Analysis. Ann Arbor Science Publishers, Inc., Ann Arbor, Michigan.
- Zenz, F. A. and D. F. Othmer. 1960. Fluidization and Fluid Particle Systems. Reinhold Publishing Corp., New York, New York.

APPENDIX A

AIRBORNE RELEASES OF POWDERS AND LIQUIDS RESULTING FROM A FREE FALL SPILL IN STATIC AIR

	Weight	Spill Height	Uranine Airborne	Ura Wall Dep	nine osition, g	Uranine Total	Percent Deposited	Calculated Mass Airborne	Total Mass Airborne and Deposition	Weight Percent	Weight Airborne and
Kun	g	111 	<u>g</u>	1 m	3 m	g	On Wall	9	9	Airborne	Deposition
37	471.9	3	2.85×10^{-3}	1.87×10^{-5}	2.9615×10^{-3}		3.8	0.9105	0.9462	0.19	0.20
38	450.4		4.38 × 10 ⁻⁴	3.44 x 10 ⁻⁵	1.10×10^{-5}	4.83 x 10 ⁻⁴	9,4	0.1475	0.1678	0.033	0.036
39	431.1		3.07 × 10 ⁻⁴	5.29 x 10 ⁻⁵	5.25 x 10 ⁻⁵	4.12×10^{-4}	25.6	0.0858	0.1151		0.027
42	460.0		1.17×10^{-3}	5.90 x 10 ⁻⁵	7.41 × 10 ⁻⁵	1.30 × 10	10.2	0.3900	0.4333	0.09	0.094
43	100		1.35×10^{-3}	1.97 × 10 ⁻⁴	1.18×10^{-4}	1.66×10^{-3}	18.7	0.1194	0.1469	0.12	0.15
44	100		1.01×10^{-3}	7.71 x 10 ⁻⁵	4.00 x 10 ⁻⁵	1.13×10^{-3}	10.3	0.0990	0.1108	0.099	0.11
4.7	1000		6.403 × 10 ⁻³	2.357 × 10	1.64×10^{-4}	6.803 × 10 ^{-3*}	5.9	1.480	1.5725	0.15	0.15
48	1003		3.742×10^{-3}	2.263 × 10 ⁻⁴	1.464×10^{-4}	4.1147×10^{-3}	9.1	0.9659	1.062	0.096	0.11
49	25		2.926 × 10 ⁻⁴	1.445×10^{-5}	2.95 x 10 ⁻⁵	3.3655×10^{-4}	10.8	0.0240	0.0276	0.10	0.11
50	25		1.855 × 10 ⁻⁴	ND	5.58 × 10 ⁻⁰	1.911×10^{-4}	2.9	0.01656	0.0171	0.07	0.08
45	441.1	- 1	1.48 × 10 ⁻⁶	7.45 x 10 ⁻⁵	1.21 × 10 ⁻⁴	1.68 x 10	11.6	0.0345	0.0391	0.008	0.008
46	451		1.81 × 10 ⁻⁴	1.59×10^{-5}	7.49 × 10 ⁻⁰	2.04 × 10 ⁻⁴	12.9	0.0360	0.0406	0.008	0.009
51	25		3.7882 × 10 ⁻⁵	ND	ND	3.7882×10^{-5}	ND	0.0030	-	0.01	
52	25		5.0123 × 10 ⁻⁵	ND	NŰ	5.0123×10^{-5}	ND	0.0040		0.02	
53	1000		2.2477 × 10-3	5.06 x 10 ⁻⁵	4,18 x 10 "	2.34 × 10 ⁻³	3.9	0.4984	0.5188	0.05	0.05
54	1000		1.0852×10^{-3}	1.39×10^{-5}	4.965 × 10-7	1.10×10^{-3}	1.3	0.1742	0.1766	0.017	0.018
56	1000		5.3340 × 10 ^{-b}			5.3348 × 10 ⁻⁶		0.0009			
57	1000		3.6031 × 10 ^{-b}			3.6031×10^{-6}		0.0006			

TABLE A.1. Airborne Releases of TiO2 Powder Traced with Uranine, Releases Resulting from a Free Fall Spill in Static Air.

(a) Calculate from 1 sq ft deposition sample. (b) Ceiling deposition 1.98×10^{-5} g not included.

TABLE A.2. Airborne Releases of 10 g/g Uranine Solution, Releases Resulting from a Free Fall Spill in Static Air

Run	Source Volume cc	Source Weight Uranine g	Spill Height m	Urani Airbo	ine orni	e	Estin Tota Depo	mated 1 Wall sition g	Percent Collected On Wall	Calcu Total Airbo	lated Mass orne	Weight Percent Mass Airborne
30	500	5	3	1.94	x	10-4		ND		1.94	× 10 ⁻²	0.0039
31				1.567	x	10-4	1.55	x 10 ⁻⁴	49.7	1.57	× 10 ⁻²	0.0031
32				1.534	x	10 ⁻⁴	6.79	× 10 ⁻⁵	30.8	1.53	× 10 ⁻²	0.0031
33				2.79	x	10-4	4.58	× 10 ⁻⁴	39.1	2.79	× 10 ⁻²	0.0056
34				2.55	x	10 ⁻⁴	3.02	× 10 ⁻⁴	15.4	2.55	× 10 ⁻²	0.0051
35				1.63	x	10 ⁻⁴	2.41	× 10 ⁻⁴	32,4	1.63	x 10 ⁻²	0.0033
40 ^(a)				2.04	x	10-4	4.31	x 10 ⁻⁵	17.4	2.04	× 10 ⁻²	0.0041
41 ^(a)				1.85	×	10 ⁻⁴	7.35	× 10 ⁻⁶	3.8	1.85	× 10 ⁻²	0.0037
55	1000	10		1.270	х	10 ⁻³	1.23	× 10 ⁻⁴	9.7	1.27	× 10 ⁻¹	0.013
58	1060	10	1	3.037	x	10-4		ND		3.0374	10-2	0.003
59	1000	10	3	8.0237	x	10-4	5.079	× 10 ⁻⁵	6.0	8.0237	× 10 ⁻²	0.008
60 ^(b)	125	1.25	3	2.00	x	10 ⁻⁴		ND		2.00	× 10 ⁻²	0.016
61	500	5	1	2.913	х	10 ⁻⁵		ND		2.913	$\times 10^{-3}$	0.0004
62	í25	1.25	1	7.7685	x	10-6		ND		7,7685	× 10 ⁻⁴	0.0006
63	125	1.25	3	4.147	x	10 ⁻⁵		ND		4.147	× 10 ⁻³	0.003

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(a) Spill initiated by pulling stopper(b) High release due to splashesND Not Detectable

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TABLE A.3.	Airborne Re	leases	of DU	0 Powder,	Releases	Resulting
	from a Free	Fall	Spill	in Static	Air	

DUO Weight, g	Spill Height, m	Total DUO Airborne, g	Weight Percent Airborne
1000	3	0.23122	0.023
100	1	0.00337	0.003
500	- 1	0.01924	0.004
100	3	0.00350	0.004
1000g TiO ₂	1	0.000100 ^(a)	
500	3	0.17579	0.04
1000	1	0.05589	0.006
100	3	0.04489	0.04
500	1	0.03081	0.008
1000g Ti02	3	0.000211 ^(a)	
100	1	0.00670	0.007
1000	1	0.07703	0.008
1000	3	1.19463	0.12
500	3	0.54383	0.11
	DU0 Weight, 9 1000 100 500 1000 1000g Ti0 ₂ 500 1000 1000 500 1000g Ti0 ₂ 100 1000g Ti0 ₂ 100 1000 500	DU0 Weight, gSpill Height, m100031000150011000g TiO2 5001100031000g TiO2 500110003100011000g TiO2 50031000g TiO2 100031000g TiO2 1000310001100035003100035003	DU0 Weight, gSpill Height, mTotal DU0 Airborne, gymg100030.2312210010.0033750010.0192410030.003501000g Ti0210.000100(a)50030.17579100010.05589100010.030811000g Ti0230.000211(a)1000g Ti0230.000211(a)100010.07703100010.07703100031.1946350030.54383

 (a) TiO₂ standard indicated 0.00011 g interference in uranium quality assurance tests. Therefore this collection is not significant.

Run	Source Volume, cc	Source Weight Uranium, g	Spill Height, m	Uranium Airborne, µg	Weight Percent Airborne	Total Mass Airborne(a), G
1	1000	208.7	3	2271.4	0.0011	1.85×10^{-2}
2	125	26.1	1	141.1	0.0005	1.15×10^{-3}
3	500	104.4	1	137.5	0.0001	1.12×10^{-3}
4	125	26.1	3	346	0.0013	2.82×10^{-3}
5	1000cc H ₂ 0		3	22.7		
6	500	104.4	3	1067.6 ^(b)	0.0013	1.12×10^{-2}
7	1000	208.7	1	223.2	0.0001	1.82×10^{-3}
8	125	26.1	3	514	0.0020	4.19×10^{-3}
9	500	104.4	1	161.5	0.0002	1.32×10^{-3}
10	1000cc H ₂ 0		3	42.36		
11	125	26.1	1	43.4	0.0004	3.50×10^{-4}
12	1000	208.7	1	254.0	0.0001	2.07×10^{-3}
13	1000	208.7	3	2378.2	0.0011	1.94×10^{-2}
14	500	104.4	3	1412.5	0.0014	1.15×10^{-2}

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TABLE A.4. Airborne Releases of 208.7 g/g Uranium Solution, Releases Resulting from a Free Fall in Static Air

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(a) Calculated using solution density of 1.7 g/cc.
(b) Q2 dropped and contaminated. Collection estimated at 313 µg would raise the value to 1380.6. The weight percent airborne and total mass airborne were calculated using this value.

APPENDIX B

MEDIAN AERODYNAMIC EQUIVALENT DIAMETER PARTICLE SIZE GENERATED BY A FREE FALL SPILL IN STATIC AIR

Run	Source Weight, g	Spill Height, m	Median Diameter, µm	σg	Percent of Collection 10 µm and Less	Weight Percent of Source Airborne 10 µm and Less
37	471.9	3	21.0	6.2	34	0.065
38	450.4		5.8	5.6	62	0.020
39	431.1		16.0	6.6	40	0.008
42	460		17.2	6.9	40	0.036
43	100		27.0	7.5	31	0.037
44	100		13.0	5.9	46	0.046
45	441	1	15.3	7.7	41	0.033
46	451	1	16.8	6.5	42	0.034
47	1000	3	36.0	6.7	25	0.038
48	1000		17.0	7.4	40	0.038
49	25		9.6	3.7	50	0.050
50	25		9.8	4.7	50	0.035
51	25	1	4.7	2.7	62	0.006
52	25		7.2	3.1	58	0.012
53	1000		9.1	2.8	53	0.027
54	1000		16.5	5.9	40	0.068

TABLE B.1. Median Aerodynamic Equivalent Diameter Particle Size Generated by a Free Fall Spill in Static Air, Traced TiO₂ Powder

Run	Source Volume, cc	Uranine Weight, g	Spill Height, m	Median Diameter, µm	σg	Percent of Collection 10 µm and Less	Weight Percent of Source Airborne 10 µm and Less
30	500	5	3	8.0	3.5	56	0.0022
31				11.0	2.8	44	0.0014
32				4.5	2.4	80	0.0025
33				4.8	2.6	82	0.0046
34				5.4	2.6	74	0.0038
35				4.8	3.0	70	0.0023
40				7.3	3.2	59	0.0024
41				6.8	3.1	64	0.0024
55	1000	10		11.5	3.1	45	0.0058
58	1000	10	1	8.6	4.1	53	0.0016
59	1000	10	3	10.0	2.6	50	0.0040
60	125	1.25	3	9.2	3.7	52	0.0083
61	500	5	1	6.0	2.1	62	0.0002
62	125	1.25	1	4.2	1.8	72	0.0004
63	125	1.25	3	7.2	3	63	0.0019

TABLE B.2. Median Aerodynamic Equivalent Diameter Particle Size Generated by a Free Fall Spill in Static Air, 10 g/& Uranine Solution

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Run	Source Weight, g	Spil: Height, m	Median Diameter, µm	σg	Percent 10 µm and Less	Weight Percent of Source Airborne 10 µm and Less
1	1000	3	3.1	5.1	25	0.006
13	1000		10.5	3.9	49	0.059
6	500		6.1	3.1	70	0.028
14	500		16	5.2	38	0.042
4	100		<0.1	50	91	0.004
8	100		12.0	3.2	44	0.017
7	1000	1	11.0	2.75	46	0.003
12	1000		9.6	3.2	50	0.004
3	500		3.2	3.5	83	0.003
9	500		9.5	2.6	52	0.004
2	100		0.65	5.2	93	0.003
11	100		11.5	2.9	51	0.004

TABLE B.3. Median Aerodynamic Equivalent Diameter Particle Size Generated by a Free Fall Spili in Static Air, DUU Powder

Run	Source Volume,	Spill Height, m	Median Diameter, µM	σg	Percent 10 µm and Less	Weight Percent of Airborne 10 µm and Less
1	1000	3	34	4.1	21	0.0002
13	1000		32	3.6	19	0.0003
6	500		4.2 ^(a)	2.2	99.4	0.0013
14	500		34	2.1	16	0.0002
4	125		20	7	36	0.0005
8	125		30	6	26	0.0005
7	1000	1	9.6	5	51	0.00005
12	1000		40	5.8	24	0.00002
3	500		2.3	1.9	85	0.00009
9	500		27	6.6	30	0.00006
2	125		3.2	2.3	62	0.0003
11	125		5.0	7.8	61	0.0002

TABLE B.4. Median Aerodynamic Equivalent Diameter Particle Size Generated by a Free Fall Spill in Static Air, 208.7 g/L Uranium Solution

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(a) Collected in second half of run, so the size

distribution is in smaller range, not used in analysis

APPENDIX C

SOURCE MATERIALS USED IN FREE FALL EXPERIMENTS

APPENDIX C

SOURCE MATTALS USED IN FREE FALL EXPERIMENTS

Two liquid solutions and two powders were used as sources in the free fall experiments.

Liquids

Uranine (sodium fluorescein) solutions were used in the first experiments. A 100 g/ ℓ uranine stock solution was prepared by dissolving the powder in 200 cc water and making to a one liter volume with ethyl alcohol. Aliquots of the stock solution were used to make the 1 g/ ℓ and 10 g/ ℓ dilutions used in our experiments. The density of this solution was about 1 g/cc.

Sample analysis was performed by fluorometry. Dilutions of the stock solution representing different concentrations were made and the results read on a fluorometer. The lower detection limit is about 1×10^{-9} g/cc. The results can be plotted and the sample unknown values read from this plot. For our work, equations for the plot at each of the four fluorometer ranges were formulated and used for calculating results.

The R^2 (variability) values, approaching 1, indicate a good fit at all attenuations, as Table C.1 shows. The 30X range is the most sensitive.

The second solution used was about 200 g/ ℓ uranium in nitric acid solution, density about 1.7 g/cc. It was made from a 510 g/ ℓ stock solution and the concentration determined by chemical analysis to be 208.7 g/ ℓ , using standard Davies-Gray (Davies 1964) uranium titration methods accurate to ±0.1%. Sufficient uranium solution for all the experiments, eight liters in all, was prepared in one batch, insuring source uniformity.

TABLE C.1. R² Values for Plots at Different Fluorometer Autenuations

 $R^2 = \frac{30X}{0.997} = \frac{10X}{0.999} = \frac{3X}{0.9998} = \frac{1X}{0.9997}$

C.1

Samples collected in an experiment were dissolved in 0.1 N HNO₃ and analyzed using laser fluorometry. This method employs a pulsed nitrogen laser to excite uranium in a solution containing a pyrophosphate reagent. The fluorescent signals are amr^{***}ied and integrated, and the results displayed on a meter. In evaluations at New Brunswick Laboratory (Collins 1979) precision and accuracy were within 5% error with a sensitivity of 0.005 µg uranium. Standards prepared by dotting a collection filter with dilutions of the bulk solution confirmed a recovery of 96% and better for our samples.

Powders

Titanium dioxide, TiO₂, was used in the first powder spills. For analysis of the collected samples, the powder was traced with uranine solution, and total powder mass airborne calculated from this content. An aliquot of the stock solution in ethyl alcohol was mixed uniformly and thoroughly with TiO₂ powder. The mixture was dried, ground in a mortar and pestle, and mixed again. For each run, four portions of the source powder were analyzed fluorometrically, and the results averaged to confirm the uranine source concentration. The maximum variability was about 6%.

Depleted uranium dioxide was the second powder spilled in experiments. These samples were also analyzed for uranium using laser fluorometry, since good correlation with other methods had been exhibited at high uranium levels. Process streams containing as much as 20 g/ ℓ uranium were analyzed using this method (Robbins 1978) and indicated it would be satisfactory for all samples. Samples were dissolved in a mixed acid (5N HNO₃, 0.1 N HF) each for analysis. An error of ±10% or less for our samples was established by preparing and analyzing standards. Known amounts of DUO were placed on the filter, air pulled at 50 CFM for 1/2 hour, the filter removed, uranium dissolved in the mixed acid leach, and a portion sent for analysis. Total DUO powder airborne was calculated using the uranium content of 87.6%.

Powder Characterization

Particle size, morphology, density and bin-flow tests were performed on the powders.

Particle Size

The size distributions of the source powders were determined using sedimentation methods that record the cumulative mass settled in a column of liquid. These are shown in Figure C.1. The mass median diameter (MMD) of the TiO₂ was $.7 \mu$ m; DUO, 1.0μ m; σ g was 2 for both powders. Ninety-five percent of the TiO₂ powder was between 0.425 and 6.8 μ m, MMD; the DUO between 0.25 and 4.0 μ m. Using theoretic density of 4.26 g/cc for TiO₂, 10.76 g/cc for DUO, aerodynamic diameters of 3.5 and 3.3 μ m, respectively, were calculated. The DUO was thus a somewhat finer powder as tabulated in Table C.2. A size distribution considered representative of dry PuO₂ powders (Schwendiman 1977) is included for comparison.



Density

A bulk, or pour, density for both powders was determined by pouring powder into a tared graduated cyclinder and reweighing. The density of the powder was determined as 0.63 g/cc, for TiO_2 and 1.5 g/cc for DUO. This determination of bulk density was considered representative of the source powder as used in these experiments.

Morphology

The morphology of the powders is shown in the scanning electron microscopic photographs, Figures C.2 and C.3. The samples are briefly dispersed ultiasonically, yet many apparent agglomerates are visible in the upper photos for both powders. Agglomeration appears less in the TiO₂ powder.

Bin-Flow Test

In an effort to gain the innate "flowability" of the powders, a rheological test (Zenz and Othmer 1960) was performed, demonstrating that the powders were cohesive and did not tend to flow. A rheological test evaluates the interparticle friction (angle of internal friction, α) that is important in gravity flow and could play a role in powder leaks and spills. The DUO and TiO₂ powders were both tested and compared to tests on sand with a mass median diameter of 64 µm.

TABLE C.2. Source Powder Particle Size Dis	stribution
--	------------

Size	Measured Mass	Cumulative Percent	
μm	Ti02	DUO	Pu02
20	98	97	100
10	97	95	81
8	96	94	56
6	94	93	39
4	88	92	20
2	60	85	4
1	16	50	1 .
0.8	11	40	

C.4





10,000X





5000X



20,000X



A bin-flow test measures the angle with the horizontal assumed by the moving core of solids in a vessel provided with a central opening in the bottom through which the contents can flow in free fall. The vessel is rectangular with a clear front wall, as illustrated in Figure C.4. The angle, a, can be measured at the line of demarcation between stationary and flowing solids.

A clear, plastic bin measuring 21.6 \times 2.54 \times 17.8 cm was fabricated. A 1.27- \times 2.54-cm hole in the bottom of the bin was covered with a sliding plastic cover that could be opened to allow powder flow. The bin was filled with powder to a depth of approximately 12.7 cm. As the bottom cover was removed, a marginal amount (<5 g) of the experimental powders dropped out. Vigorous, constant agitation was required to make either DUO or TiO₂ powder flow.

In order to compare the experimental powders with powder that flows, sand with a mass median diamter of 64 μ m was tested in the same bin. As soon as the bottom hole was opened, fast flow was apparent; the line demarcation between the cone of flowing solids and the stationary solids was visible; and the angle of internal friction was measured as 80°



FIGURE C.4. Bin-Flow Test

APPENDIX D

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SAMPLING PROCEDURES

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APPENDIX D

SAMPLING PROCEDURES

Two general sets of experiments measured the aerosol produced by a free fall in static air. The first experiments used 47-mm open-face filters, the second used high-volume samplers. The 47-mm filters would create a minimal change in the quiescent air volume but require calculation of the total release. The high-volume samplers might create airflows but should collect all of the particles that become airborne. Sampling with an airflow was selected rather than gravity settling to enable comparison with future experiments with room air movements, where gravity settling was considered unsuitable.

Four sets of experiments investigated the following effects: source height and volume, time, location, and amount of sampling air volume. Highvolume samplers were determined to be the most satisfactory experimental design and were used in experiments with DUO and UNH.

Height and Source Volume Effects

The first eight experiments investigated the effect of fall height and source volume on the release. Two different volumes of 1 g/& uranine solution, 500 cc and 50 cc (0.5 g and 0.05 g uranine) were spilled from two heights, 1 m and about 3 m (at high as possible in the RART). Side-by-side open-face samplers (14 lpm) were located at 61, 152, and 244 cm above the RART floor. The sampler locations in the RART are illustrated in Figure D.1. Impactors with 28 lpm throughput at the same levels collected samples to evaluate particle sizes. Aluminum foil on the RART walls and floor defined mass distribution at these locations. Sampling was limited to insure minimal disturbance of the static air.

Time Effects

The second series of experiments investigated changes in aeroso? concentration with time.



FIGURE D.1. Height and Volume Effects Sampling

The samplers were located at the same level as the first experiments--61, 152, and 244 cm above the RART floor. Five open-face filters were located side-by-side at each level. Four of the samplers had collection at times ranging from 5 minutes to 3 hours. The remaining samplers were exposed to the RART air for the entire experiment with no collection ("zero" airflow). This sample was background for all the samplers located at each level.

Location Effects

These experiments tested the assumption that samples collected in one section were representative of the aerosol distribution in the RART. This was important since this assumption was made when the results of the experiments were calculated. One experiment evaluated the difference in samplers located side-by-side, 5 open-face filters sampled at 61, 152, and 244 cm above the RART floor. Other experiments defined the distribution in the RART by sampling equal volumes of the RART. This was achieved by locating samplers in the middle of these volumes as shown in Figure D.2. They were at two levels, 75 and 225 cm, above the floor at four radial locations 18 and 75 cm from the wall. All sampling times were one hour.

Sampling Several RART Air Volumes

These experiments sampled all of the RART air, pulling eight "RART volumes" of air through samplers in a 30-minute run. This sampling collected almost air aerosol generated by a spill. It was used in the statistical matrix and was described in detail in the report section. A schematic, Figure 2 (p. 9), shows the configuration. The samplers were equipped with 8- x 10-in. glass fiber filters with 99.9% retention of 0.3 μ m particles and sampled at 1.4 m³/min, pulling 168 m³ of particle-laden air through the filters. A high-volume cascade impactor (0.56 m³/min) sampled for size distribution.



FIGURE D.2. Location Effects Sampling

APPENDIX E

PROBABLE SAMPLING ERROR CAUSED BY PARTICLE SETTLING

APPENDIX E

PROBABLE SAMPLING ERROR CAUSED BY PARTICLE SETTLING

Estimating the sampling error due to settling of the experimental measurements in this study can only be done for an ideal case. This case is the "well mixed" volume. The RART volume after the spill is not well mixed although the high volume samplers help to mix the air. From this "well mixed" compartment the samplers compete with gravita ional settling for the airborne particles. Once settled, the particle is assumed lost from the air and cannot be part of the sample. The particles of concern settle according to the Stokes' Law expression for terminal velocity, V_{t} , where

$$v_t = D^2 (\rho_P - \rho_a)g/18\mu$$

D = particle diameter (spherical equivalent diameter)

g = gravitational acceleration

 $\mu = air viscosity$

 $p_p = particle density$

 $\rho_a = air density$

Thus, the ratio of particles lost due to settling of the particles sampled is simply

V_t A_h/F

where

 A_{h} = horizontal surface area of the RART

F = total sampler volumetric flow rate.

This ratio is plotted versus D $\sqrt{\rho_p}$, the aerod namic equivalent diameter.

Thus if one desires to account for possible settling losses in defining the initial airborne source term using the RART data, Figure F.1 can be used for any part of the particle size distribution. Since the correction is highest for the largest particles which are least likely to challenge the pathway to the environment (because of settling), we have ignored correcting the data of this report because corrections are not extremely significant.





APPENDIX F

SPILL VELOCITY

APPENDIX F

SPILL VELOCITY

High speed photographs of the 3-m, 1000-g spills with TiO₂ and DUO were taken, but reproductions were not of the quality required for a report. There-fore, they are not included here. However, they were used to estimate the time required to complete the spill.

The TiO_2 powder spill required 1.36 sec; DUO, 1.2 sec. This corresponds to an average fall velocity of 220 and 250 cm/sec, respectively, over 3 m.

Individual particle velocities of the average $20.5 \ \mu m \ TiO_2$ particle would be about 11 cm/sec; $20.8 \ \mu m$ DUO 12 cm/sec. The spill front thus fell faster than individual detached particles.
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NRC FORM 335 U.S. NUCLEAR REGULATORY COMMISSION BIBLIOGRAPHIC DATA SHEET		1. REPORT NUMBER (Assigned by DDC) NUREG/CR-2139 PNL-3786 2. (Leave blank)			
4. TITLE AND SUBTITLE (Add Volume No. (I appropriate)					
Aerosols Generated by Free Fall Spiils of Powders and			3. RECIPIENT'S ACCESSION NO.		
Solutions in Static Air					
7 AUTHORIS)			5. DATE REPORT C	OMPLETED	
S.L. Sutter, J.W. Johnston, J. Mishima		December	December 1981		
9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)			DATE REPORT IS	DATE REPORT ISSUED	
Pacific Northwest Laboratory			MONTH	MONTH YEAR	
Richland, WA 99352		December	1981		
		6 (Leave blank)			
			8. (Leave blank)		
12 SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)		10. PROJECT/TASK/WORK UNIT NO.			
Division of Risk Analysis Office of Nuclear kegulatory Research			11. CONTRACT NO		
U.S. Nuclear Regulatory Commission Washington, DC 20555		FIN B2287			
13. TYPE OF REPORT		PERIOD COVER	RED (Inclusive dates)		
5. SUPPLEMENTARY NOTES		14. (Leave Diank)			
16. ABSTRACT (200 words or less)				244 C	
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13. KEY WORDS AND DOCUMENT ANALY	5)S.	17a DESCRIPTOR	85		
17b. IDENTIFIERS/OPEN-ENDED TERMS		19. SECURIT	Y CLASS (This report)	21. NO OF PAGES	
		Unc1	assified		
Unlimited 20 SECONT Uncla		assified	22 PRICE S		

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