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TITLE: AEROSOLIZED U AND Be FROM LASL DYNAMIC EXPERIMENTS

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AEROSOLIZED U AND Be FROM LASL DYNAMIC EXPERIMENTS

Summary

At the request of OSD/ALO, the Los Alamos Scientific Laboratory conducted a field investigation to estimate the atmospheric release of toxic metals due to dynamic experimental activities at LASL. These dynamic experiments are routinely conducted on LASL controlled areas. These experiments typically employ conventional high explosives and may contain quantities of potentially toxic metals including beryllium, lead, mercury, and uranium.

The objective of the investigation was to determine the fraction of the total mass of uranium and beryllium aerosolized and transported in the debris cloud from each of three experiments conducted in early November 1974. Up to that time it was assumed the majority of the metals were aerosolized. The investigation was conducted by LASL with aircraft sampling support from EPA/ Las Vegas.

Aerosolized uranium and beryllium concentration and size distribution information was obtained by aircraft penetrations through the debris cloud. Cloud size and trajectory information were derived from photographic records taken from two orthogonally located camera sites. The total material in each cloud was then estimated using total cloud volume estimates, aircraft sample volumes, and observed concentrations of uranium and beryllium. This experimental technique is subject to significant uncertainties arising from cloud sizing, material distribution in the cloud, and aircraft penetration path. However, this approach appeared to be the most feasible and straightforward way to obtain a reasonable estimate of material contained in the debris clouds.

Results indicated that approximately 10% of the total mass of uranium and 2% of the beryllium were contained in the debris clouds. The aircraft impactor data on aerosolized uranium showed a log-normal distribution with an aerodynamic mass median diameters of 0.1 to 1 μm and standard geometric deviations of

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about 8 for the three experiments. Each of the three experiments sampled were different in character and thus the results should be reasonably representative of the spectrum of experiments routinely conducted at LASL.

These experimental results were then used to estimate the theoretical contribution of dynamic experimentation to atmospheric concentrations of these metals for a typical year in the Los Alamos environs. Annual atmospheric concentration estimates were obtained using the measured aerosolized values and through the use of a time-integrated version of the Gaussian puff model. For calculation purposes the following assumptions were made:

1. Initial cloud diameter and height of 100 meters
2. Average wind speed of 3 m/s
3. Equal probability of transport in all directions
4. Slightly unstable atmosphere.

The table below gives results on an annual basis:

ESTIMATES FOR 1976

Element	Annual Usage (Kg)	Percent Aerosolized	Annual Avg. Conc. (ng/m^3) 4 km	8 km	Applicable Standard (ng/m^3)
Uranium (D-38)	1023	10	0.1	0.04	9000
Be	25.5	2	0.0007	0.0002	10 (30 day avg)
Hg	36.1	100*	0.05	0.02	None
Pb	18.6	100*	0.02	0.08	None
		Totals	0.17	0.068	10000 [For total heavy metals N > 21]

*Assumed values.

A national emission standard for Hg is 1 $\mu g/m^3$ averaged over one day. The time integral for a single experiment consuming the total of 16.4 kg of Hg (1976 monthly maximum) is 0.8 $\mu g\text{-}d/m^3$ (80% of standard) with the assumptions below:

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1. Neutral atmosphere,
2. Average wind speed + 2 m/s,
3. Downwind sampling distance = 4 km, and
4. Initial cloud diameter and height = 100 m.

While no standards exist for Pb the annual concentration average at 4 km is about 5% of the amount expected from the resuspension of continental dust.

Approximately 100,000 kilograms of uranium have been used in dynamic experiments at LASL since 1943. The average yearly use for the last 22 years is 2466 (σ = 1309) kilograms. In recent years use has dropped to approximately 1000 kg/yr. Atmospheric uranium concentrations have been routinely measured at LASL by a 26 station air sampling network. The spatial average concentration for uranium in 1976 was 0.06 ng/m^3. The expected levels of uranium due to the resuspension of continental dust is 0.08 ng/m^3 ($\pm$ a factor of 2). Network sampling results for airborne uranium (0.06 ng/m^3 annual average) for 1976 are reassuring because the aerosolization percentages and crude dispersion model used do not underestimate dynamic experimentation contributions (calculational estimates ranged from 0.04-0.10 ng/m^3).

Specific information about the experiments conducted and computational analysis performed are contained in the attached appendices.

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APPENDIX A

AIRCRAFT SAMPLING FOR URANIUM AND BERYLLIUM FROM LASL DYNAMIC EXPERIMENTS

I. Introduction

The Los Alamos Scientific Laboratory is engaged in dynamic experimentation. These experiments are conducted on LASL controlled areas. The experiments typically employ conventional high explosives and may contain quantities of potentially toxic metals including beryllium, lead, mercury, and uranium. It was the object of this investigation to determine the amounts and nature of beryllium and uranium released to the atmosphere as a consequence of such experimentation. Beryllium and uranium were selected for several reasons. One, they are commonly released elements of significant environmental interest. Secondly, they display different chemical behavior: uranium being chemically reactive and frequently pyrophoric, whereas beryllium is more chemically inert and refractory. Additionally, these elements have good analytical detectability.

II. Sampling Techniques

Three dynamic experiments were sampled in early November 1974. Since the experiments disperse material into the atmosphere by way of a buoyant, explosive-products cloud, air samples were collected by aircraft rather than by conventional ground-based sampling. Although there is a considerable "wake" of dust and debris blown out along the ground following a detonation, the subsequent dust cloud was believed to be the major source of atmospherically dispersed material. Aircraft sampling was conducted by the Radiation Monitoring Branch of the U.S. Environmental Protection Agency, National Environmental Research Center, Las Vegas, NV using the EPA twin turbine Beechcraft airplane. Aircraft sampling equipment included an on-board high volume (hi-vol) air filtration system and a wing mounted cascade impactor sampler for particulate size analysis.

The sampling probe for the hi-vol sampler consisted of a tube extending from the nose of the craft to the sample collection area located near the center of the plane. The air flow of the hi-vol sampler was automatically maintained at a rate

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of 4.8 ± 0.2 x 10^{-2} m^3/s (102 CFM), a flow rate approximately 70% of an isokinetic sampling rate. Atmospheric aerosol samples from the hi-vol sample were collected on 20-cm x 25-cm, Whatman No. 41 cellulose filter paper. Rapid filter changing capabilities permitted sequential hi-vol sampling.

The wing-mounted cascade impactor was of the hi-vol, multistage, multislit design, incorporating five impaction stages and a standard hi-vol backup filter. Particles were collected on approximately 14-cm x 14-cm, parallel slit impaction paper, also of Whatman No. 41 cellulose. For sub-micron particles, a backup filter of 20-cm x 25-cm Whatman No. 41 was used. The sampling pod containing the cascade impaction sampler had an aerodynamic flow which maintained the impaction sampling rate at 1.9 ± 0.1 x 10^{-2} m^3/s (40 CFM) for the aircraft sample collection speed. The impaction system was designed for isokinetic aerosol sampling, and calibration at one atmosphere and 298K gave the following particle size cut-off diameters (in μm) at 50% collection efficiency for spherical particles of unit density: Stage 1, 8.2 and greater; Stage 2, 3.5 to 8.2; Stage 3, 2.1 to 3.5; Stage 4, 1.0 to 2.1; Stage 5, 0.5 to 1.0; and Filter Stage, below 0.5.

III. Sampling Missions

The aircraft sampling missions were flown on two days in November 1974. Experiment I, sampled during the afternoon of the first day, contained uranium-238 (D-38) and beryllium in addition to stainless steel and other materials. Prior to detonation, the aircraft circled the firing point at a distance of about 3 km. Radio communications were established between a central ground observation point and the firing site, the aircraft, and two ground photographic observation points. At detonation, the aircraft began its first approach to the visible explosion cloud with synchronized still photography commencing at the two photographic observation points. Standard 6-cm x 6-cm color positive slides were taken at 30-s intervals and at times of visible aircraft penetration through the cloud. A movie camera also chronicled the event. Continuous tape recordings were made of activities and communications at each of the three ground sites so that the data could be synchronized to a common time frame.

At 65 s after detonation, the aircraft made its first penetration of the cloud at an altitude of approximately 122 m above the firing point. A total of five cloud penetrations were made, the last of which was approximately 7.6 min after detonation. Aircraft cloud penetration data are presented in Table I. The visibility of the dust cloud was greater from

0007808

the vantage point of the aircraft than from the ground observation points. Thus, aircraft samples were taken even though ground visual observation had been lost. Similarly, photographic visual observations were inferior to aircraft sighting.

On the second day, two experiments were sampled; Experiment II in the morning and Experiment III in the afternoon. The sequence of events for these mission paralleled that of Experiment I. Pertinent aircraft sampling data are given in Table I. Experiment II contained D-38 and beryllium; Experiment III contained D-38.

Following each penetration through a cloud, the hi-vol filter was changed and packaged, providing an independent sample for each penetration. Each hi-vol filter sample represented a sampling period of approximately 0.5 min, although only a few seconds were actually spent traversing the cloud. Preceding each shot, a "background" hi-vol sample was taken which represented approximately 0.5 min of sample collection of ambient air. The cascade impactor remained open during the entire airborne mission, and integrated size fractionated samples representing the composite of all passes through a cloud were obtained.

IV. Analysis

The air filter samples were chemically analyzed for beryllium and uranium by Group H-5. Filter samples were first wet ashed to dryness with concentrated sulphuric and nitric acids (and with H_2O_2 and $HClO_4$ as needed). After dryness the samples were brought to a standard volume of 10 mℓ with 0.1 N H_2SO_4. An aliquot of from 1. to 2 mℓ was used for the beryllium analysis, performed with a Perkin-Elmer Model 303 Atomic Absorption Spectrophotometer with nitrous oxide flame. From the remainder of the sample solutions, a standard aliquot was taken for uranium analysis. Dried uranium aliquots were fused with NaF and analyzed fluorometrically with a Jarrell Ash Fluorometer. This instrument is insensitive to isotopic variations and measures only total uranium mass.

Table II gives the results of the chemical analyses of the filter samples collected during the sampling missions. Errors associated with mass analyses are one standard deviation (σ). For the uranium analysis, a filter "blank" value of 0.9 ± 0.6 μg/hi-vol filter and 0.3 ± 0.2 μg/impactor filter was subtracted from the data. For beryllium, the filter blank is below the level of detection, but the errors associated with these values reflect the uncertainty of the blank

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value. The 2σ 95% confidence detection limit for beryllium is 0.1 μg.

It is apparent from Table II that the levels of uranium collected were much larger than the levels of beryllium. This was expected since the experiments generally contain several kg of D-38 and around one-half kg of beryllium. Note that the beryllium data for Experiment III confirm the absence of beryllium.

Photographic triangulation was used to determine cloud size and location at 30-s synchronized intervals after detonation. An attempt was made to locate the camera sites for orthogonal photographs. This was limited somewhat by access restrictions and available viewing platforms. Each camera had a 6-cm x 6-cm (2-1/4 x 2-1/4 in.) format and a 150-mm lens focused at infinity. This resulted in a 3.78° angular change per cm of movement on the film. In order to maintain an absolute frame of reference, each photograph shared some distinct topographical feature with the preceding photograph to permit the cameras to be reaimed without losing the absolute frame of reference. This was necessitated by the rapid movement of the clouds.

Photographic analysis consisted of computing the azimuthal angle between the cloud center and alternate camera site for each pair of frames. The two azimuthal angles were then used to located a vertical line of azimuthal intersection. The elevation angle of cloud center for each of the frame pairs was used to compute a separate cloud height. These two values were averaged and the result used as a cloud center location. Angular data of cloud horizontal and vertical width were then converted to length dimensions for each paired frame. The resultant four width values were then available for cloud volume estimation.

V. Results

In estimating the fraction of uranium and beryllium aerosolized during a dynamic experiment, the following methodology for hi-vol data was employed: (1) an estimate of the cloud diameter at the time of aircraft sampling was obtained from the photographs, (2) the volume of the cloud was calculated assuming sphericity, (3) a sample collection time was determined given the true air speed of the aircraft and and approximate diameter of the cloud, (4) of the total cloud volume, the volume actually sampled by the aircraft was calculated from the aircraft collection time and the sample collection rate, (5) an average uranium or beryllium concentration in the volume sampled was calculated, (6) a total uranium or beryllium mass in the cloud at the time of sampling

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was estimated, assuming the aircraft sample was representative of the entire cloud, and (7) the dispersed fraction of the total mass of uranium or beryllium in the experiment was calculated. The most uncertain aspects of these determinations were estimating the cloud diameter and the assumption of cloud sphericity.

The cloud diameters at time of penetration are presented in Table III; diameter uncertainties were approximately 20%. Upon calculation of a spherical cloud volume, errors from 50% to 100% were propagated. There was essentially no ground visual cloud observation for aircraft passes three through five of Experiment I, two and three of Experiment II, and two through four of Experiment III, because of dispersion. Consequently, estimates of an equivalent cloud diameter and diameter error were established from systematic extrapolation of cloud expansion based on photographic cloud size data. From the photographs and triangulational measurements, it was possible to trace the vertical and horizontal movement of the "center" of the clouds. The actual shapes of the clouds, and variations in dust density within the clouds, were undeterminable with photographic evidence from ground observation points. It was apparent from the photographs and ground observations that the clouds dispersed somewhat randomly with a tendency toward an elongated, oblate shape. It is obvious that the aircraft was unable to fly directly through the centroid of the cloud. In order to compensate for the lack of knowledge of the true shape of the cloud, and the non-central penetration by the aircraft, an equivalent "diameter" of the cloud was determined by averaging the vertical and horizontal widths established from triangulation measurements. The standard deviation of these four measurements was then used as the conservative error of the equivalent cloud diameter. Sample collection times were determined by dividing the equivalent cloud diameter by the true air speed. In order to further compensate for cloud shape and penetration representativeness uncertainties, conservative errors of from 100% to 300% were assumed for aircraft sample collection times for passes where the cloud size was estimated from systematics.

Table III verifies the intuitively obvious notion that for later aircraft passes through a cloud, the amount of useful information obtained diminishes. For pass five of Experiment I, for example, the element concentrations in the fraction of the cloud sampled dropped erratically, apparently as a result of poor visibility, poorly defined cloud shape, and nonrepresentative samples. Passes two and three of Experiment II and passes three and four of Experiment III also appear to be nonrepresentative samples. Interpretation of data from these passes was therefore avoided.

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In order to determine the fraction of uranium and beryllium which was aerosolized and suspended in the cloud, the amounts of these elements prior to detonation were needed. Table IV gives the quantity of D-38 and beryllium used in the experiments.

Despite the large errors associated with the mass of uranium and beryllium in the clouds (columns 9 and 10 of Table III), these values are relatively small percentages (columns 11 and 12) of the total element mass available for dispersion. For Experiment I (excluding the nonrepresentative pass 5), integrating samples from passes one through four gives 314 ± 31 μg of uranium and 2.63 ± 0.14 μg of beryllium collected from a cumulative cloud fraction of 26 ± 13 x 10^{-8}. Assuming these are representative samples, a total of 1.2 ± 0.6 kg of uranium and 10 ± 5 g of beryllium were then dispersed, or 10 ± 5% and 1.6 ± 0.8%, respectively, of the initial amounts.

For Experiment II, only the first pass can be trusted, and it was calculated that the cloud contained 0.13 ± 0.10 kg of uranium and 9 ± 7 g of beryllium. This corresponds to 3 ± 2% and 1.9 ± 1.5%, respectively, of the initial amounts of uranium and beryllium in the device. Passes one and two of Experiment III were composited to establish a total uranium mass in the cloud of 0.31 ± 0.27 kg which represents 12 ± 10% of the total D-38 in the device. There was no beryllium in Experiment III as the chemical analyses corroborate. The maximum limit of beryllium resuspended in the cloud by the explosion was 1 g.

An estimate of the elemental component of the particle size distributions was obtained from the hi-vol impactor samples. Detectable quantities of uranium were collected on nearly all impaction stages for each test shot (see Table II). These data are represented in Fig. 1 as cumulative distribution plots using the aerodynamic diameter particle size and uranium mass. The aerodynamic diameter is the size of a unit density aerosol particle with inertial characteristics equivalent to the particle actually collected. Because of the density difference, the aerodynamic diameter of a uranium particle is larger than the real diameter. Aerodynamic diameter is generally used because of its greater significance in inhalation/ingestion studies. The activity median aerodynamic diameters AMAD (the aerodynamic diameter for which half of the uranium mass is above and half is below) for Experiment II and III are 1 μm and 0.9 μm, respectively. As is evident from their relatively straight curves, Experiment II and III demonstrate a characteristic log-normal size distribution. The standard geometric deviations σg for these log-normal distributions are approximately 8, indicative of a highly skewed particle size distribution. The

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size distribution plot of Experiment I is less amenable to a log-normal treatment but clearly demonstrates a wide range of particle sizes. The AMAD for Experiment I was not measurable but can be estimated at 0.1 μm or less. Analogous particle size distribution plots for beryllium were not made because of detection limitations. Nevertheless, the data of Table II indicate that the most significant fraction of the atmospheric beryllium had an aerodynamic diameter of less than 0.5 μm.

In conclusion, for the experiments studied, the percent of uranium (D-38) dispersed in the detonation cloud was approximately 10%. For beryllium, the percent dispersed was approximately 2%. Furthermore, the uranium particle size distribution of these dispersed aerosols is approximately log-normal with mass median diameters in the range 0.1 to 1 μm and standard geometric deviations of about 8.

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TABLE I

AIRCRAFT SAMPLING MISSION DATA

Experiment I: PM

Cloud Sampling Pass	Time After detonation(s)	Elevation Above firing point(m)	True Air speed(m/s)	Visual Clarity of cloud
1	65	122	67	Good
2	145	104	67	Fair
3	225	137	67	Poor
4	300	137	67	Very Poor
5	455	229	67	None

Experiment II: AM

1	85	159	75	Fair
2	165	214	76	Poor
3	270	217	78	Very Poor

Experiment III: PM

1	50	107	72	Fair
2	120	107	69	Poor
3	185	168	70	Very Poor
4	255	183	70	None

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TABLE II

CHEMICAL ANALYSIS OF AIR FILTER SAMPLES

Filter Identification		Beryllium Mass Collected (μg)	Uranium Mass Collected (μg)
Experiment	Type		
I	Background Hi-Vol	<0.1	0.2±0.03
I	1st penetration Hi-Vol	1.64±0.07	206 ± 30
I	2nd penetration Hi-Vol	0.39±0.07	42.2±3.1
I	3rd penetration Hi-Vol	0.32±0.07	37.5±3.1
I	4th penetration Hi-Vol	0.28±0.07	27.9±3.1
I	5th penetration Hi-Vol	<0.1	8.7±0.7
I	1st Stage Impactor	<0.1	7.6±0.4
I	2nd Stage Impactor	<0.1	9.7±0.4
I	3rd Stage Impactor	<0.1	6.1±0.4
I	4th Stage Impactor	<0.1	4.9±0.4
I	5th Stage Impactor	0.12±0.07	2.4±0.4
I	Backup Impactor Filter	0.44±0.07	48.8±6.0
II	Background Hi-Vol	<0.1	1.1±0.03
II	1st Penetration Hi-Vol	1.80±0.07	25.5±3.1
II	2nd Penetration Hi-Vol	0.37±0.07	1.5±0.7
II	3rd Penetration H-Vol	<0.1	5.0±0.7
II	1st Stage Impactor	<0.1	6.3±0.4
II	2nd Stage Impactor	<0.1	5.7±0.4
II	3rd Stage Impactor	<0.1	4.2±0.4
II	4th Stage Impactor	<0.1	5.2±0.4
II	5th Stage Impactor	<0.1	4.4±0.4
II	Backup Impactor Filter	0.24±0.07	17.4±0.4
III	Background Hi-Vol	<0.1	1.3±0.03
III	1st Penetration Hi-Vol	<0.1	57.1±3.1
III	2nd Penetration Hi-Vol	<0.1	15.1±0.7
III	3rd Penetration Hi-Vol	<0.1	0.1±0.6
III	4th Penetration Hi-Vol	<0.1	0.3±0.6
III	1st Stage Impactor	<0.1	3.3±0.4
III	2nd Stage Impactor	<0.1	3.3±0.4
III	3rd Stage Impactor	<0.1	2.4±0.4
III	4th Stage Impactor	<0.1	2.1±0.4
III	5th Stage Impactor	<0.1	2.5±0.4
III	Backup Impactor Filter	<0.1	9.4±0.4

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TABLE III

URANIUM AND BERYLLIUM DETERMINATION FOR DEBRIS CLOUDS

Experiment	Pass	Cloud Diameter (m)	Cloud Volume ($10^6 m^3$)	Collection Time (s)	Volume Sampled (m^3)	Elemental Conc. of Sample ($\mu g/m^3$)		Total Element Mass in Cloud		Dispersed Fraction of Total Mass	
						U	Be	U(kg)	Be(g)	U(%)	Be(%)
I	1	98±22	0.5±0.3	1.5±0.3	0.07±0.02	$(2.9\pm0.8)\times10^3$	23±6	1.4±1.1	11±8	11±9	1.8±1.3
I	2	158±28	2.1±1.1	2.4±0.4	0.11±0.02	$(3.7\pm0.7)\times10^2$	3.5±0.8	0.8±0.4	7±4	6±3	1.2±0.7
I	3[a]	202±32	4.3±2.1	3.0±100%	0.15±0.15	$(2.6\pm2.6)\times10^2$	2.2±2.2	1.1±1.2	9±11	9±10	1.5±1.8
I	4[a]	216±32	5.3±2.4	3.2±200%	0.16±0.31	$(1.8\pm3.6)\times10^2$	1.8±3.6	1.0±2.0	10±12	8±16	1.6±2.0
I	5[a]	240±40	7.2±3.6	3.6±300%	0.17±0.52	$(0.5\pm1.5)\times10^2$	≲ 0.6	0.4±1.1	≲ 4	3±9	≲ 0.7
II	1	78±20	0.3±0.2	1.0±0.3	0.05±0.01	$(5.1\pm1.5)\times10^2$	36±9	0.13±0.10	9±7	3±2	1.9±1.5
II	2[a]	104±34	0.6±0.6	1.4±200%	0.07±0.13	$(2.3\pm4.7)\times10^1$	6±11	0.01±0.03	3±7	0.2±0.7	0.6±1.5
II	3[a]	118±40	0.9±0.9	1.5±300%	0.07±0.22	$(6.9\pm21)\times10^1$	≲ 1.4	0.06±0.19	≲ 1	1.5±4.6	≲ 0.2
III	1	88±22	0.4±0.3	1.2±0.3	0.06±0.02	$(9.9\pm2.6)\times10^2$	< 1.7	0.4 ±0.3	<0.6	16±12	-
III	2[a]	136±40	1.3±1.2	2.0±200%	0.10±0.19	$(1.6\pm3.2)\times10^2$	≲ 1.1	0.2 ±0.5	≲1.4	8±20	-
III	3[a]	168±60	2.5±2.7	2.4±300%	0.12±0.35	0.9±5.8	≲ 0.9	0.002±0.014	≲2.2	0.1 ±0.6	-
III	4[a]	190±80	3.6±4.5	2.7±300%	0.13±0.39	2.3±8.3	≲ 0.8	0.01 ±0.03	≲2.8	0.4 ±1.2	-

[a] For this aircraft penetration, there was no direct photographic determination of cloud diameter. The diameter was estimated from systematics of cloud expansion. Greatly increased errors in cloud traversal time (fifth column) reflect the uncertainty of this assumption.

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TABLE IV

BERYLLIUM AND D-38 IN EXPERIMENTS

	Mass of Element in Experiments (kg)	
	Be	D-38
Experiment I	0.61	12.3
Experiment II	0.48	4.1
Experiment III	-	2.5

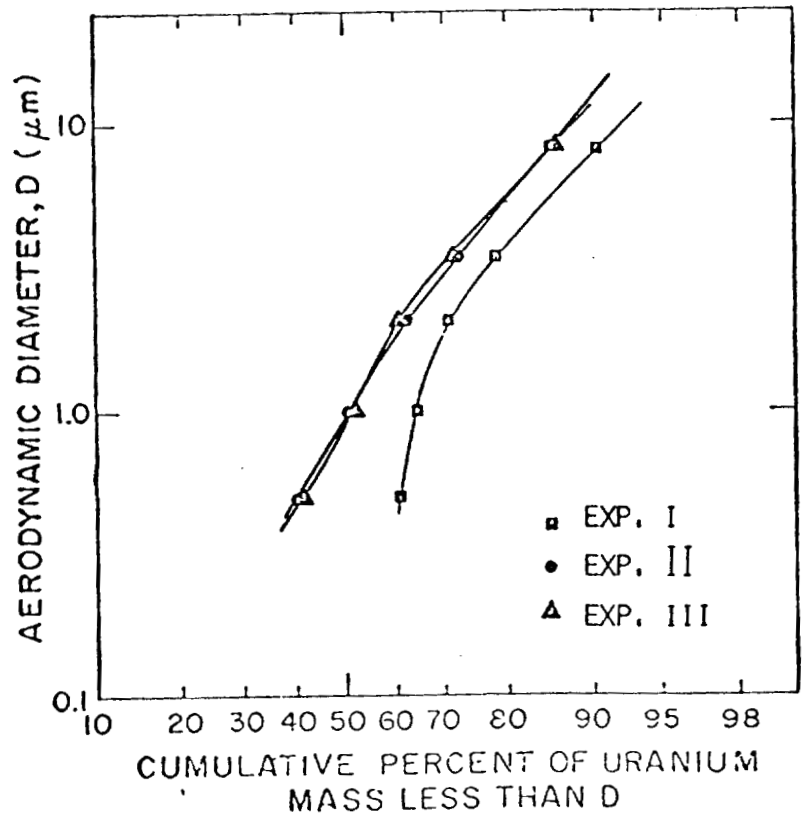

Fig. 1. Uranium particle size distributions.

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APPENDIX B

IMPLICATIONS OF AIRCRAFT SAMPLING FOR URNAIUM AND BERYLLIUM FROM LASL DYNAMIC EXPERIMENTS

I. INTRODUCTION

Appendix A contains the details of a study to determine the percent of uranium and beryllium carried aloft by the dust cloud from dynamic experiments at LASL. From this study, we obtained a qualitative description of the nature of an explosives products cloud, and we have an estimate of an atmospheric dispersion factor for two important species, uranium and beryllium. Given this albeit limited "source term" information, we can now make some etimates of atmospheric dispersion from these operations for potentially harmful elements.

II. DISPERSION MODEL

We employ here the modified Sutton equation for Gaussian diffusion as proposed by Turner[1] for instantaneous sources:

$$\chi(x,y,0;H) = \frac{2Q_T}{(2\pi)^{3/2}\,\sigma_x\,\sigma_y\,\sigma_z} \exp\left[-1/2\left(\frac{x-ut}{\sigma_x}\right)^2\right] \exp\left[-1/2\left(\frac{H}{\sigma_z}\right)^2\right] \exp\left[-1/2\left(\frac{Y}{\sigma_y}\right)^2\right]$$

where χ is the concentration in mass/m^3,

$z = 0$ = ground level, Q_T is the total mass of the element of interest which is present in the "puff", H is the effective height in meters of release of the puff (i.e., the height at which the puff ceases to be super-buoyant), u is the wind speed in m/s, t is the time after release, and the σs are empirical Gaussian dispersion parameters (which are different from those used for the dispersion estimates of a continuous source). Our findings of activity mean aerodynamic diameter values of $\lesssim 1$ μm for uranium and beryllium in the dispersion clouds permit treatment of these elements as gasses.

Fig. 1 shows cloud diameter as a function of time, and Fig. 2 gives the relation of cloud height and time after detonation for the three

[1] D. B. Turner, "Workbook of Atmospheric Dispersion Estimates", U. S. Dept. Health, Education, and Welfare, Public Health Service Publication No. 999-AP-2, revised 1969.

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experiments studied. It is apparent from these curves that there is an initial rapid increase in cloud diameter and height which is a function of its heat and buoyancy factors. After a few minutes, however, the puff becomes somewhat stabilized, and normal dispersion factors begin to take affect. For these dispersion estimates, then, we can consider a puff at time t = 0 (≈ detonation time) with diameter D and height H. A program has been written for the calculation of $\chi_{max}(x,0,0;H)$ using the above equation. The values of $\chi_{max}(x,0,0;H)$ are for maximum concentration, i.e., when t = x/u along the wind direction axis where y = 0. In this program, initial dimensions of the puff are used to extrapolate an up-wind "virtual" point source for the puff. The virtual source location increases in the upwind direction for increasing initial diameter of the puff. Estimations of dispersion parameters for a quasi-instantanous source, taken from Slade[2], are as follows:

Meteorological condition	x = 100 m		x = 4 km	
	σ_y	σ_z	σ_y	σ_z
unstable	10	15	300	220
neutral	4	3.8	120	50
very stable	1.3	0.75	35	7

We have used a standard power function $\sigma = Ay^B$ to fit these points in order to obtain interpolated and extrapolated values of σ_y and σ_z. Also, it is assumed that $\sigma_x = \sigma_y$. Consequently this model becomes less valid for distances greater than ≈10 km.

Plots of χ_{max}/Q_T obtained from the program are shown in Figs. 3 and 4 (the computer graphics routine is responsible for the slightly irregular shape of the dashed curve of Fig. 4). From Figs. 1 and 2 it is reasonable to assume an initial puff diameter of between 100 and 200 m and an initial height also between 100 and 200 m. Fig. 3 is for a puff with initial height and diameter equal to 100 m, and for Fig. 4, the initial height and diameter are 200 and 100 m, respectively. In Fig. 4 the curve for χ_{max}/Q_T under stable conditions is not given because it would be off scale for regions where the model is applicable. For 20 km, the value of χ_{max}/Q_T is $3 \times 10^{-29}/m^3$.

APPLICATION TO RELEASE OF D-38

Applying this model to the release of uranium (D-38) in an explosives products cloud, we can estimate a time average off-site concentration. For a location directly down wind from the source, the time integral of the concentration can be approximated by

$$\int_0^\infty \chi(x,0,0;H)dt = \chi_{max}(x,0,0;H) \frac{(2\pi)^{1/2}\sigma_x}{u} \text{ ; where } \chi_{max}(x,0,0;H)$$

[2] D. H. Slade, Ed., "Meteorology and Atomic Energy - 1968", U.S. Atomic Energy Commission, 1968.

0007819

is the maximum concentration, i.e., for time $t = x/u$. An average distance from the firing points to populated off-site areas is about 8 km, whereas 4 km is approximately the minimum distance to populated off-site areas. For a distance of 8 km, considering the worst meteorological condition (neutral condition, Pasquill category D) for the dispersion of a puff with the conservative initial diameter and height of 100 m, a χ_{max}/Q_T of 1.2×10^{-8} m^{-3} is expected. For a typical D-38 Q_T value of 1 kg as observed in the aircraft sampling study, and a wind speed of 2 m/s, the maximum concentration would be 12 $\mu g/m^3$, and the time integral of the puff would be 3.8×10^{-3} $g\text{-}s/m^3$. When averaged over the period of a month, the average concentration from this release would be 1.5 ng/m^3.

For a distance of 4 km from the source to a populated area, a neutral meteorological condition is again most restrictive. The approximate χ_{max} and time integral for the above incident at a distance of 4 km are 24 $\mu g/m^3$ and 4.2×10^{-3} $g\text{-}s/m^3$, respectively.

Most of the dynamic experiments conducted at the LASL firing points are between the hours 10 AM and 4 PM. Hence a slightly unstable meteorological condition (Pasquill category C) with wind speed around 3 m/s could be considered as a conservative dispersion condition. For 1974, the total D-38 used in the shots was 1020 kg, and based on the aircraft sampling study, about 1/10 of this was released to the atmosphere via the puffs. An annual average D-38 concentration at a distance of 8 km would then be 0.04 ng/m^3 (using an equi-directional wind dilution factor of $(2\pi)^{1/2}\sigma_y/2\pi x$). For a distance of 4 km, the annual average for the same conditions would be 0.1 ng/m^3. (Using a conservative average meteorological condition of neutral with a wind speed of 2 m/s, the annual average D-38 concentration at 8 km and 4 km are 0.15 and 0.2 ng/m^3, respectively.)

The 1974 annual average uranium concentrations observed in the air monitoring network ranged from 0.04 ($\pm 2\sigma = 0.02$) to 0.15 ($\pm 2\sigma = 0.04$) ng/m^3. The spatial average concentration was about 0.09 ng/m^3 which is approximately equal to the expected uranium concentration of 0.08 ng/m^3 ($\pm$ a factor of 2) resulting from the resuspension of continental dust. (For a given release incident, the air sampling network may "miss" the puff, but it is believed that the annual average data adequately represent the real atmosphere being sampled.) Thus, it would seem that the model is applicable, in that it does not underestimate expected concentrations. It is reasonable to deduce that annual average D-38 concentrations in the LASL environs resulting from diagnostic test operations are in the range 0.001 to 0.1 ng/m^3 with typical off-site maximum episodic concentrations in the range 1 to 100 $\mu g/m^3$.

III. APPLICATION TO RELEASE OF BERYLLIUM

The 1974 annual diagnostic tests usage of Be was about 14 kg. We observed about 10 g (2%) of the initial mass of Be to be aerosolized in the puffs studied. Applying the assumptions used in the D-38 calculations, we can then estimate maximum episodic Be concentrations at 4 and 8 km to be 0.2 and 0.1 $\mu g/m^3$, respectively. Annual average off-site Be concentrations at 4 and 8 km are approximately 0.4 and 0.1 pg/m^3, respectively. These concentrations incorporate the 2% atmospheric-dispersion factor, but if this factor is significantly different for the annual average release, then the above concentrations would have to be appropriately corrected.

National Emission Standards for stationary Be sources may not apply to these tests, but the restriction of 10g/d is probably being exceeded, even though the 30-d average concentrations limit of 0.01 $\mu g/m^3$ is apparently satisfied. The National Emission Standard for Be from rocket motor firing is 75 $\mu g\text{-}min/m^3$ accumulated during any two-week period. A typical off-site (4 km downwind) episodic Be concentration integral is about 0.2 $\mu g\text{-}min/m^3$ and hence this standard is apparently being met.

IV. FURTHER CONSIDERATIONS

Other metals of interest which are released to the atmosphere by diagnostic tests are mercury, lead, and antimony. The 1974 annual usage for these elements, and for D-38 and Be, is shown in Table I. Applying similar arguments to the release of these elements, calculated annual average and episodic concentrations are shown in Table I. In the absence of experimental information, we assume atmospheric dispersion factors of 100% for Pb, Hg, and Sb. The National Emission Standard for Hg of 1 $\mu g/m^3$ averaged over one day is apparently not being violated since a worst-case (neutral meteorological condition, $u = 2$ m/s, $x = 4$ km, cloud height = diameter = 100 m, mass = 1 kg Hg) episodic concentration time integral is about 0.05 $\mu g\text{-}d/m^3$. As there are no applicable emission standards for Pb and Sb, the calculated annual average Pb and Sb concentrations should be compared to the expected ambient concentrations of 0.4 ng/m^3 and 6 pg/m^3, respectively, resulting from the resuspension of continental dust. There is a New Mexico Standard for ambient air restricting total combined heavy metals ($N > 21$) to less than 10 $\mu g/m^3$.

It should be remembered that the model employed in these determinations is at best qualitative. The atmospheric dispersion of contaminants in complex terrain in all probability has scant resemblance to dispersion estimated with the Gaussian puff model. These estimates could therefore be erroneous by as much as one or possibly two orders of magnitude. However, for atmospheric uranium,

0007821

the only element for which we have atmospheric elemental concentration data, it appears that the model has not underestimated expected off-site concentrations resulting from dynamic experiments.

In a recent meeting with A. J. Toy of LLL, a further complication concerning the atmospheric dispersion of D-38 was revealed. Moving pictures of weapons test shots taken at LLL's Site 300 indicated that shrapnel D-38 could be a significant source of aerosolized uranium. Upon explosion, ignited pieces of what were undoubtedly D-38 were dispersed, some of which dramatically disintegrated by a "star burst" in mid air. Such aerosolized uranium would in general not be observed by our aircraft sampling missions. Conceivably, this secondary, pyrogenic source of uranium aerosol could be as important as the initial blast. Our "source term" for aerosolized uranium does not consider this secondary mechanism. Nevertheless, our aerosolization factor of ≈10% cannot be erroneous by more than a factor of ten, and this error is probably less than that which is inherent in the meteorological dispersion assumptions.

TABLE I

CALCULATED ATMOSPHERIC CONCENTRATIONS OF ELEMENTS USED IN LASL DYNAMIC EXPERIMENTS (1974 DATA)

Element[1]	1974 Annual Usage (kg)	Annual Avg. Conc. at 4 km (ng/m^3)	Max Episodic Conc.[2] at 4 km (mg/m^3)	Annual Avg. Conc. at 8 km (ng/m^3)	Max. Episodic Conc.[2] at 8 km (mg/m^3)
D-38	1020	0.1	0.02	0.04	0.01
Be	14	0.0004	0.0002	0.0001	0.0001
Hg	11	0.02	0.02	0.005	0.01
Pb	39	0.05	0.02	0.02	0.01
Sb	0.2	0.0005	0.005	0.0001	0.003

[1] The atmospheric dispersion factors for D-38, Be, Hg, Pb, and Sb are assumed to be 10%, 2%, 100%, 100%, and 100%, respectively.

[2] The total episodic releases for D-38, Be, Hg, Pb, and Sb are assumed to be 1, 0.01, 1, 1, and 0.2 kg, respectively.

0007822

CLOUD DIAMETERS FOR EXPERIMENTS

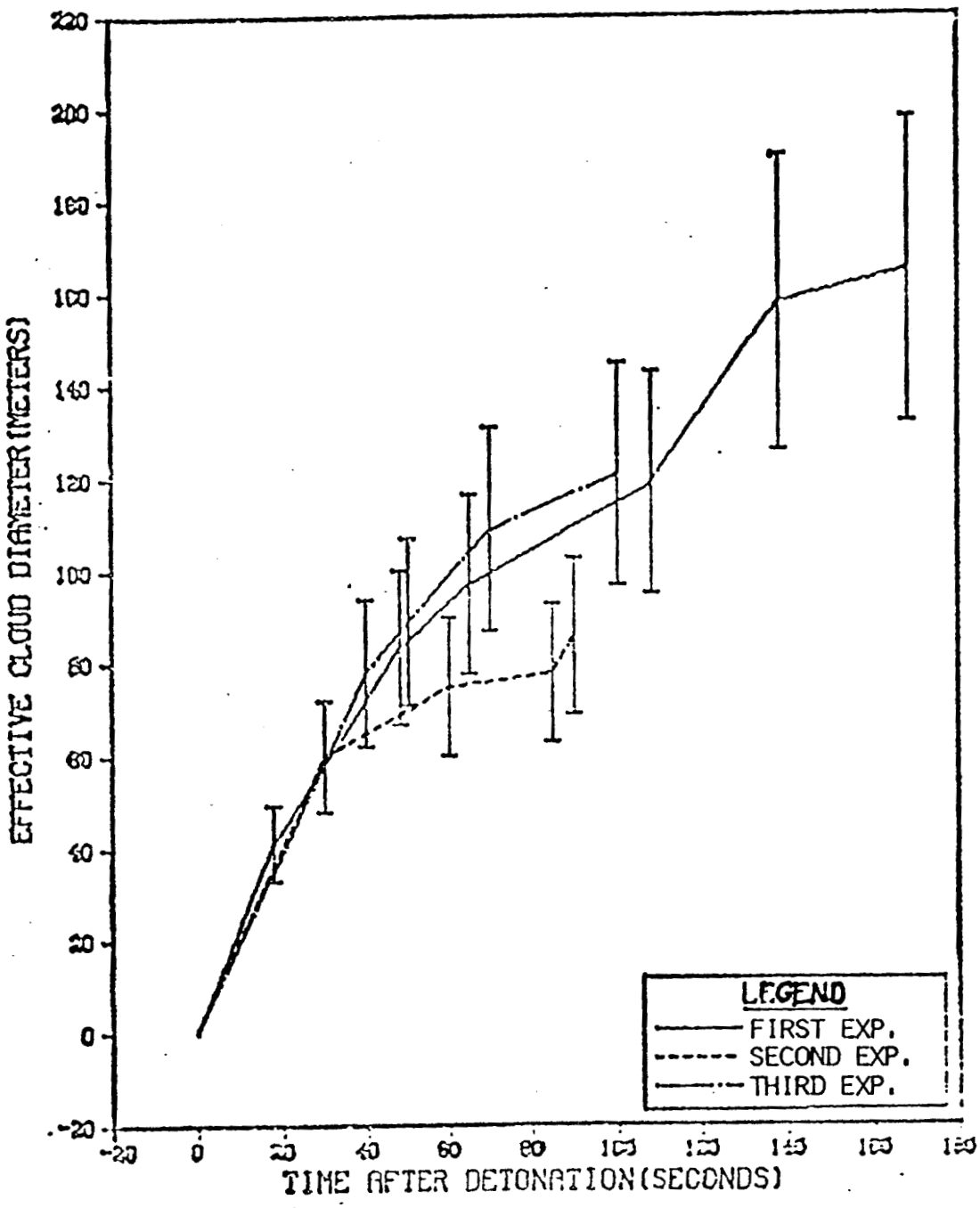

Fig. 1

CLOUD HEIGHTS FOR EXPERIMENTS

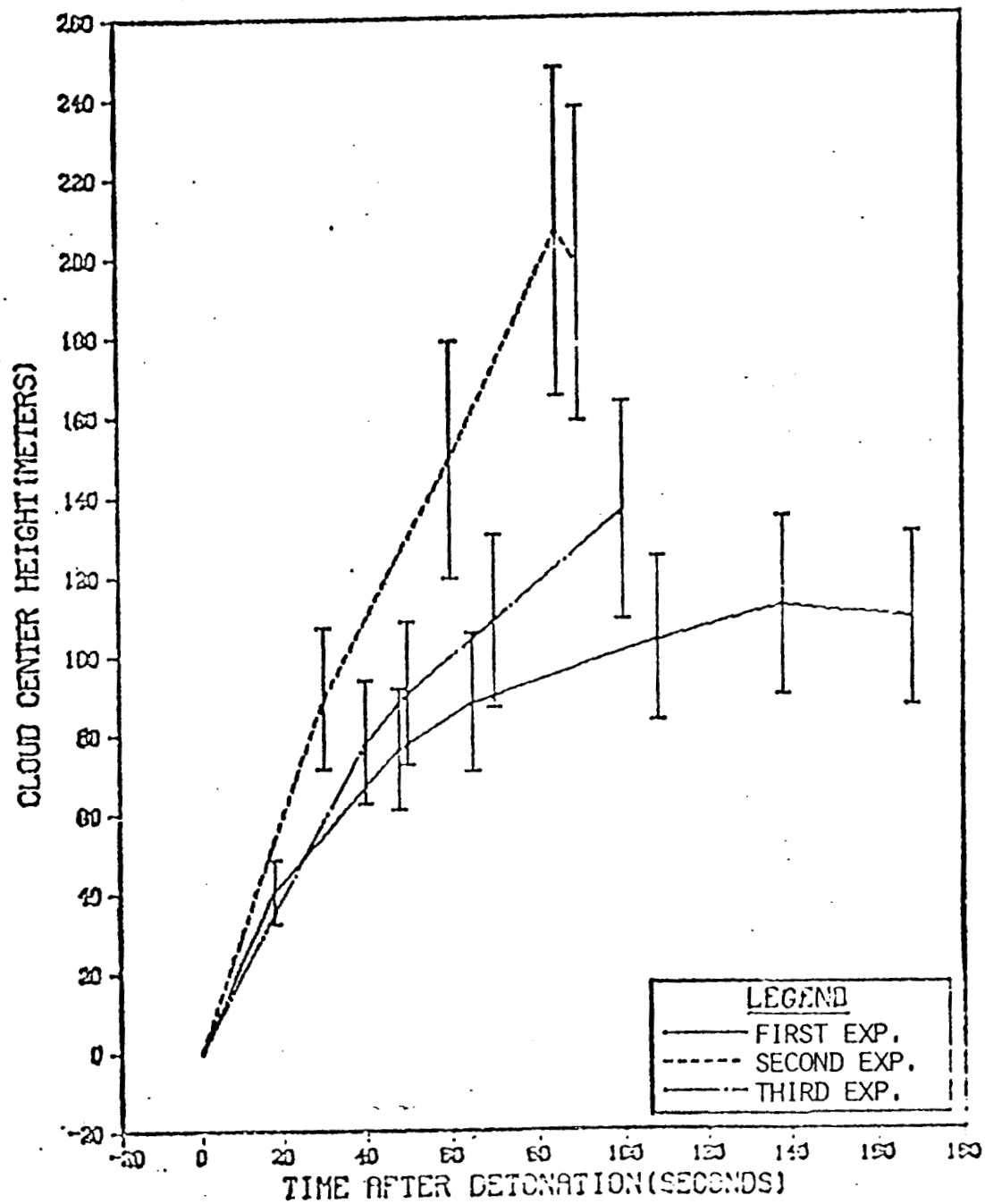

Fig. 2

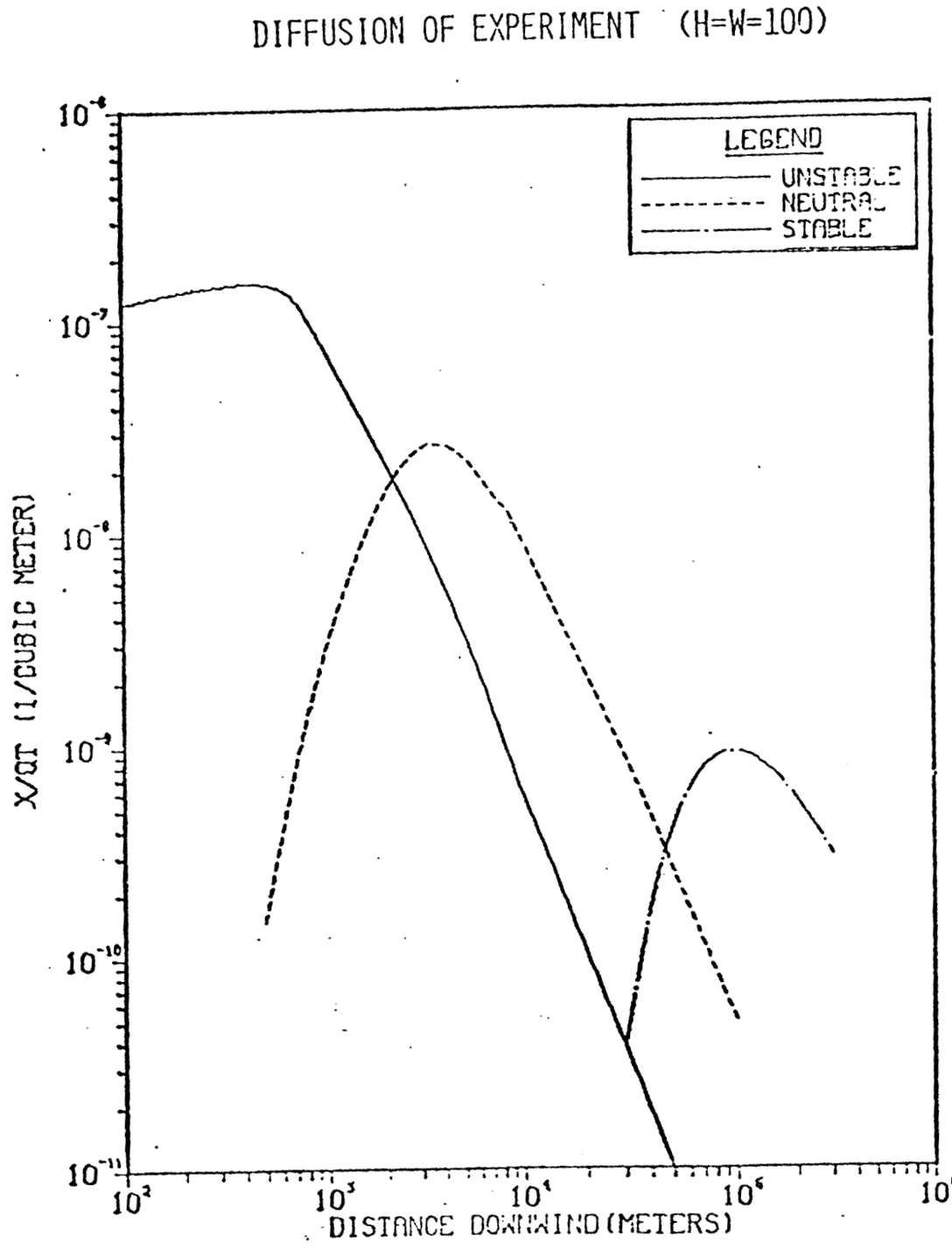

Fig. 3

0007825

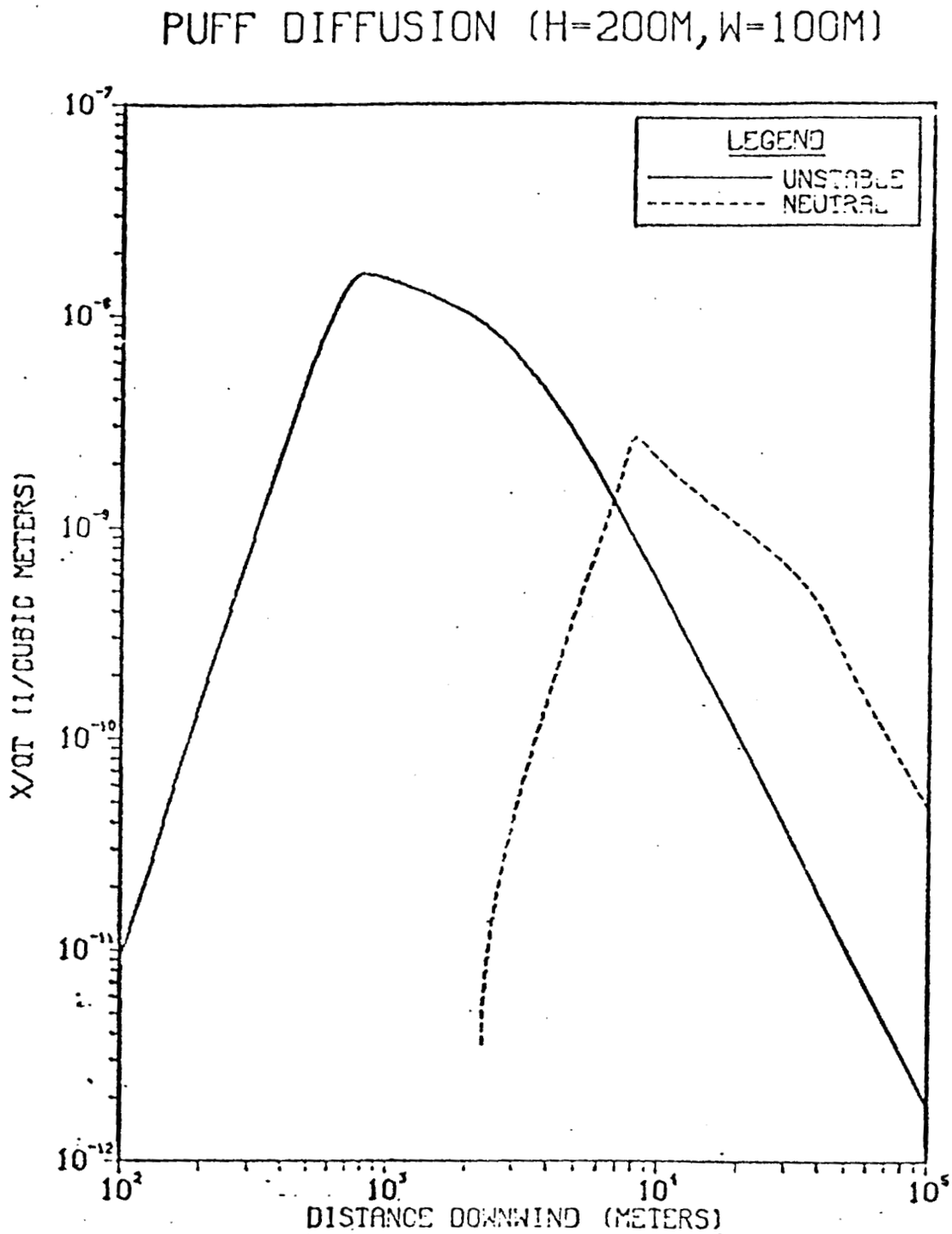
PUFF DIFFUSION (H=200M,W=100M)
LEGEND
UNSTABLE
NEUTRAL
X/QT (1/CUBIC METERS)
DISTANCE DOWNWIND (METERS)

Fig. 4