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A New Analysis of the Vixen A Trials*

by

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ABSTRACT

A new analysis of the air sampler data from the two Vixen A Trials experiments involving chunks of plutonium exposed to hydrocarbon fuel fires indicated a respirable release fraction (referenced to total plutonium involved) of 0.0001 to 0.0003 (1E-4 to 3 E-4) depending on the parameters used in the atmospheric transport and dispersion model used. A value of 3 E-4 is recommended as an upper limit for use in safety assessment studies involving similar accident scenarios.

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Introduction

The source term for assessing events that involve the combustion of metallic plutonium (Pu) presents a continuing need in carrying out safety evaluations in support of DOE programs involving nuclear weapons. For production, storage, transport and decommissioning operations, an accident sequence that frequently must be evaluated involves metallic Pu being exposed in a fire environment. There are significant data on which to base an estimate for the source term which has been surveyed recently by Mishima et al (Mishima, 1993), but much of the surveyed work relates to well controlled laboratory experiments with relatively small amounts of Pu.

One of the most relevant sources of information was the work by Stewart (Stewart, 1959) of the UK Atomic Weapon Research Establishment at Aldermaston. That work, referred to as the Vixen A Trials provides direct experimental data on the aerosolization and environmental releases from relatively large metallic Pu shapes immersed in a "petrol" fire. While Stewart made some estimates of the aerosolized material released, he did not feel sufficiently confident in the data to recommend his estimated respirable release fraction value (3.5×10^{-5}) because it turned out to be smaller than that yielded by some laboratory experiments he had done earlier (5×10^{-4}). Given Stewart's concern it might be considered imprudent to try to extract more information from the Vixen data. However, the experiment is the only large scale experiment that is directly related to many of the scenarios being analyzed in safety studies and, as such, deserves additional efforts to extract useful information. The aim of this note is to provide an independent estimate of the total aerosolized and respirable release fraction for Pu generated in the two Vixen experiments and to estimate that part which was respirable.

Vixen Overview

The details of the experiments will not be repeated here, but two experiments were run outdoors in Australia. Each exposed nominally 200 gram cylinders of Pu metal supported in a mesh basket to a petrol fire in a makeshift chimney arrangement. Downwind of the fire, a grid of air samplers collected aerosol at distances of 200, 500, 1000, 1500, and 2000 yards. At 500 yards, balloon borne samplers provided vertical definition of the cloud at heights up to 300 feet. In addition to the air samplers there was definition of the fallout of the Pu from the aerosol plume at similar distances from the events. Crosswind average data from the air samplers at each range and aloft excerpted from the Vixen report are provided in Tables 1, 2, and 3.

Measurements were made to define the meteorological conditions during the experiments; these are excerpted in Table 4. The direction data indicate large variations in the wind direction that tended to "paint" the aerosol widely over the quarter circle air sampling grid in both experiments. Wind speed, time of day (early afternoon) and the standard deviation of wind direction in the vertical and crosswind directions (Sig y and Sig z) are consistent with unstable or possibly neutral atmospheric dispersion conditions for both tests.

Table 1: Air Sampler Data at Ground Level for VIXEN Experiment 1**

Distance (yards)	Stage *	Mean Collection (pCi)	Estimated Resp. Fraction	No. Samplers	Total# Collection (pCi)	SE of the Mean	Standard Deviation	Est. SD from Count Stat
200	1	248.50			5467.70			
	2	17.15			377.40			
	3	6.80			149.50			
	4	0.15			3.18			
	Filter	0.26			5.74			
	Total	272.86	7%	22	6003.52	NA	NA	NA
500	1	0.08			1.76			
	2	0.91			19.08			
	3	0.14			2.87			
	4	0.02			0.41			
	Filter	0.04			0.74			
	Total	1.18	74%	21	24.85	0.59	2.74	0.18
1000	1	0.04			0.92			
	2	0.04			0.90			
	3	0.02			0.45			
	4	0.06			1.23			
	Filter	0.03			0.59			
	Total	0.19	72%	21	4.09	0.031	0.144	0.1
1500	1	0.07			1.97			
	2	0.08			2.46			
	3	0.10			2.92			
	4	0.04			1.18			
	Filter	0.04			1.15			
	Total	0.32	73%	30	9.67	0.031	0.169	0.12
2000	1	0.02			0.33			
	2	0.02			0.41			
	3	0.06			1.19			
	4	0.09			1.70			
	Filter	0.04			0.89			
	Total	0.23	91%	20	4.52	0.038	0.172	0.11

Sum of Air Sampler Collections by Stage for all Samplers at Same Distance

* Stage Mass Median Diameters (in micrometres): 1=12; 2=4.0; 3=1.3; 4=0.5; Filter=all remaining

** Extracted From Table 10 of AWRE Report T15/60

Table 2: Air Sampler Data at Ground Level for VIXEN Experiment 2

Distance (yards)	Stage*	Mean Collection (pCi)	Estimated Resp. Fraction	No. Samplers	Total# Collection (pCi)	SE of the Mean	Standard Deviation	Est. SD from Count Stat
200	1	0.39			9.76			
	2	0.46			11.55			
	3	0.69			17.28			
	4	0.33			8.24			
	Filter	0.03			0.64			
	Total	1.90	73%	25	47.46	0.46	2.3	0.2
500	1	0.11			2.31			
	2	0.27			5.57			
	3	0.27			5.41			
	4	0.04			0.85			
	Filter	0.03			0.60			
	Total	0.72	75%	21		0.21	0.88	0.15
1000	1	0.02			0.39			
	2	0.03			0.61			
	3	0.06			1.27			
	4	0.04			0.83			
	Filter	0.02			0.49			
	Total	0.17	85%	21		0.044	0.2	0.1
1500	1	0.03			0.94			
	2	0.02			0.48			
	3	0.05			1.57			
	4	0.05			1.57			
	Filter	0.03			1.01			
	Total	0.19	81%	30		0.036	0.196	0.11
2000	1	0.02			0.41			
	2	0.08			1.56			
	3	0.06			1.25			
	4	0.06			1.11			
	Filter	0.06			1.27			
	Total	0.28	86%	20	5.59	0.033	0.147	0.011

Sum of Air Sampler Collections by Stage for all Samplers at Same Distance

* Stage Mass Median Diameters (in micrometres): 1=12; 2=4.0; 3=1.3; 4=0.5; 5=all remaining

** Extracted From Table 10 of AWRE Report T15/60

Table 3: Air Sampler Data at Elevations from 30 to 300ft at Range of 500 yd**

Sampler Height (feet)	Stage	Stage Collection (pCi)	Estimated Resp. Fraction	Stage Collection (pCi)	Estimated Resp. Fraction
Sampler A 30	1	0.04	76%	Sampler B 0.00	99%
	2	0.09			
	3	0.00			
	4	0.11			
	Filter	0.03			
	Total	0.27			
Grid Ref.	3549			3551	
150	1	0.05	80%		
	2	0.06			
	3	0.06			
	4	0.04			
	Filter	0.12			
	Total	0.33			
Grid Ref.	3550				
200	1	0.00	87%		
	2	0.14			
	3	0.05			
	4	0.00			
	Filter	0.09			
	Total	0.27			
Grid Ref.	3552				
250	1	0.15	43%		
	2	0.08			
	3	0.02			
	4	0.05			
	Filter	0.00			
	Total	0.29			
Grid Ref.	3550				
300	1	0.00	98%		
	2	0.02			
	3	0.00			
	4	0.11			
	Filter	0.14			
	Total	0.26			
Grid Ref.	3552				

* Stage Mass Median Diameters (in micrometres): 1=12; 2=4.0; 3=1.3; 4=0.5; 5=all remaining

** Extracted From Table 10 of AWRE Report T15/60

Table 4 - Meteorological Conditions for the Vixen Experiments

Experiment	Duration (minutes)	Avg. Wind Spd at 30 ft ht. (mph)	Mean Dir.	Direction Variation	Sig y (degrees)	Sig z (degrees)
Pu 1	23	9.2	93	+34 to -40	10	6
Pu 2	40	9.7	73.5	+68 to -68	27	9.8

*relative to mean direction

Analysis Concept

The air sampler data from the Vixen experiments provide the information needed to infer the source term for the two experiments. If a reasonably realistic model for the atmospheric transport of the aerosol is available, a correct model source term magnitude will predict the general features of the spatial distribution of air sampler data. "Realistic" in this context means a model that is simple enough to have a few parameters that can be adjusted to provide a fit and complex enough to be consistent with the type, quantity and accuracy of the data to be fitted. Selecting a realistic model was accomplished by noting certain characteristics of the air sampler data. The cross-arc data (not shown here) indicate collection of Pu aerosol on virtually all samplers over the entire array from the shortest to the longest distance. The angular width of the pattern confirms the wide wind direction variation over the duration of the experiment. The full quarter circle instrumented array received aerosol. Some small fraction may have passed outside the instrument array. The crossarc data at each range show little symmetry about the mean wind direction, but the highest collections are not at the edges of the array. This indicates that most of the material passed over the array. If the makeshift chimney were successful in producing a well defined aerosol plume, it might be expected that little aerosol would be collected near the ground at the shortest distance (200 yds). The presence of aerosol at the closest distance indicates that some fraction of the aerosol was released at, and moved along the ground. In the downwind direction, crosswind average sampler aerosol collections decrease as distance squared until 500 yd. Beyond about 500 yd., collections turn up, indicating increasing contribution from an elevated plume of aerosol from the chimney. The elevated samplers indicate a relatively flat profile from the ground to 300 feet which indicates relatively good vertical mixing.

Based on these observations, a two component model was postulated. One part was a ground level point source emitting aerosol into a sector of 90 degrees. The second was an elevated point source emitting aerosol into a 90° sector at a height which was a variable parameter in the model. The plume height, and magnitude of the respirable source term

and proportioning of source between the ground level and elevated sources was varied to achieve the best possible fit to the air sampler data.

Models Used

The models used for the calculation are Gaussian representations of aerosol dispersion taken from Meteorology and Atomic Energy (AEC, 1968) pages 405&406. The equations are:

$$TIC = \sqrt{\frac{2}{\pi}} \cdot QR \cdot \frac{n}{2\pi x U \sigma_z} \cdot \exp - \frac{1}{2} \left(\frac{h}{\sigma_z} \right)^2 ; \text{ see para 10, p405 of (AEC, 1968)}$$

$$SC = SR \cdot [a \cdot TIC(h) + b \cdot TIC(h = 0)]$$

$$\sigma_z = 0.53x^{0.73} \text{ (unstable) or } \sigma_z = 0.15x^{0.70} \text{ (neutral); p406; Table A2 of (AEC, 1968)}$$

$$QR = Q \cdot ARF \cdot RF = Q \cdot RRF$$

where:

- TIC = time integrated concentration (in source units - seconds/ metre cubed)
- QR = source strength of aerosol emitted in the experiment (in source units - e.g., grams, Curies, etc.)
- n = number of times the angular segment of aerosol exposure fits in a circle; for this effort = 4
- x = distance from emission point (in metres)
- U = wind speed (in metres/second)
- σ_z = vertical spread of aerosol in a Gaussian representation (in metres)
- h = plume height of emission (in metres)
- SC = sampler collection predicted (in source units)
- SR = volumetric rate for air sampler (in cubic metres per second)
- a,b = weighting factors for fitting to sampler data
- Q = mass of Pu exposed to fire
- ARF = aerosol release fraction
- RF = respirable fraction of ARF
- RRF = respirable release fraction

Determining the RF was accomplished by fitting the computed estimates of sampler collections to the data by visual evaluation. An initial estimate of the emission height needed to increase the ground level aerosol collection at ranges beyond 500 yards for unstable dispersion conditions indicated approximately 150 metres for Experiment 1 and 175 metres for Experiment 2 provided the best fit. For neutral dispersion conditions, the

necessary emission height was found to be about 50 metres. The values used for aerosol release fraction (ARF) for the two experiments were those determined by Stewart, that is, 1% and 3% for Experiments 1 and 2 respectively. The respirable fractions (RF) and the coefficients a and b were determined for each of the two Vixen experiments under both unstable and neutral conditions by trial and error in fitting the crosswind average air sampler collections for the 5 distances from 200 to 2,000 yards. The relative goodness of fit was judged by minimizing the sum of the logarithms of the ratios of measured and calculated collections over the 5 ranges (like fitting a curve on a log-log plot). Estimates obtained for RRF are shown in Table 5. The largest values of RF were found for unstable conditions. The RF for neutral conditions was found to be a factor of about 3 lower, but was still above the estimate made by Stewart based on a cross-arc and vertical integration of sampler collections to determine the mass of aerosol accounted for on the samplers at 500 yards. Stewart's RRF estimate of 3×10^{-5} is consistent with an assumption that there was no material above 300 feet or beyond the edges of the sampling array. The assumption of neutral conditions used here limited the vertical spread of aerosol to below 300 ft as Stewart assumed. Using unstable dispersion conditions, which is also justified by the meteorological data, acknowledges that some aerosol might have gone higher than 300 feet. While conditions more unstable than neutral seemed probable, it seems unlikely that fully unstable dispersal conditions were present. Thus, it seems prudent to base the respirable release fraction on the more conservative choice of dispersal conditions.

Figure 1 shows the Vixen data and the calculated estimates for unstable conditions that were judged to be the best fit for this study. The fit is not exact but the trend with distance, especially the decreasing slope of the curves beyond 500 yards is reasonably well portrayed. Uncertainty in the data and range to range variability makes achievement of a closer fit problematical.

Table 5. Fitting Parameters for Vixen Experiments

Experiment	a	b	RRF	h
1 unstable	0.35	0.65	1.9E-4	150m
2 unstable	0.06	0.94	3.0E-4	175m
1 neutral	0.15	0.85	1E-4	50
2 neutral	0.04	0.96	1.2E-4	50

Figure 1 - Fitting of Vixen Data with Dispersion Model (Unstable Conditions)

Conclusion

The results of this reevaluation of the Vixen Trial data for plutonium respirable release fraction (RRF) yielded values between $1\text{E-}4$ to $3\text{E-}4$. While these are above the $3.5\text{E-}5$ Stewart derived from the experiments but didn't recommend, they are also below the $5\text{E-}4$ which Stewart did recommend as a "prudent choice." This analysis suggests that Stewart's caution was well founded, but also suggests that analyses that require an expected value might use a somewhat lower value of $3\text{E-}4$ with a range of $\pm 2\text{E-}4$. This study also suggests that an RRF of $5\text{E-}4$ appears to be a reliable upper limit to Pu aerosolization from exposure to hydrocarbon fuel fires.

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