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Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air

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ABSTRACT

Safety assessments and environmental impact statements for nuclear-fuel-cycle facilities require an estimate of potential airborne releases caused by accidents. Aerosols generated by accidents are being investigated by Pacific Northwest Laboratory to develop the source terms for these releases. An upper boundary accidental release event would be a pressurized release of powder or liquid in static air. Experiments were run using various source sizes and pressures and measuring the mass airborne and the particle size distribution of aerosols produced by these pressurized releases. Two powder and two liquid sources were used: TiO_2 and depleted uranium dioxide (DUO); and aqueous uranine (sodium fluorescein) and uranyl nitrate solutions. Results of the experiments showed that pressurization level and source size were significant variables for the airborne powder releases. For this experimental configuration, the liquid releases were a function of pressure, but volume did not appear to be a significant variable.

During the experiments 100 g and 350 g of DUO (1 μm dia) and TiO_2 (1.7 μm dia) powders and 100 cm^3 and 350 cm^3 of uranine and uranyl nitrate solutions were released at pressures ranging from 50 to 500 psig. The average of the largest fractions of powder airborne was about 24%. The maximum amount of liquid source airborne was significantly less, about 0.15%.

The median aerodynamic equivalent diameters (AED) for collected airborne powders ranged from 5 to 19 μm; liquids ranged from 2 to 29 μm. All of the releases produced a significant fraction of respirable particles of 10 μm and less.

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SUMMARY AND CONCLUSIONS

Pacific Northwest Laboratory conducted experiments to measure aerosols generated by pressurized releases of powders and liquids. Data from these experiments will be used in developing models to predict accidental airborne releases. The mass airborne and particle size distribution of the aerosols generated by pressurized releases were measured as a function of source size and chamber pressurization level. Powder and liquid sources were placed in a container, pressurized to selected levels, then released to the ambient air by rapid depressurization through ruptured discs.

Two TiO_2 and DUO masses (100 g and 350 g), and two uranine and UNH solution volumes (100 cm^3 and 350 cm^3), were released from a pressurized chamber at 50, 250, or 500 psig. Releases were made in the Radioactive Aerosol Release Tank (RART) which has a volume of 20 m^3--about the size of a small room. The airborne material was collected by high volume samplers. Particle size distributions were calculated from samples collected using a high volume cascade impactor.

Pressurization level and source size were significant variables for the airborne powder releases, with pressure having a more pronounced effect. Releases from the two types of powders were comparable. The average weight percent airborne for these powders ranged from about 2% to 24%. Although there was less airborne mass from the 100-g powder source, the percentage airborne was greater than from the 350-g powder source.

Liquid mass releases were a function of pressure, but for this experimental configuration (rupture discs above the liquid) the source volume did not appear significant. The average weight percent of liquid source that became airborne ranged from 0.0004% to 0.15%, and as with powder, represented a larger fraction of the smaller sources.

Other findings were:

- All releases produced a significant fraction of particles 10 μm and less in median aerodynamic equivalent diameter (AED). These are considered respirable.
- Powder aerosols ranged from an average of 5 μm to 19 μm AED.
- Liquid aerosols ranged from an average of 2 μm to 29 μm AED.
- Uranine solutions produced the finest, most homogeneous aerosols.

Results of these experiments are now being used to develop models to estimate the airborne particulate material generated by the energy in these events.

Some suggestions for future pressurized release investigations are included in this report.

INTRODUCTION

Aerosol generation characteristics of accidental particle releases 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 Nuclear Regulatory Research. Safety analysis reports and environmental impact statements must evaluate the consequence of postulated accidents in or involving the facility in question. During an accidental release from a nuclear fuel cycle facility the dominant pathway to man is usually through airborne particles, so it is necessary to determine an aerosol source term (i.e., the quantity of material initially airborne from an accident).

Assessments of the potential consequences of accidents are based on scenarios that define the sequence of largely inadvertent conditions that result in airborne releases. Typical events range from spilling the materials in free fall through air, to releases caused by fire and explosion. It is difficult to make reasonable assessments of accident scenarios, 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 to assess the aerosol generation and behavior under these conditions, and to perform experimental studies to provide new data.

To give us an indication of the upper and lower limits of accidental airborne releases, we have suggested upper and lower boundary events. Free-fall spills in static air are considered a lower bound release event (Sutter et al. 1981) and releases of pressurized powders and liquids from ruptured containers are suggested as an upper bound event. This report describes a series of experiments performed to investigate aerosols generated by these releases. Such events are of concern and have occurred in the nuclear industry. An example of a pressurized release scenario happened in a plutonium oxide storage facility (Committee Report 1979). Presence of decomposition products and water in a can of plutonium oxide powder led to pressurization and subsequent rupture of the can and resulted in a plutonium release.

Data to calculate the release of pressurized powders and liquids from containers were not found in a literature review (Sutter 1982). The only information available to estimate the magnitude of this type of release has been based on estimations of airborne powder concentrations. Swain and Haberman (1961) reviewed data from nonnuclear sources and calculated that 33 mg/m^3 was the maximum airborne powder concentration a few minutes after an accident. Castleman, Horn and Lindauer (1969) generated fine particles using an exploding wire technique and found concentrations as high as 71 mg/m^3 plutonium a few minutes after generation. Mishima (1975) anticipated an upper concentration of 100 mg/m^3 for quasi-stable, accident-generated, airborne concentrations of dry powders.

Because the literature review did not reveal adequate information to characterize pressurized releases, an experimental program was implemented. Experimental pressurized releases of powders and liquids were conducted in static air in an enclosure, and the airborne mass and particle size distribution determined. We measured pressurized releases of two powders, titanium dioxide (TiO_2) and depleted uranium dioxide (DUO) and two liquids, sodium fluorescein solution (uranine) and uranyl nitrate hexahydrate (UNH), at three pressures 50, 250, and 500 psig. Abbreviations for the compounds are used throughout this report to avoid confusing uranine and uranium. Data from these experiments will be used in ongoing work to develop release models based on physical parameters. Pressurized releases resulting from closed containers that rupture when exposed to excessive heat (i.e., fires) are not considered in this study. Liquid spray data can be used to evaluate pressurized releases of liquids with the failure point below the liquid level.

General experimental procedures and the results are discussed in the main portion of the work. This study is part of a continuing experimental program to measure accident-generated particulate materials. Therefore, the same source materials, sampling procedures, and sample analysis techniques used in an earlier work were employed here. These were described in detail by Sutter (1981) in a report documenting free fall spill experiments. Airborne mass measurements are in Appendix A. Appendix B lists the median aerodynamic equivalent diameters.

EXPERIMENTAL PROCEDURES

Experiments were performed to determine the amount of airborne material and the aerodynamic particle size of the aerosols generated by releases of pressurized powders and liquids in static air. We were interested in determining the airborne mass generated during the release event rather than the time-dependent behavior of the aerosol. Through our experiments 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 experimental parameters was not within the scope of this project. Experiments were limited by the availability of equipment and planning decisions. We are interested in indoor releases, so most of the experiments were performed in a room-size enclosure.

We used a matrix of experiments using TiO_2 and DUO powders and uranine and UNH liquids to investigate the effect of source size, density, and chamber pressure on the airborne release. We ran experiments at three pressures (50, 250, and 500 psig), with 100 g and 350 g of powder or 100 cm^3 and 350 cm^3 of liquid. Radioactive DUO and UNH were used because they are found in nuclear fuel cycle facilities and are considered suitable surrogates for PuO_2 powder and plutonium solutions. This was important because the information from these experiments is being developed for use in evaluating hazards in nuclear fuel cycle facilities.

EXPERIMENTAL CHAMBER

A chamber called the PARE (Pressurized Airborne Release Equipment) was designed and fabricated for use in these experiments. The PARE, shown schematically in Figure 1, is approximately 800 cm^3 in volume and can be loaded with a maximum of 524 cm^3 of powder. TiO_2 is a light, loosely compacted powder and 350 g filled the chamber. This established the maximum powder mass of both TiO_2 and DUO used in the experiments; 100 g was the second source quantity used.

After pressurization with air to a preselected experimental level, material contained in the PARE can be released explosively by the rupturing of discs. A rupture disc is a thin, circular, metallic membrane clamped between metal flanges. The complete assembly is called a safety head, because it is usually used as a relief valve to protect a system from excessive pressure. The discs are constructed to rupture at a specified pressure. The PARE uses a double disc safety head to give precise control of the chamber pressure at the time of rupture. With this disc arrangement the intermediate chamber pressure can be varied to keep the pressure difference across the disc within safe limits. To cause the powder release, a solenoid valve is opened letting the intermediate chamber depressurize. The pressure difference across the lower disc quickly becomes too great and the disc ruptures, causing the upper disc to rupture microseconds later.

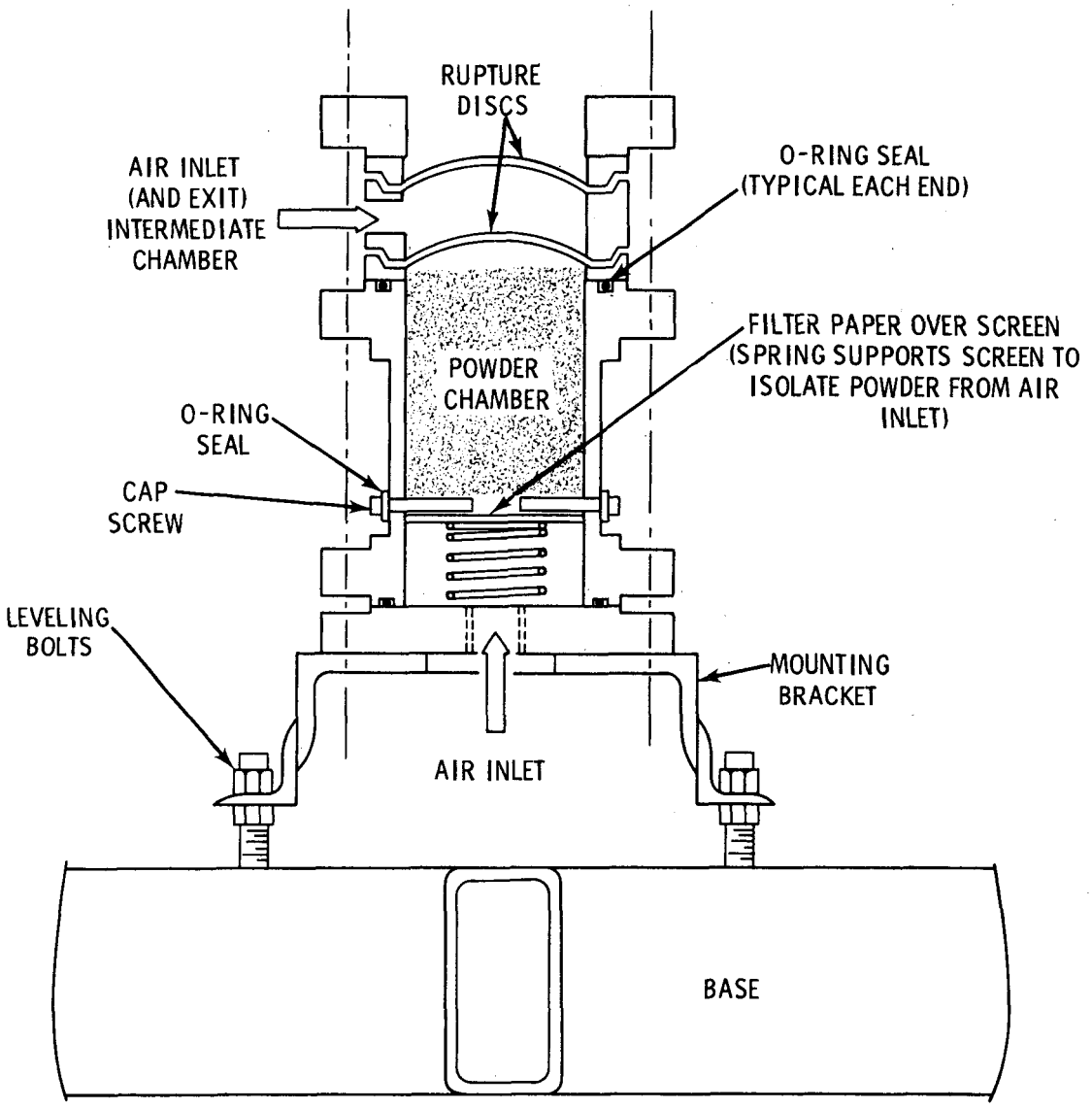

FIGURE 1. PARE, Pressurized Airborne Release Equipment

PROCEDURE

The general sampling procedure for conducting the experiments, shown in Figure 2, was to pressurize both chambers and then depressurize the intermediate chamber thus ejecting the powder. Samplers were immediately turned on and particle laden air pulled onto filters, as indicated by the solid arrows in Figure 2. Clean air exits from the rear as indicated by the light arrows. Figure 3 is a photo of one of the experiments showing a typical powder release.

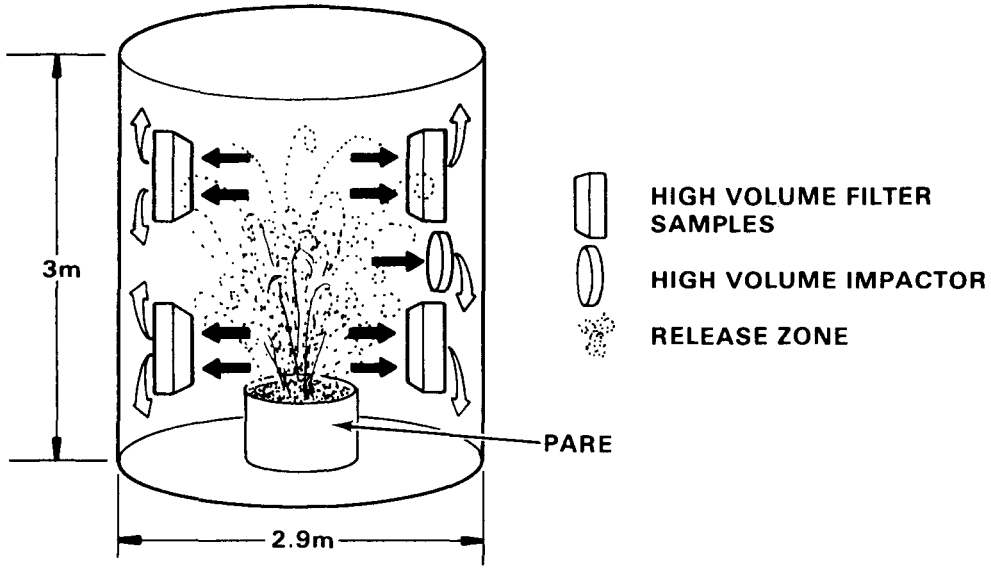

FIGURE 2. Sampling for a Pressurized Release with High Volume Samplers

The experiments were conducted in a cylindrical enclosure called the Radioactive Aerosol Release Tank (RART). It is a stainless steel vessel approximately 3 m high and 2.9 m in diameter with a 20-m^3 volume, about the size of a small room.

Figure 2 shows four high volume samplers at the 1-m and 2-m level and one high volume cascade impactor at the 1.5-m level, equipped with appropriate glass fiber filters, as they were used to sample aerosol. Eight RART volumes of air were pulled through the samplers in a 30-minute run. After 30 minutes, 99% of the airborne material should be collected since the aerosol concentration decayed exponentially. Background samples after early runs showed that in experiments generating large amounts of airborne powder, 1.5% of the aerosol mass could remain airborne after sampling. Therefore, two additional high volume samplers were included and sampling time increased to one hour. Analysis of overnight deposition samplers placed in the RART after the high volume sampling established that less than 1% of the collection remained airborne. [Some of the airborne material settles during the sampling. As in the case of the free-fall spills data (Sutter 1981), corrections can be made for this settling loss, but they are not extremely significant.]

Experimental pressures were monitored with a transducer indicator which was calibrated before use to an accuracy of ±0.5%. The chamber pressure level was displayed as a digital readout.

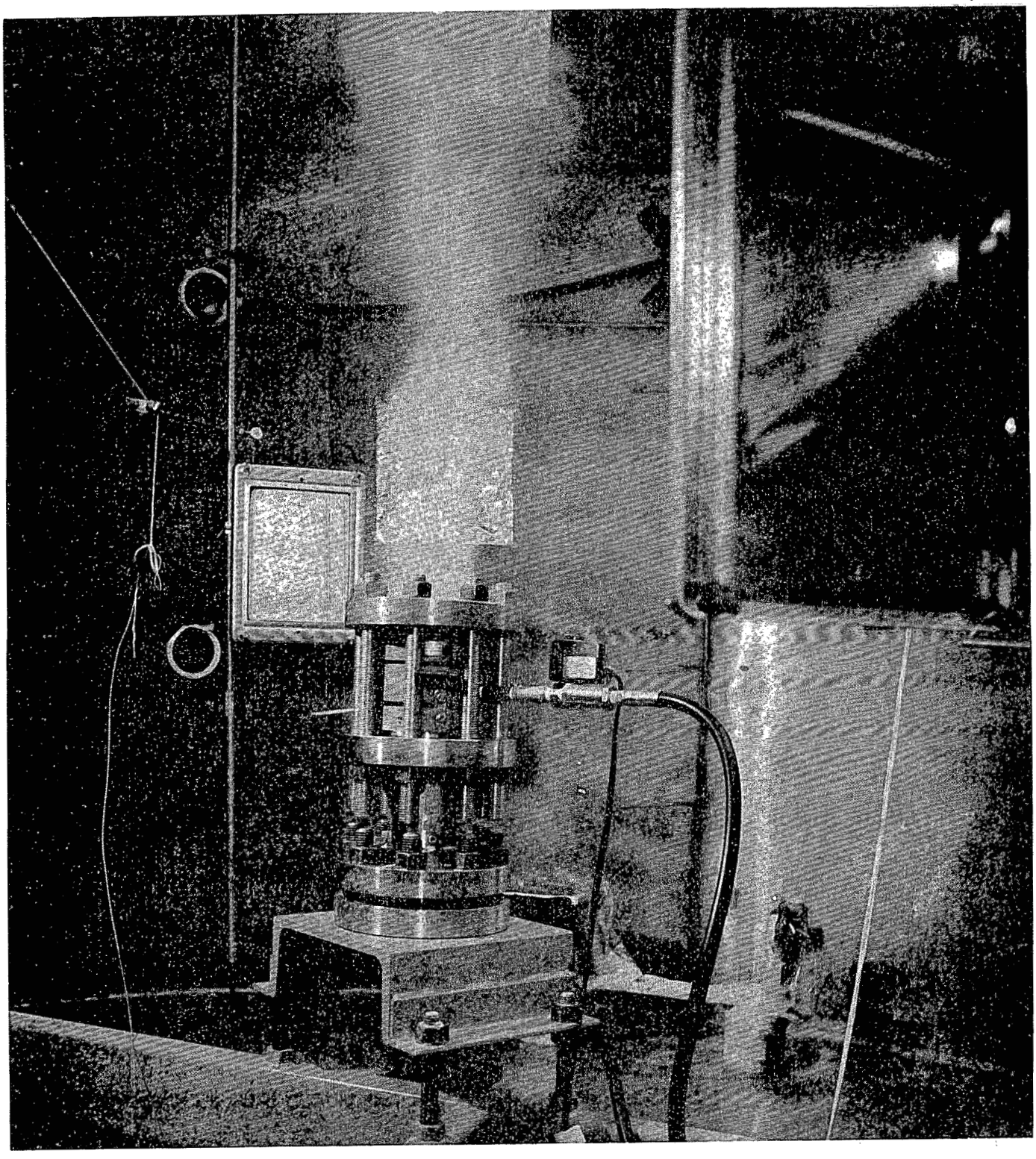

FIGURE 3. PARE Experiment

Experimental Design

The experimental matrix was designed for analysis of variance (AOV) techniques. AOV divides the total variation of the dependent variables (mass airborne) 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 the uncertainty of the results. Replicate experimental runs are included at all levels.

Wall Deposition

In some of the initial experiments, foil squares were placed on portions of the RART walls to collect aerosol deposition samples. Our work focuses on measuring airborne material so sampling of wall deposition was limited.

Sample Analysis

Uranine aerosols collected on the filters were dissolved in water and analyzed by fluorometry. Dilutions of a stock solution representing different concentrations were made and the results read on a fluorometer. The lower detection limit is about 1×10^{-9} g/cm^3. The results were plotted and the unknown sample values read from this plot. We formulated equations for the plot at each of the four fluorometer ranges and used these for calculating results. R^2 (correlation between observed and predicted values) values approached one, indicating a good fit for the standard curve at all attenuations.

We dissolved filter samples containing collected uranium aerosols in acid and analyzed the samples using laser fluorometry. This method employs a pulsed nitrogen laser to excite uranium in a solution containing a pyrophosphate reagent. The fluorescent signals are amplified and integrated, and the results are displayed on a meter. We analyzed standards that were prepared using our DUO results and established that the error was less than 10%.

OTHER POWDER RELEASES

Initial experiments revealed an enclosure effect from the high pressure jetting powder out and impacting it on the RART ceiling. This led us to the question of how high an unrestricted release would go. Two experiments (350 g TiO_2 at 50 and 1000 psig) were conducted outdoors and the release height was measured using triangulation methods. The release volume was estimated from high speed photographs.

Unpressurized powder above a pressurized chamber can be involved in an accident. The PARE was modified as shown in Figure 4 so powder could be placed above the chamber. Only a limited number of these PRAC (Powder Release Above Chamber) experiments were performed since the PARE type event is considered more important in our accident scenarios. TiO_2 was released once at each pressure and source size level.

LIQUID RELEASES

The PARE was modified as shown in Figure 5 to allow pressurized liquid releases from the same configuration used in powder releases. Experiments with two source quantities (100 cm^3 and 350 cm^3) at three pressures (50, 250, and 500 psig) were completed using uranine and UNH.

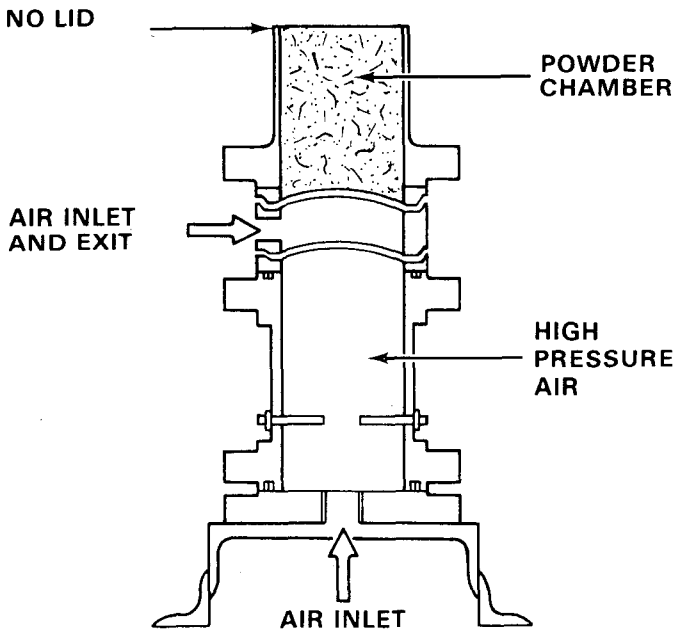

FIGURE 4. PARE Modification for Powder Release Above Chamber (PRAC) Experiments

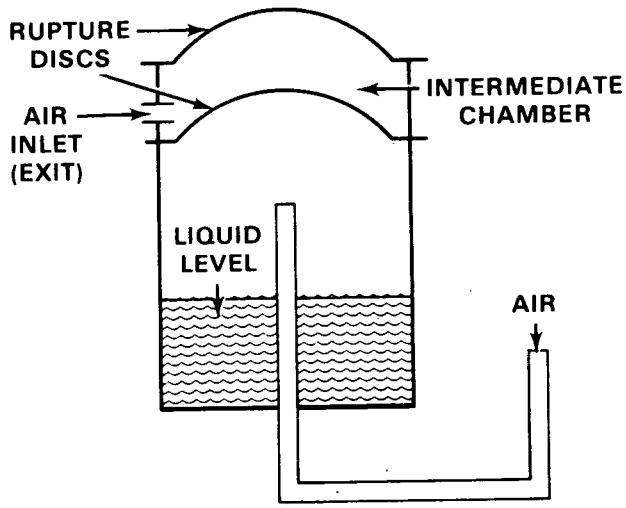

FIGURE 5. PARE Modification for Pressurized Liquid Release Experiments

RESULTS AND DISCUSSION

Included in the following discussion are results of the high volume sampling of the powder and liquid release experiments. These results will be used to develop models which will estimate airborne releases of accident-generated aerosols. These models, which are to be based on physical parameters, will be described in a later report.

A complete listing of the measured release values for the mass airborne is in Appendix A and values for particle size are in Appendix B.

POWDER RELEASES -- PARE EXPERIMENTS

PARE releases in the RART were the primary thrust of the pressurized powder experiments. In the following paragraphs we discuss measurements of the total mass airborne, weight percent of source powder airborne, and particle size distribution, resulting from pressurized powder releases.

Mass Airborne

The average powder mass airborne from replicate runs (for individual run results see appendix) of TiO_2 and DUO release experiments are listed in Table 1 and plotted in Figure 6. Two exploratory experiments using TiO_2 at 1000 psig are included.

TABLE 1. Average Mass Airborne from Pressurized Powder Releases

Pressure, psig	350 g Source		100 g Source	
	TiO_2, g	DUO, g	TiO_2, g	DUO, g
1000	15.9		12.4	
500	27.8	30.2	24.1	19.8
250	18.3	20.5	18.2	13.0
50	3.3	6.9	4.4	3.6

The results in Table 1 for TiO_2 make it appear that the release quantity was independent of source quantity. A review of the experimental data, however, suggest that this is an effect of time. Two significantly different releases were averaged. Five exploratory experiments were performed in the summer to check out the PARE apparatus, and then further experiments were postponed because of other work requirements. As was found in the free fall spill experiments (Sutter 1981), there was a time effect. Later runs had higher levels of TiO_2 than did early runs. Replicate runs yielding larger releases were made at lower temperatures, 14°C to 19°C, and early runs at as much as 23°C.

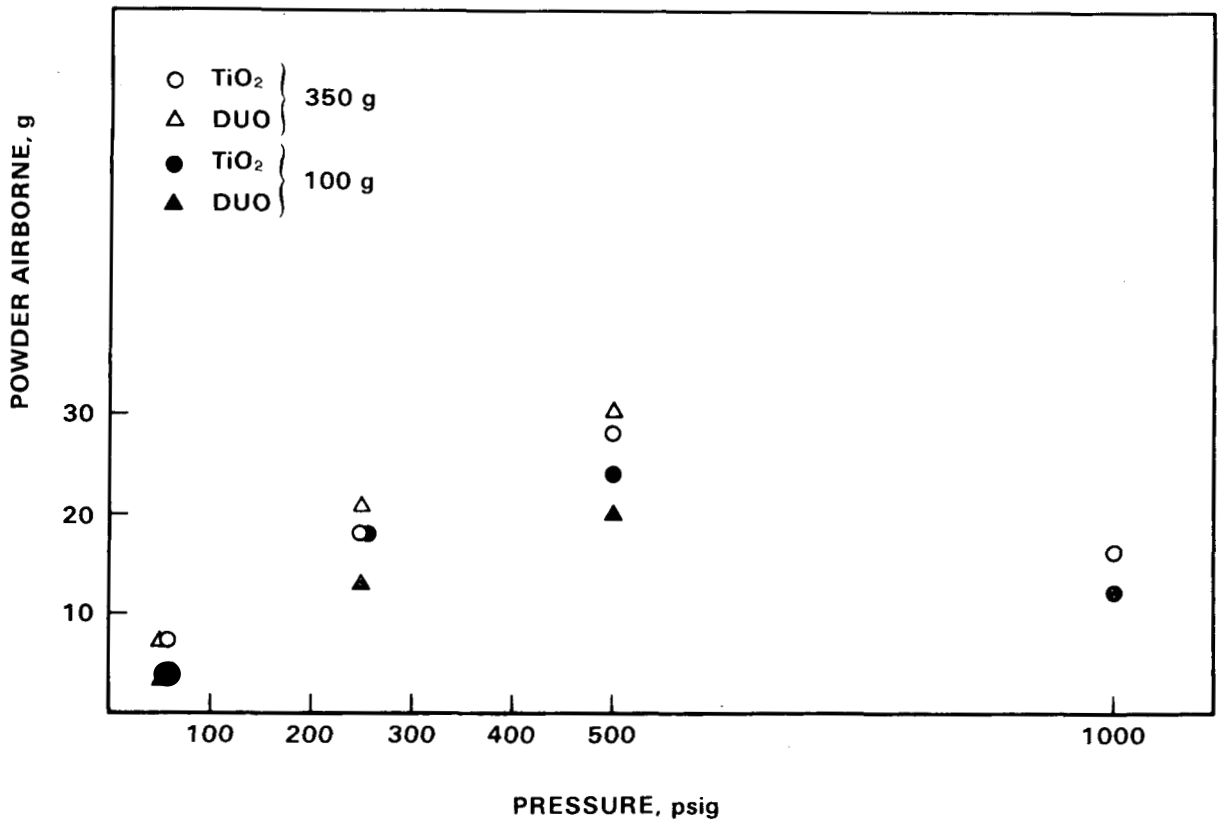

FIGURE 6. Mass Airborne from the Pressurized Release of Two Powders

The average mass airborne from the PARE experiments increased as a function of pressure for both TiO_2 and DUO powders. This increase continued up to 500 psig as shown in Figure 6. The 1000 psig exploratory runs using both source quantities of TiO_2 had a lower measured mass airborne than the 500 psig runs. This is attributed to an enclosure effect because the powder impacted on the center of the ceiling where a portion was retained. After impacting on the ceiling, the remainder spread to the sides of the RART where there was further impaction, and consequently, less of the release was in the airborne fraction. Because the focus of our work is to identify the maximum airborne release, runs at 1000 psig were not continued. Ceiling impaction was a factor at all pressures, however.

A material balance would be helpful to explain the above discrepancies, but attempts made to collect the ejected powder were not satisfactory. The ejected material was so widely dispersed that the technician was unable to recover it completely. For example, during the first run at 250 psig, 10 grams became airborne, but only 288 g were recovered from the RART. This leaves 50 g of the 350 g source unaccounted for.

Chamber pressure and source quantity were significant variables for aerosol generation for both powders, with the pressure having a more pronounced effect. Release from the different types of source powders were comparable for the same experimental parmeters. Each high volume sampler had about the same mass collection, indicating that the aerosol achieved a fairly uniform distribution in the RART.

There was no pressure rise evident in the RART. This was anticipated since we calculated the ΔP using the expression:

$$\Delta P = \frac{V_P}{V_R} \left(\frac{Ta}{273^0 K}\right) P_g$$

where

ΔP = pressure rise, psi
V_P = PARE volume
V_R = RART volume
T_a = ambient temperature, °K
P_g = pressure of compressed gas, psi.

Total volume of the PARE is about 862 cm^3 (812 cm^3 for the chamber and an additional 50 cm^3 for the dome shaped rupture disc). The RART volume is about 2.0×10^7 cm^3, so even for the empty chamber at 1000 psig the ΔP is 0.05 psi.

Weight Percent Airborne

The weight percent of source that becomes airborne is a direct method used to estimate releases, so our data (average of replicate runs) for powder are presented as weight percents in Table 2 and plotted in Figure 7.

The weight percent of the source that became airborne in the enclosure was larger for the smaller sources. The weight percent of airborne source at different pressures was comparable for both powders. The maximum average value of 24% might be used to make a conservative estimate of pressurized powder releases within pressure limits used in this work. It is also possible that in an unconfined experiment, the weight percent airborne might increase as pressure is increased.

TABLE 2. Average Weight Percent Airborne from Pressurized Powder Releases

Pressure, psig	350 g Source		100 g Source	
	TiO_2	DUO	TiO_2	DUO
1000	5		12	
500	8	9	24	20
250	5	6	18	13
50	2	2	4	4

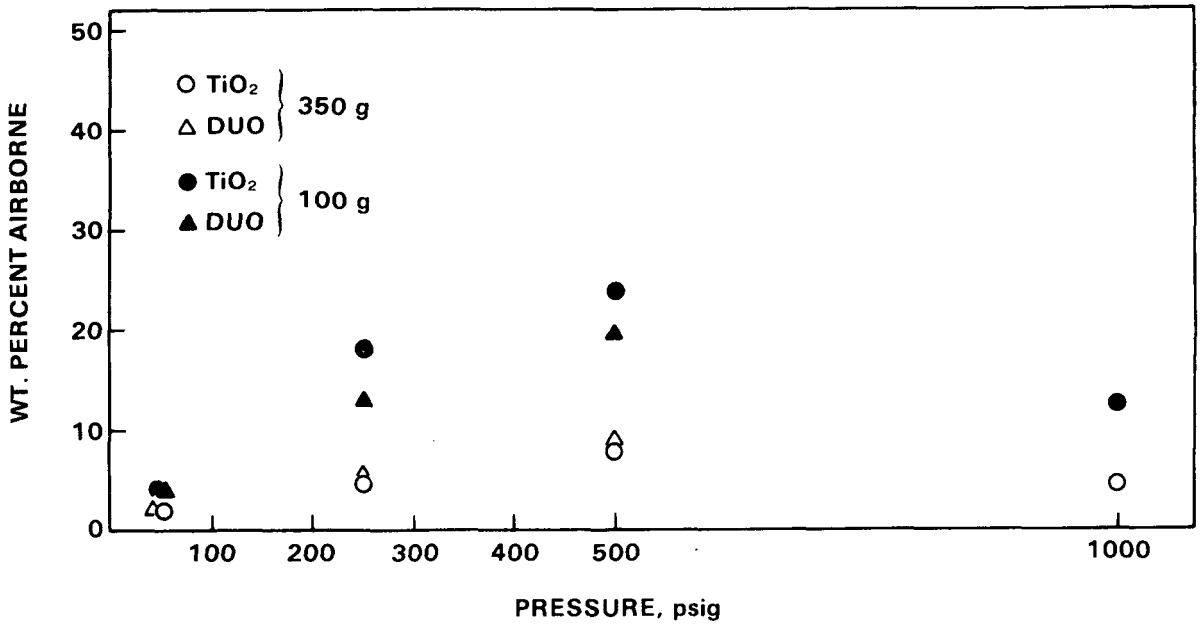

FIGURE 7. Average Weight Percent of Powder Airborne as a Function of Pressure

Particle Size Distribution

The median aerodynamic diameters for the particles for all experiments are listed in Appendix B. The average values from replicate, powder-release experiments are shown in Table 3. These powders are similar in terms of diameter.

TABLE 3. Average Median AED of Particles Generated by Pressurized Powder Releases, μm

Chamber Pressure, psig	350 g		100 g	
	TiO_2	DUO	TiO_2	DUO
500	14	16	5	14
250	14	15	10	11
50	18	19	8	19

Some of the calculated diameters are beyond the range of the impactor sampler and were estimated by extrapolation of the data. These particles are probably agglomerates since they are larger than the source powder particle size. The confined release in the enclosure would not give opportunity for

dispersion of agglomerates. It has been observed that the effectiveness of dispersion from this type of event is limited, and therefore, results in larger mean particle sizes (Seefeldt 1977). Seefeldt concluded that an accidental explosion will have the greatest consequence in the absence of confinement because the particle sizes would be smaller.

The geometric standard deviation (σg) of the particle size distribution is a measure of the aerosol homogeneity. These values ranged from 1.9 to 14 for the PARE powder experiments indicating a disperse particle size distribution for the aerosols. DUO aerosols were the most uniform with σg values ranging from 1.9 to 4.8; TiO_2 ranged from 4.1 to 14.

All of the releases generated a significant fraction of particles with diameters 10 μm and less, particles that can be inhaled and retained in the lung. This portion ranged from 25% of the source mass for a DUO release at 50 psig to 72% for 100 g TiO_2 released at 500 psig. Respirable particles represented from 1 to 21% of the aerosol.

Wall Deposition

An enclosure effect can be anticipated from this event. During early runs we placed aluminum squares on the RART walls to collect aerosol deposition samples. Table 4 shows the total wall deposition calculated from replicate TiO_2 runs and the percent of source deposited. The largest fraction of the ejected material fell to the floor of the RART.

TABLE 4. Calculated Average TiO_2 Wall Deposition

	350 g Source		100 g Source	
Pressure	g	%	g	%
500	2.25	0.6	0.98	0.98
250	2.31	0.7	1.14	1.14
50	0.94	0.3	0.16	0.16

About 1% of the source was deposited on the walls in all experiments. The presence of walls can cause two effects: 1) the trajectory of the initial impact is shortened and this may eliminate added shear stresses on the particles that could cause more airborne fines, and 2) the impact itself probably creates some aerosol that would not be generated in the unrestricted path. These two effects counterbalance one another and their net contribution is likely to be small.

Powder Ejected

At 500 and 250 psig, all the powder was ejected from the PARE. At 50 psig, some powder sometimes remained in the PARE. Based on the amount recovered from the PARE, 77 to 97% of the source powder could be ejected from the PARE during a release at 50 psig.

POWDER RELEASES -- PRAC EXPERIMENTS

The masses airborne from the PRAC experiments are listed in Appendix A, and median particle sizes in Appendix B. Figure 8 is a plot of the mass releases. It appears from the plot that a maximum release would be about 38 g for this configuration.

The venting of pressurized gases through unpressurized powder resulted in an airborne mass ranging from 1.2 to 2.3 times greater than corresponding releases from the PARE, as shown in Table 5. Median particle sizes ranged from 7 to 15 μm, similar to that from PARE releases.

Because we consider the PARE type release (pressured powder and container) more important in accident scenarios, this work was limited to a few exploratory experiments.

POWDER RELEASES -- OUTDOOR TESTS

Two outdoor tests measured the plume height and volume produced by an unrestricted PARE release of 350 g TiO_2 powder at 50 and 1000 psig. Data generated from these tests are shown in Table 6. Plume heights measured using

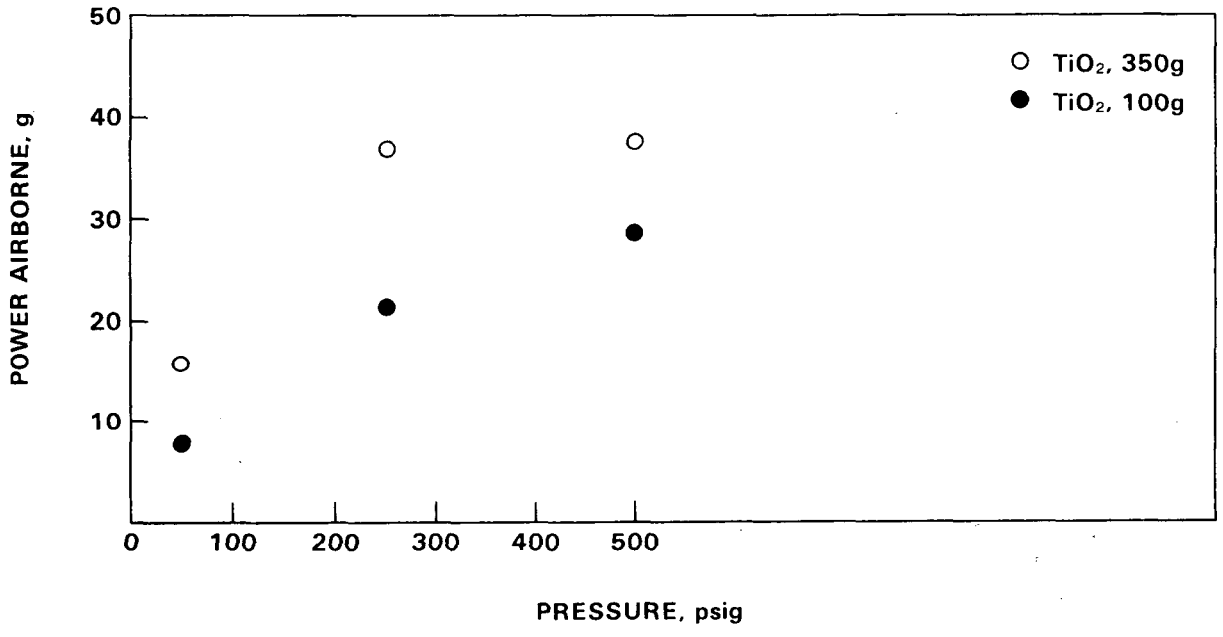

FIGURE 8. Mass Airborne from PRAC Experiments

TABLE 5. Comparison of PARE and PRAC Releases

Pressure	350 g TiO_2 PARE, g	350 g TiO_2 PRAC, g	x Greater	100 g TiO_2 PARE, g	100 g TiO_2 PRAC, g	x Greater
500	27.8	38.2	1.4	24.1	28.7	1.2
250	18.3	36.7	2.0	18.2	21.3	1.2
50	7.4	16.9	2.3	4.4	8.2	1.9

TABLE 6. Results of Unrestricted PARE Pressurized Powder Releases

Parameters	Pressurized Powder Releases 50 psig	Pressurized Powder Releases 1000 psig
Plume height	4.5 m	10.7 m
Plume volume	2.2 m^3	10 m^3
Expelled from PARE	~200 g	350 g
Plume initial calculated concentration	91 g/m^3	35 g/m^3
Amount deposited in ~2.25 m radius of PARE	30 g	2 g
Percent Expelled and deposited near PARE	15	<1

triangulation techniques were 4.5 and 10.7 m for 50 and 1000 psig, respectively. Volumes listed in Table 6 were estimated from high speed photographs. Deposition samplers placed on the ground near the PARE were analyzed to determine the amount deposited in the immediate vicinity of the release.

There was no measurable windspeed at the 50-m elevation during these outdoor tests, however, the release cloud drifted away. As anticipated, an unconfined event would have more severe consequences, going higher and further than a release in the enclosure. Even more important, if the amount collected in a 2.25-m radius represents that not airborne, then the unconfined release can result in 100% airborne at a high pressure.

LIQUID RELEASES

The modified PARE equipment shown in Figure 5 was used for pressurized liquid releases. Experiments measured releases from two source volumes at

three pressures. In the following paragraphs we discuss the measurements of mass airborne, weight percent airborne, liquid ejected, and particle size distribution resulting from pressurized releases of liquids.

Mass Airborne

The average from replicate runs of the solution mass airborne is listed in Table 7 and plotted in Figure 9. These values were calculated using measured solution densities of 0.99 g/cm^3 for uranine, 1.54 g/cm^3 for UNH.

The mass airborne increased as a function of pressure for both uranine solutions and UNH. The source volume did not appear significant in determining the magnitude of the release. Releases were also lower than we had anticipated. High speed photography revealed that the release seemed to be a vaporization of the liquid caused by the rapid depressurization. The only noticeable release was a condensed vapor cloud. However, since uranine and UNH did become airborne, some surface disturbance had to occur to entrain the solutes. The differences in the mass airborne from the different volumes may be a result of experimental variability. These measured releases were in the same range for both solutions.

These results were low enough to suggest comparison with free fall spill releases (Sutter 1981). It can be suggested that airborne releases from the two accident types we have studied might be related. In other words the lowest pressurized release measurements could be similar to the higher release values from spills. This hypothesis was examined for both powders and liquids.

While verification of this hypothesis was not evident in powder experiments, a review of the liquid releases indicates a close match in results. Releases resulting from 3 m spills of 125 cm^3 uranine and UNH were measured and found to be essentially the same as the release of 100 cm^3 of pressurized liquid. All of these releases ranged from 3×10^{-3} to 5×10^{-3} g. This close agreement might be coincidental. There are different stresses being placed on the liquid in the two types of experiments.

TABLE 7. Average Mass Airborne from Pressurized Liquid Releases

Pressure, psig	350 cm^3 Source		100 cm^3 Source	
	Uranine, g	UNH, g	Uranine, g	UNH, g
500	0.17	0.13	0.15	0.13
250	0.03	0.05	0.06	0.08
50	0.003	0.002	0.005	0.003

Weight Percent Airborne

The weight percent of the source liquids that become airborne is listed in Table 8 and plotted in Figure 10.

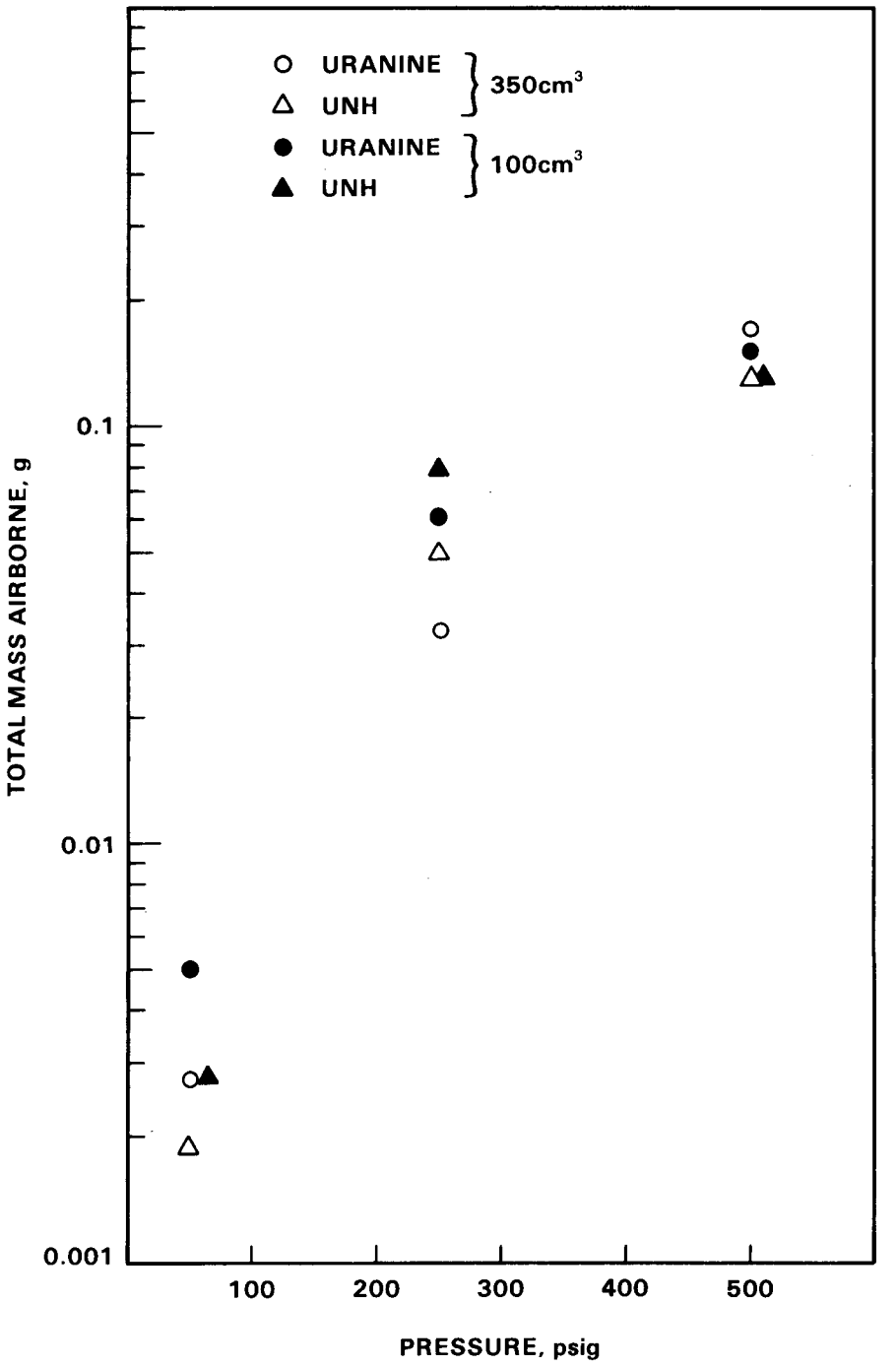

FIGURE 9. Mass Airborne from the Pressurized Release of Two Solutions

TABLE 8. Average Weight Percent Airborne from Pressurized Liquid Release

Pressure, psig	350 cm^3 Source		100 cm^3 Source	
	Uranine	UNH	Uranine	UNH
500	0.05	0.025	0.15	0.08
250	0.01	0.01	0.06	0.05
50	0.0008	0.0004	0.005	0.002

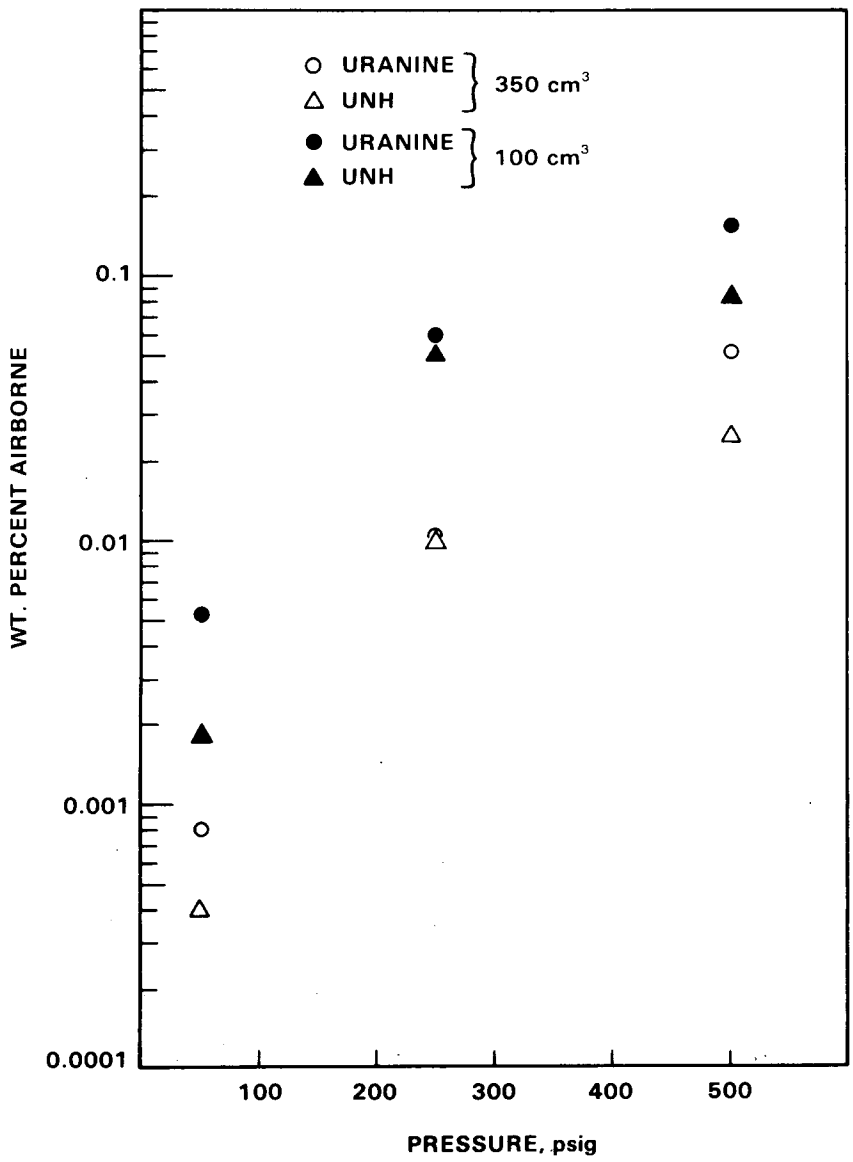

FIGURE 10. Average Weight Percent of Liquid Airborne as a Function of Pressure

The weight percent of airborne material was comparable between the two solutions and increased with increasing pressure. The solution densities were measured and used to calculate the source mass for calculations of weight percent. At a density of 1.54 g/cm^3, the source UNH is 539 and 154 g for 350 and 100 cm^3, respectively. Uranine solution density is 0.99 g/cm^3, which can be calculated to 347 and 99 g sources. The uranine source mass is about the same as that of the source powders, and can be used to compare the percent liquid and powder releases from the same source mass. This comparison is made in Table 9, using an average of the TiO_2 and DUO values.

TABLE 9. Comparison of Weight Percent of Source Mass Airborne from Pressurized Releases of Uranine Solution and Powder

	~350 g Source		~100 g Source	
psig	Powder	Uranine	Powder	Uranine
500	8.5	0.05	22.1	0.15
250	5.6	0.01	15.6	0.06
50	2.1	0.0008	4.2	0.005

The percent of powder released from the same source mass ranges from 150 to about 2600 times greater than comparable liquid releases. Dispersal mechanisms play a role in this difference. Small cavities in the powder bulk are filled with pressurized air. This acts to shatter the powder and energetically disperse it on release. The force acting on the liquid was rapid depressurization leading to vaporization and entrainment of the solution.

Liquid Ejected

In liquid releases a significant amount remains in the apparatus after the pressurized release. This amount was measured to determine the fraction of released material aerosolized. These values are listed in Table A.6 of the appendix. The average amount ejected is shown in Table 10, and the weight percent that became aerosolized is listed in Table 11.

TABLE 10. Average Amount of Liquid Ejected from PARE

	350 cm^3 Source				100 cm^3 Source			
Pressure,	Uranine,		UNH,		Uranine,		UNH,	
psig	cm^3	g	cm^3	g	cm^3	g	cm^3	g
500	14	14	10	15	5	5	8	12
250	8	8	7	11	6	6	6	9
50	7	7	6	9	3	3	5	8

TABLE 11. Average Weight Percent of Liquid Released that Became Aerosolized

Pressure, psig	350 cm^3 Source		100 cm^3 Source	
	Uranine wt%	UNH wt%	Uranine wt%	UNH wt%
500	1.2	0.9	3.0	1.0
250	0.4	0.5	1.0	0.09
50	0.04	0.02	0.2	0.03

Particle Size Distribution

Table 12 lists the average particle size distributions that were calculated from cascade impactor samples generated in replicate runs.

TABLE 12. Average Median AED of Particles Generated by Pressurized Liquid Releases, μm

Chamber Pressure, psig	350 cm^3		100 cm^3	
	Uranine	UNH	Uranine	UNH
500	4	16	4	14
250	4	29	3	13
50	2	4	2	7

Releases of uranine solutions generated the finest, most uniform particles. Median diameters were 4 μm and less, with σ_g values of 1.6 to 4. These particles were predominantly (70 to 90%), in the respirable size range of 10 μm and less. If there is a pressure or source quantity effect on the airborne particle size, the experimental variation between the 2 and 4 μm values precludes a conclusion.

The UNH releases produced larger particle sizes and more polydispersed aerosols. Average median diameters ranged from 4 to 29 μm and σ_g values from 2.5 to 7.5. Fewer respirable particles (10μm and less) were generated, forming 34 to 76% of the aerosol.

The above particle size distributions were measured by using the cascade impactor samples, and may not be representative of the size distribution of the airborne liquid droplets. Significant evaporation of the droplets captured in the impactor could mean that the measured size distribution does not correspond to that of the actual airborne aerosol. This problem will be handled in the model development and discussed in that report.

FUTURE WORK

The experiments discussed in this report are a continuation of our work investigating accident generated aerosols. Through our work, other potential research areas dealing with pressurized releases have been identified and are discussed in this section.

The behavior of liquid releases from pressurized containers, particularly, suggests the need for further experimental work. In our experimental configuration the pressurized air entered the bottom of the chamber, stabilized in the volume above the solution, and was released when the discs at the top of the chamber ruptured. The release seemed to be caused by the rapid depressurization vaporizing and entraining some of the liquid. Most of the solution, 85% or more, was retained in the PARE. The airborne release was low, (i.e., close to the level of spill releases). It would be appropriate to investigate a leak below the liquid level with pressure forcing the solution out. (This is a viable powder experiment also). These releases might be described satisfactorily using traditional spray relationships, and this could be confirmed by the suggested experimental work.

Our literature review (Sutter 1982) indicated little experimental work has been done to investigate flashing sprays. These releases occur in accidents where liquids pass from a region of high pressure to a low pressure environment. If the liquid vapor pressure exceeds the low pressure, some of the mass shifts into the vapor state to maintain equilibrium. Superheating of the upper zone relative to the lower zone of a system produces flashing. This type of release would relate particularly to fire accident scenarios where heated solutions pressurize and subsequently rupture containers. Thus, hot solutions are jetted into the air producing aerosols of fine particles. Currently, available empirical data are used to estimate drop sizes, requiring multiple models. Volume flow in flashing sprays has been calculated by summing over drop size ranges (Ostrowski 1966). Calculating from another study (Brown 1961) of flashing sprays, Mishima (1976) estimated that <0.3% of the amount released would be in a size range that could be transported downwind and become an inhalation hazard.

Because enclosure size limited our experiments, it would be desirable to perform pressurized releases in a larger enclosure, representing additional realistic situations. Using a larger enclosure would allow investigation of the release as a function of distance from the event.

Another matter of interest is the use of other particulate materials such as ion exchange resin in the PARE. Contaminated ion exchange resins have been involved in pressurized release accidents. A radioactively traced resin could be ejected from the chamber and particle size of the release compared to the original size. Work using this type of material indicates that the fraction less than 10 μm is insignificant (Ames 1959, Swift 1962). However, resins might be shattered in an event and produce fines. Or the resin could be degraded prior to the event and contain fines that would become airborne. The possibility that resins burn upon release exists, and a fire-generated aerosol is possible.

Pressurized releases into flowing airstreams are another potential study area. Since additional stresses can be anticipated in this situation, larger releases might be expected.

Thus, there are many options for continuing investigations of pressurized release accident scenarios. This project is not designed to be a study of all parameters, but rather an empirical look at lower and upper boundary airborne releases (with minimum parameter variation) within a nuclear fuel cycle manufacturing facility. Therefore, a few pressurized release accident scenarios could be identified to study for ongoing work.

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APPENDIX A

MASS AIRBORNE RESULTING FROM PRESSURIZED POWDER AND LIQUID RELEASES IN STATIC AIR

TABLE A.1. Mass Airborne from Pressurized Releases of DUO Powder

Source g	Rep.[a]	Run[b]	psig	Weight, g	Average, wt g	wt% Airborne	Avg wt% Airborne
350	1	1	500	29.7028		9	
	2	13	500	30.7083	30.2056	9	9
	1	6	250	19.1341		6	
	2	14	250	21.9318	20.5330	6	6
	1	4	50	7.0006		2	
	2	8B	50	6.7570	6.8788	2	2
100	1	7	500	21.2210		21	
	2	12	500	18.3406	19.7814	18	20
	1	3	250	12.6345		13	
	2	9B	250	13.3580	12.9963	13	13
	1	2	50	3.3166		3	
	2	11	50	3.7947	3.5557	4	4

(a) This is the replicate identification number
(b) Run number, used to identify experiment since they were done in a random sequence.

TABLE A.2. Mass Airborne from Pressurized Releases of TiO_2 Powder

Source, g	Rep.	Run	psig	Weight, g	Average, wt g	wt% Airborne	Avg wt% Airborne
350	1	5[a]	1000	15.8805	15.8805	4.5	4.5
	1	4	500	21.3782		6.1	
	2	16	500	34.2035	27.7909	9.8	7.9
	1	3	250	10.1143		2.89	
	2	14	250	26.4866	18.3005	7.6	5.2
	1	2[a]	100	3.2783	3.2783	0.94	0.94
	1	1	50	2.1848		0.62	
	2	15	50	12.5796	7.3822	3.6	2.1
100	1	7[a]	1000	12.3933	12.3933	12.4	12.4
	1	13	500	29.4976		29.5	
	2	18	500	18.7731	24.1354	18.8	24.1
	1	11	250	19.8395		19.8	
	2	9	250	16.5683	18.2039	16.7	18.2
	1	10[b]	189	11.0536	11.0536	11.1	11.1
	1	6	50	3.6082		3.6	
	2	7	50	5.1346	4.3714	5.1	4.4
50	1	8[a]	50	0.51	0.51	1.0	1.0

(a) Single value only
(b) Premature rupture

TABLE A.3. TiO_2 Powder Airborne, Powder Release Above Chamber (PRAC)

Source, g	Run	psig	Weight, g	wt% Airborne
350	1	500	38.2198	10.9
	5	250	36.6774	10.5
	4	50	15.8639	4.5
100	6	500	28.7369	28.7
	3	250	21.3226	21.3
	2	50	8.1773	8.2

TABLE A.4. Mass Airborne from Pressurized Releases of Uranine Solutions

Source cm^3	Rep.	Run	psig	Uranine Collected on Samples, g	Mass Airborne, g	Avg Mass Airborne, g	wt% Airborne	Avg wt% Airborne
350	1	1	500	2.0648×10^{-3}	0.206		0.06	
	2	13	500	1.2855×10^{-3}	0.129	0.167	0.04	0.05
	1	6	250	2.2858×10^{-4}	0.023		0.007	
	2	14	250	4.1983×10^{-4}	0.042	0.033	0.012	0.010
	1	4	50	1.2902×10^{-5}	0.0013		0.0004	
	2	8	50	4.2090×10^{-5}	0.0042	0.0028	0.0012	0.0008
100	1	7	500	1.8406×10^{-3}	0.18		0.18	
	2	12	500	1.1070×10^{-3}	0.11	0.15	0.11	0.15
	1	3	250	4.4711×10^{-4}	0.04		0.04	
	2	9	250	6.5027×10^{-4}	0.07	0.06	0.07	0.06
	1	2[a,b]	50	4.4217×10^{-6}	0.004		0.004	
	2	11	50	5.0652×10^{-5}	0.005	0.005	0.005	0.005

(a) Impactor sample suspect, removed from analysis
(b) 1g/ℓ, other runs used 10 g/ℓ solution

TABLE A.5. Mass Airborne from Pressurized Releases of UNH Solutions

Source					Uranium Airborne,	Mass[b] Airborne,	Avg Mass Airborne,	wt%	Avg wt%
cm^3	Uranium, g[a]	Rep.	Run	psig	μg	g	g	Airborne	Airborne
350	72.03	1	1	500	15,746	0.1180		0.0219	
		2	8		20,092	0.1504	0.1342	0.0279	0.0249
		1	5	250	7,261	0.0544		0.0101	
		2	10		6,386	0.0480	0.0512	0.0089	0.0095
		1	4	50	245	0.0016		0.0003	
		2	12		259	0.0022	0.0019	0.0004	0.0004
100	20.58	1	6	500	14,057	0.1052		0.0683	
		2	9		19,598	0.1466	0.1259	0.0952	0.0818
		1	3	250	9,156	0.0685		0.0445	
		2	11		12,058	0.0902	0.0794	0.0586	0.0516
		1	2	50	354	0.0026		0.0017	
		2	13		369	0.0028	0.0027	0.0018	0.0018

(a) Source = 0.2058 g/cm^3; $g/cm^3 \times cm^3$ = g in source
(b) Calculated using a measured UNH density of 1.54 g/cm^3

TABLE A.6. Volume of Liquid Ejected from the PARE

Pressure, psig	350 cm^3 Source		100 cm^3 Source	
	Uranine, cm^3	UNH, cm^3	Uranine, cm^3	UNH, cm^3
500	17, 10	10, 9	3, 6	9, 6
250	7, 8	8, 5	2, 10	5, 7
50	5, 9	4, 8	2, 4	3, 6

APPENDIX B

MEDIAN AERODYNAMIC EQUIVALENT DIAMETER (AED) OF PARTICLES GENERATED BY PRESSURIZED POWDER AND LIQUID RELEASES IN STATIC AIR

TABLE B.1. Median AED of Particles Produced by Pressurized Releases of DUO

Source, g	Rep.	Run	psig	Median Diameter, μm	σg	wt% 10 μm or Less	wt% of Source that Becomes Airborne 10 μm and Less
350	1	1	500	16	3.0	34	3.0
	2	13	500	16	3.0	31	2.7
	1	6	250	12	2.1	42	2.3
	2	14	250	18	3.0	29	1.8
	1	4	50	16	3.0	33	0.7
	2	8B	50	21	4.8	31	0.6
100	1	7	500	10	2.5	49	10.4
	2	12	500	18	4.0	38	7.0
	1	3	250	9	2.2	54	6.8
	2	9B	250	12	1.9	38	5.1
	1	2	50	13	2.9	41	1.4
	2	11	50	25	4.2	25	1.0

TABLE B.2. Median AED of Particles Produced by Pressurized Releases of TiO_2

Source, g	Rep.	Run	psig	Median Diameter, μm	σg	%10 μm or Less	wt% of Source that Becomes Airborne 10 μm and Less
350	1	5[a]	1000	7.0	4.1	64	2.9
	1	4[b]	500				
	2	16	500	15, 12	10.7, 8.6	40, 44	3.9, 4.3
	1	3[b]	250				
	2	14	250	14	8.5	44	3.3
	1	2[b]	100				
	1	1[b]	50				
	2	15[c]	50	20, 16	13, 10.3	38, 48	1.4, 1.7
100	1	7[a]	1000	5.6	8.2	60	7.4
	1	13	500	4.6, 4.2	6.3, 5.5	68, 72	20.1, 21.2
	2	18	500	6.9, 5.0	6.0, 6	54, 66	10.1, 12.4
	1	11	250	10, 8.3	9.5, 5.7	50, 54	9.9, 10.7
	2	9	250	17, 6.4	14, 5.8	42, 61	6.9, 10.1
	1	10[d]	189	14, 10	10.8, 4.5	44, 50	4.9, 5.5
	1	6	50	8.2	5.9	54	1.9
	2	17	50	9.2, 6.8	5.1, 4.3	52, 60	2.7, 3.1
50	1	8[e]	50	12	8.6	48	0.5

(a) Exploratory at 1000 psig
(b) Impactor overloaded
(c) Data based on 2 impactors where two sizes are shown
(d) Run originally set for 250 psig, went off prematurely
(e) Exploratory at 50 psig and using 50 g traced TiO_2

TABLE B.3. Median AED of Particles Produced by TiO_2 Powder Releases Above Chamber (PRAC)

Source, g	Run	psig	Median Diameter, μm	σg	%10 μm and Less	Avg wt% of Source that Becomes Airborne and is 10 μm and Less
350	1	500	20[a]	11.8	54	
			9	7	40	5.1
	5	250	11	5.5	48	
			5	3.6	72	6.3
	4	50	12	9.2	46	
			2.4	3.8	88	3.0
100	6	500	11	7.7	48	
			4.3	4.0	71	17.1
	3	250	10.1	5.3	48	
			6.2	4.4	65	12.0
	2	50	8.4	6.5	53	
			6	7.0	68	5.0

(a) Based on two impactor samples

TABLE B.4. Median AED of Particles Produced by Pressurized Releases of Uranine Solutions

Source, cm^3	Rep.	Run	psig	Median Diameter, μm	σ_g	% 10 μm and Less	wt% of Source that becomes Airborne 10 μm and Less
350	1	1	500	4	3	86	0.05
	2	13	500	4.3	2.4	85	0.03
	1	6	250	4	1.6	98	0.007
	2	14	250	3.6	3	84	0.01
	1	4	50	1.5	3	83	0.0003
	2	8	50	2.1	4	87	0.001
100	1	7	500	4	3	90	0.16
	2	12	500	4.0	4	70	0.08
	1	3	250	2.4	4	78	0.03
	2	9	250	4	3.6	77	0.05
	2	11	50	2.1	4	80	0.004

TABLE B.5. Median AED of Particles Produced by Pressurized Releases of UNH

Source, cm^3	Rep.	Run	psig	Median Diameter, μm	σg	% 10 μm and Less	wt% of source that Becomes Airborne 10 μm and Less
350	1	1	500	17	4.0	34	0.0074
	2	8		14	2.9	37	0.0103
	1	5	250	45	5.9	20	0.0018
	2	10		12	2.9	45	0.0040
	1	4	50	3	4.8	76	0.0002
	2	12		5	5.2	70	0.0003
100	1	6	500	14	2.5	46	0.0314
	2	9		14	2.9	38	0.0362
	1	3	250	14	2.7	36	0.0160
	2	11		11	4.6	45	0.0264
	1	2	50	6	7.5	61	0.0010
	2	13		8	4.6	60	0.0011

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Safety assessments and environmental impact statements for nuclear fuel cycle facilities require an estimate of potential airborne releases caused by accidents. Aerosols generated by accidents are being investigated to develop source terms for these releases. An upper boundary accidental release event would be a pressurized release of powder or liquid in static air. Experiments measured the mass airborne and the particle size distribution of aerosols produced by pressurized releases. Two powder and two liquid sources were used: TiO_2 and depleted uranium dioxide (DUO); and aqueous uranine (sodium fluorescein) and uranyl nitrate solutions. Pressurization level and source size were significant variables for the airborne powder releases. For this experimental configuration, the liquid releases were a function of pressure only. 100 g and 350 g of DUO (1 μm dia) and TiO_2 (1.7 μm dia) powders and 100 cm^3 and 350 cm^3 of uranine and uranyl nitrate solutions were released at pressures ranging from 50 to 500 psig. The average of the largest fractions of powder airborne was about 24%. The maximum amount of liquid source airborne was significantly less, about 0.15%. Powders ranged from 5 to 19 μm AED; liquids ranged from 2 to 29 μm. All of the releases produced a significant fraction of respirable particles of 10 μm and less.

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