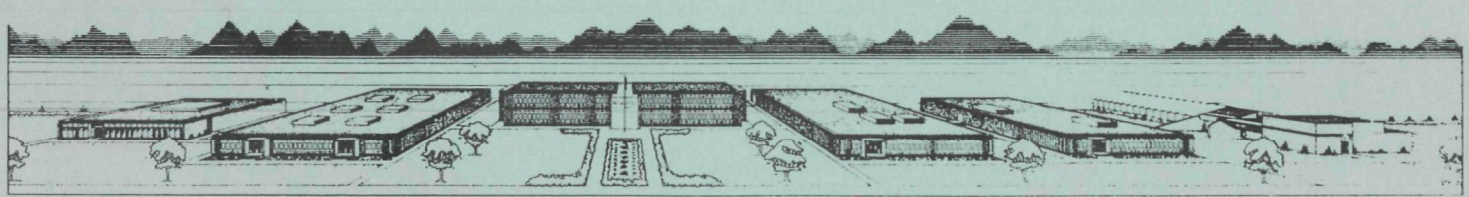
PARTICULATE EFFLUENT STUDY
PHOEBUS 1B, EP-IV

by the
Southwestern Radiological Health Laboratory

U. S. Department of Health, Education, and Welfare
Public Health Service
Environmental Health Service

April 1970

This surveillance performed under a Memorandum of
Understanding (No. SF 54 373)
for the
U. S. ATOMIC ENERGY COMMISSION



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ABSTRACT

The Southwestern Radiological Health Laboratory (SWRHL) of the U. S. Public Health Service performed, under a memorandum of understanding with the AEC, a study concerned with delineating the physical and chemical characteristics and possible hazards associated with release of particulate matter (greater than several microns in diameter) from the Phoebus IB, EP-IV reactor run conducted February 23, 1967. The reactor test was part of the Project Rover Program and was conducted at Jackass Flats, Nevada.

The particle deposition occurred in a general northerly direction from the test cell. Particles were found out to 82 miles with the results indicating a decrease in deposition (particles/unit area) with distance to about the 2.5 power. The particle size distribution, of all the particles collected, is reasonably described by a log normal distribution with a geometric mean diameter of about 12μ and a geometric standard deviation of 2.7. A breakdown of the size distribution to those particles 10μ and above gave a geometric mean of 26μ and geometric standard deviation of 2.

The density of 8 particles (12 - 28μ) was found to be about 11 g/cc. This density indicates an equivalent aerodynamic geometric mean diameter of about 40μ . Thus, the majority of the particles found and studied were larger than an equivalent diameter of 10μ - the usual cut-off for lower respiratory tract penetration. A regression analysis indicated a decrease in particle size and activity with distance.

Isotopic results showed a large degree of fractionation of the fission

products found in the particles.

Electron microprobe analysis indicated uranium, carbon, and oxygen to be present in most of the particles analyzed.

Particles were transported into the off-site area. The resulting ground concentrations were about 1 particle/100 m^2 or less and there was no known interaction of particles with people from the general population. Thus it is concluded there was no hazard to the public from the "particulate effluent."

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I. INTRODUCTION

Project Rover reactor tests are conducted at the Nuclear Rocket Development Station (NRDS), Jackass Flats, Nevada.* The station is about 90 miles NE of Las Vegas, Nevada.

The Project Rover reactors are based on a single pass gas-cooled design. The reactor cores are composed of annular fuel rods made up of UC_2 beads in a graphite matrix. The coolant, hydrogen, is passed through the core and expelled to the environment via the reactor nozzle. The hydrogen is burned after exiting from the reactor and this thermal energy, plus the effluent kinetic and thermal energy, produces a cloud or effluent rise of better than 1500 meters.

It has been noted over a period of several years that the effluent from Project Rover reactor runs contains large radioactive particulate matter in addition to gaseous and normal atmospheric size particulate matter. The large particulate matter is composed of actual segments of the reactor core released by what is termed the "corrosion process," whereas the radioactive gaseous effluent results from diffusion of the fission products in the core. The particles can contain fission product inventories up to the order of microcuries in quantity. Several of the particles were detected 30 miles downwind of the test cell after the NRX-A5, EP-IV test on June 23, 1966 (1400 - 1430 PDT). This discovery, coupled with

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*The Project Rover Program is charged with development of a nuclear reactor for rocket propulsion for deep space exploration. The program is administered by SNPO, the Space Nuclear Propulsion Office.

increasing reactor power, caused concern about the possible hazards associated with the deposition of particles in populated areas.[6]

The physical harm from interaction of these particles with individuals is not fully understood. Areas of possible concern are skin, eye, and lung doses. This report is concerned primarily with determining the physical parameters of the effluent versus other studies concerned with their biological interaction.

This report presents the results of particle studies for the Phoebus IB test series, Experimental Plan IV conducted by the Southwestern Radiological Health Laboratory.* It supersedes our previous reports on the subject. The event was conducted at 1400 PDT on February 23, 1967, at Test Cell C, NRDS. The reactor was in the inverted position, with the effluent expelled vertically upward. The power integral was about 3×10^6 Mw-sec with 30 minutes (1400-1430 PST) at full power, 1500 Mw. The work reported here was performed under a memorandum of understanding between the Public Health Service and the Atomic Energy Commission who are responsible for off-site safety. Effluent from the reactor test was distributed in a general northerly direction. Particles were found up to 82 miles from the reactor.

Project Objectives

The general objectives of the SWRHL studies were to determine the extent of downwind deposition of particles and their chemical and physical characteristics. Specifically the objectives were to determine an indication of the following:

A. Downwind concentration of the particles on the ground as

*The Phoebus IB test series was conducted by the Los Alamos Scientific Laboratory as part of the reactor development program for Project Rover.

a function of distance from the test cell. The prime emphasis was on downwind instead of crosswind concentrations.

B. Particle size distribution for all of the particles and for various distances downwind.

C. Constituent radioactive composition of the particles:

1. Gross alpha, beta, and gamma.

2. Specific isotopic composition - including the isotopic fractionation.

D. Correlation of the parameters: particle size, activity, and distance of collection.

E. Elemental and chemical composition.

F. Particle density.

G. Possible hazard to the general population in the off-site area.

After collection of the particles, a few were used in a study of biological clearance rates in rats. The primary objective of the study was to develop methodology for future studies.

II. RESULTS AND DISCUSSION

During a six-day period after the reactor test, 53 areas, generally 300 ft^2 or more in area, were surveyed for particles using an open window E-500B survey instrument held about 6 inches above the ground (see Appendix A for methods). The areas surveyed were in a northerly direction from Test Cell C at distances between 9 and 115 miles. The sampling locations were chosen on the basis of particle survey results, vegetation profiles, tracking by aircraft, and to some extent by terrain features.[1]

Particles were located in 21 of the monitored areas and 228 indications of particles were obtained.* This information is presented in Figure 1 and Appendix B. The general effluent hotline, based on three arcs where vegetation samples were collected (two on-site and one on Highways 6 and 25) and the area of highest air concentration (according to air samples taken along Highway 25), is also indicated in Figure 1. Specific results for air and vegetation samples are given in Reference 1.

Figure 2 indicates the fallout deposition pattern determined by Edgerton, Germeshausen and Grier, Inc. on February 24. The pattern was determined using a calibrated crystal in an airplane flying at about 500 feet above the surface.

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*Effluent from reactor cool-down was carried by the night-time drainage winds over the Lathrop Wells area (approximate azimuth 210°). This area, along Highway 95, was monitored on February 27 and there was no indication of particulate activity.

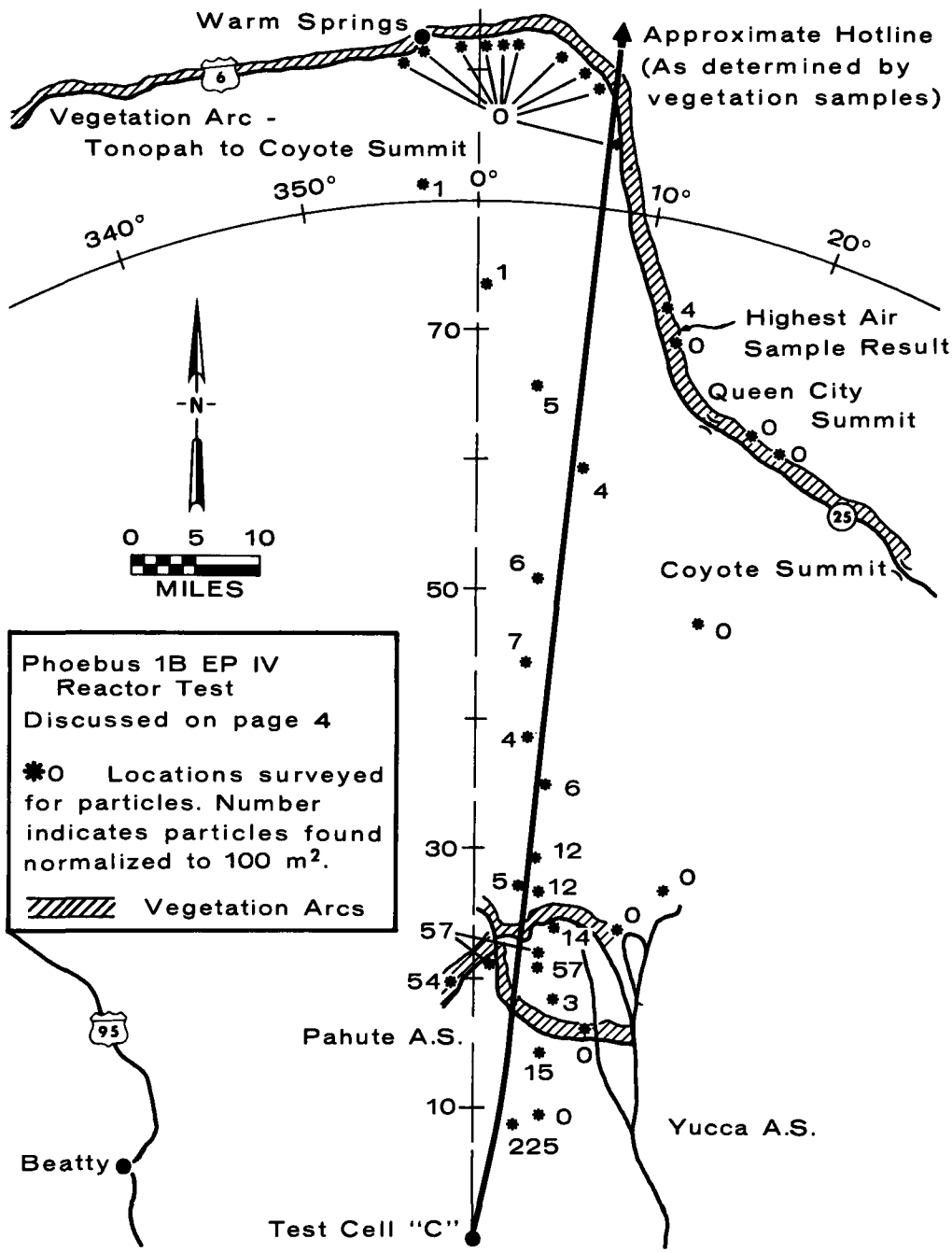

Figure 1. Particle sampling areas.

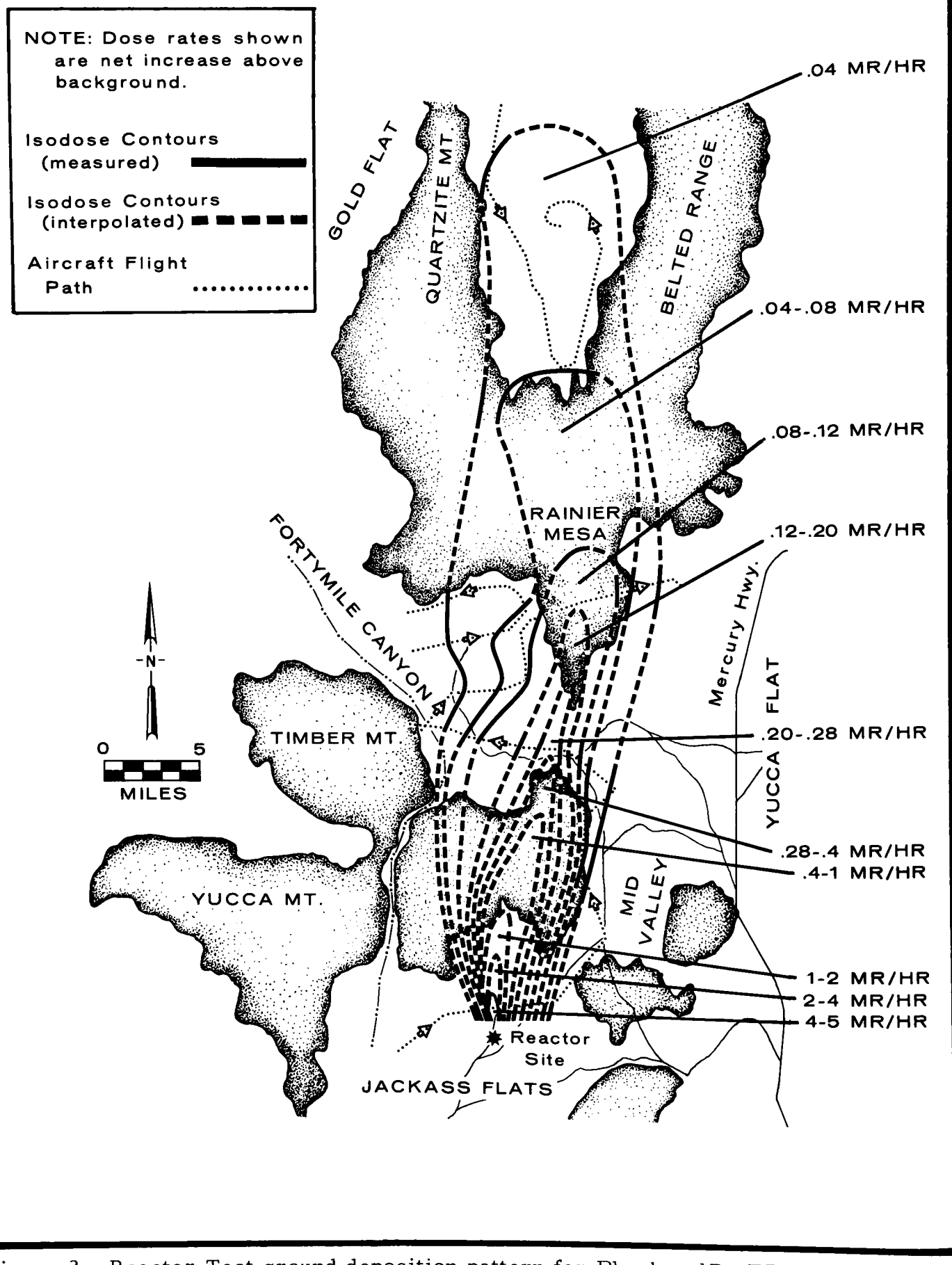

Figure 2. Reactor Test ground deposition pattern for Phoebus 1B, EP-IV, 24 February 1967. Discussed on Page 4.

A comparison of Figures 1 and 2 shows that the particle ground survey was generally performed in the area of highest deposition. Thus, although the ground survey primarily determined the downwind distribution rather than both downwind and crosswind, it is felt that it should be reasonably representative of the particle hotline. An exception is the points at less than about 25 miles from the test cell, where our surveys appear to be generally east of the hotline. A composite of the studies indicates the debris hotline was on an azimuth between 5 - 10°. [1, 2, 3] It appeared to start NNE and "back" more northerly with distance. Using ESSA/ARL upper level weather data (radar) collected at 1430 on February 23, 1967, over Jackass Flats, Nevada, an attempt was made to determine the initial height attained by the particles. The hodograph (Figure 3) shows that a particle hotline of 7° would correspond to an effective release height of 10,500 feet MSL and mean layer wind speed of about 12 mph.

A. Particle Concentration with Distance

The number of particles detected on the ground per $100m^2$ as a function of distance is presented in Figure 4. The indicated line is based on regression or least squares analysis of the data (log-log). Only those sampling points near the hotline (as indicated by this study, the Pan American study, and other effluent studies) were used in this analysis. Fourteen of the 21 sampling points were used. The slope of the line is -2.5 and the correlation coefficient for the indicated line is -0.95 or between -0.83 and -0.98 at the 95% confidence level.*

*The correlation coefficient squared is an indication of the percent of the variation of the data explained by the regression line.

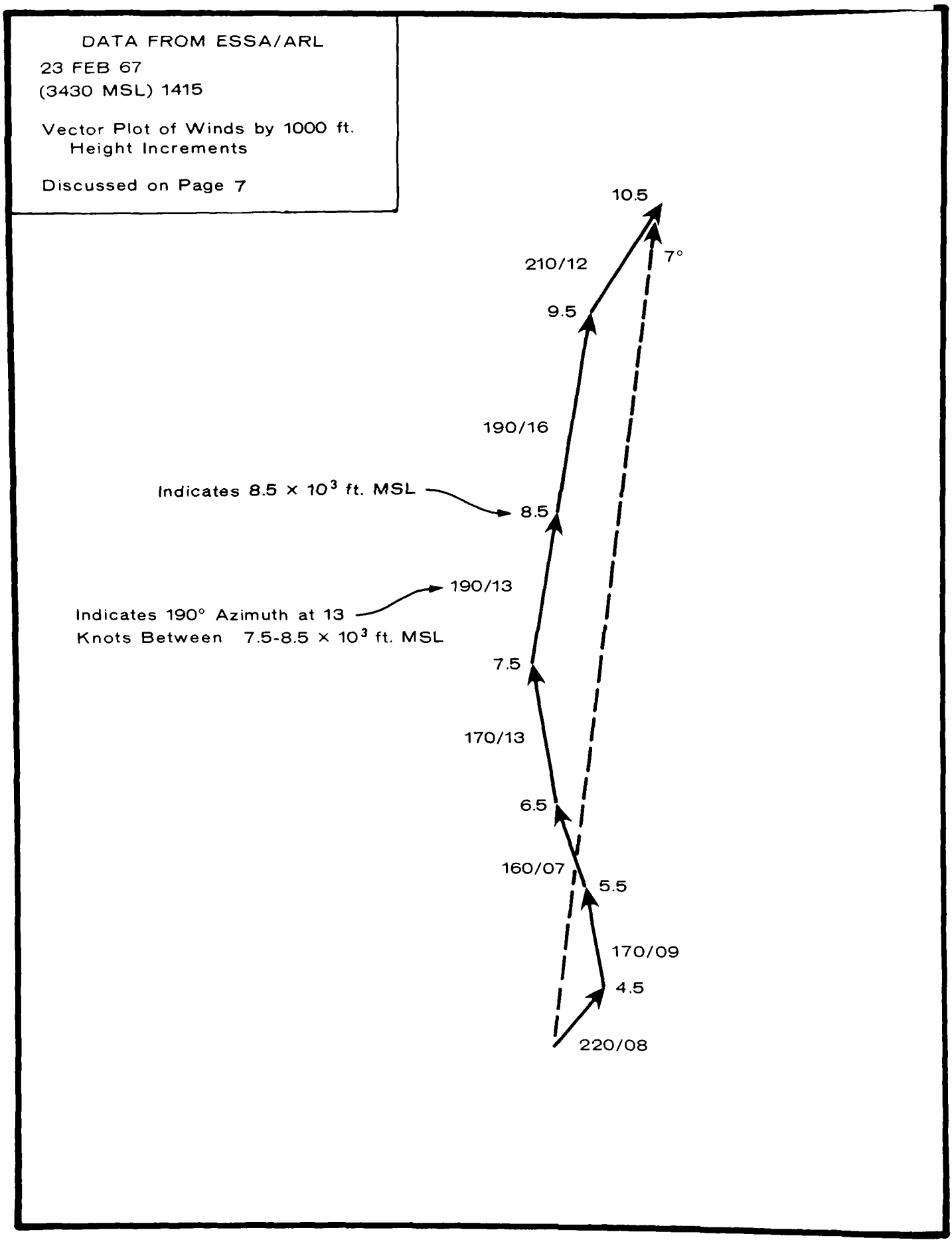

Figure 3. Hodograph for Jackass Flats, Phoebus 1B, EP-IV.

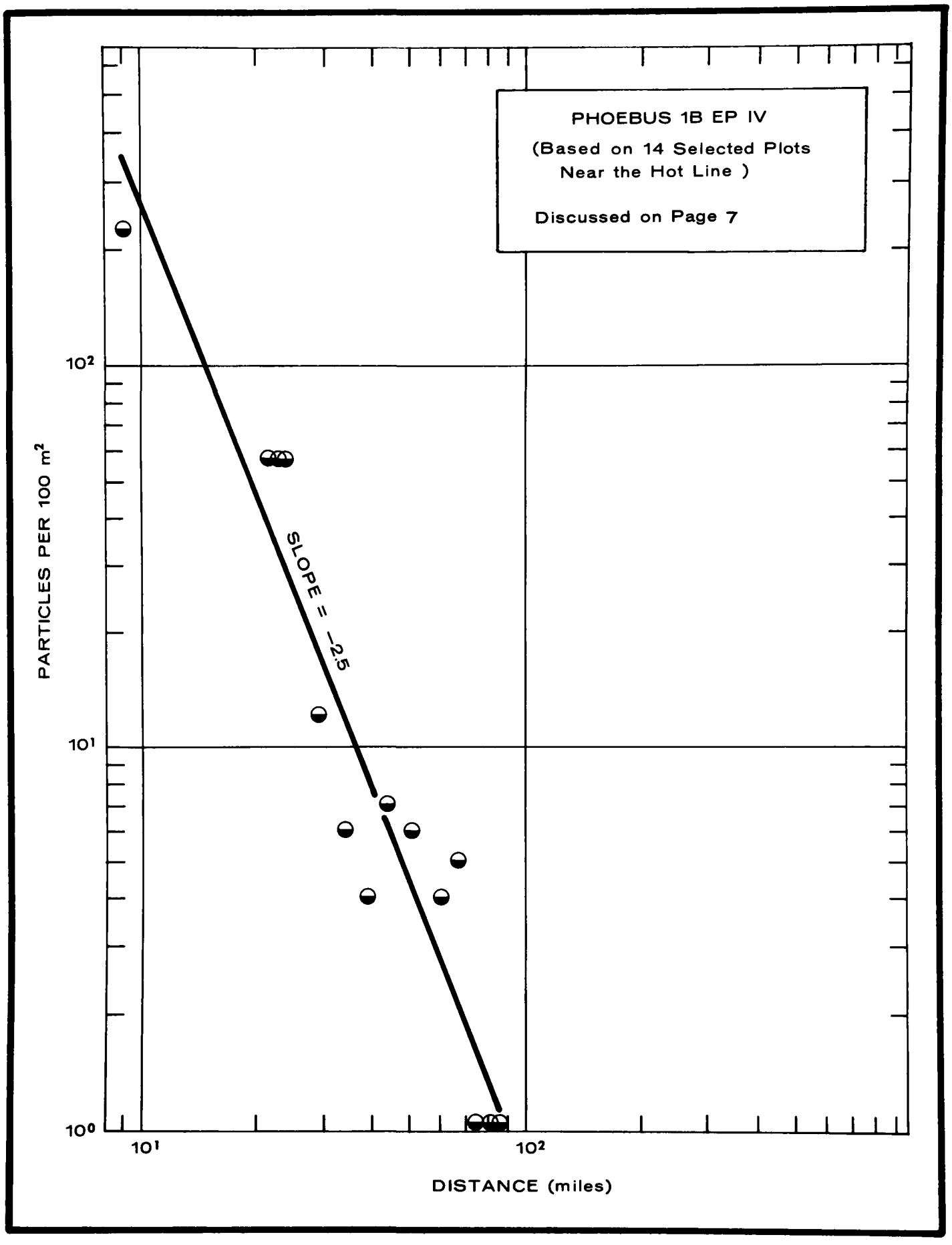

Figure 4. Number of particles per $100m^2$ versus distance (miles).

B. Particle Size Distribution

The size frequency distribution for 109 of the particles is presented as a bar graph in Figure 5 (numerical information in Appendix C). This information is also presented on log probability paper in Figure 6. It appears to give a reasonable fit to a log normal distribution with a geometric mean of about 12μ. The goodness of fit for this distribution was checked using a Chi(X) squared test. The calculated value of X^2 (95% confidence level) for 6 degrees of freedom was 8.9. Thus the data appear to be reasonably described by a log normal distribution. But, Figure 6 gives some indication of a platykurtic bimodal distribution.

The bimodal characteristic of the curve could be due to the presence of two or more distributions. That is, a basic distribution of fairly large particles and a distribution formed from fractured particles. If this is the case, it is felt that the fracturing took place in the environment.

It should be emphasized that correlations of the data may be biased by collection techniques (easier to find more active and thus possibly larger particles, collection was over a period of time, etc.); terrain features; and especially the fragile nature of the particles which made them very susceptible to fracturing during transport, collection, and analysis. Therefore it is possible that some of the particles were reduced in size, thus affecting any correlations. Care was taken in collection and handling (see Appendix A for methods) in an attempt to minimize these effects. If several particles were found in one sample, they were assumed to be parts of a fractured particle and excluded from the sizing analysis. Particles were collected with

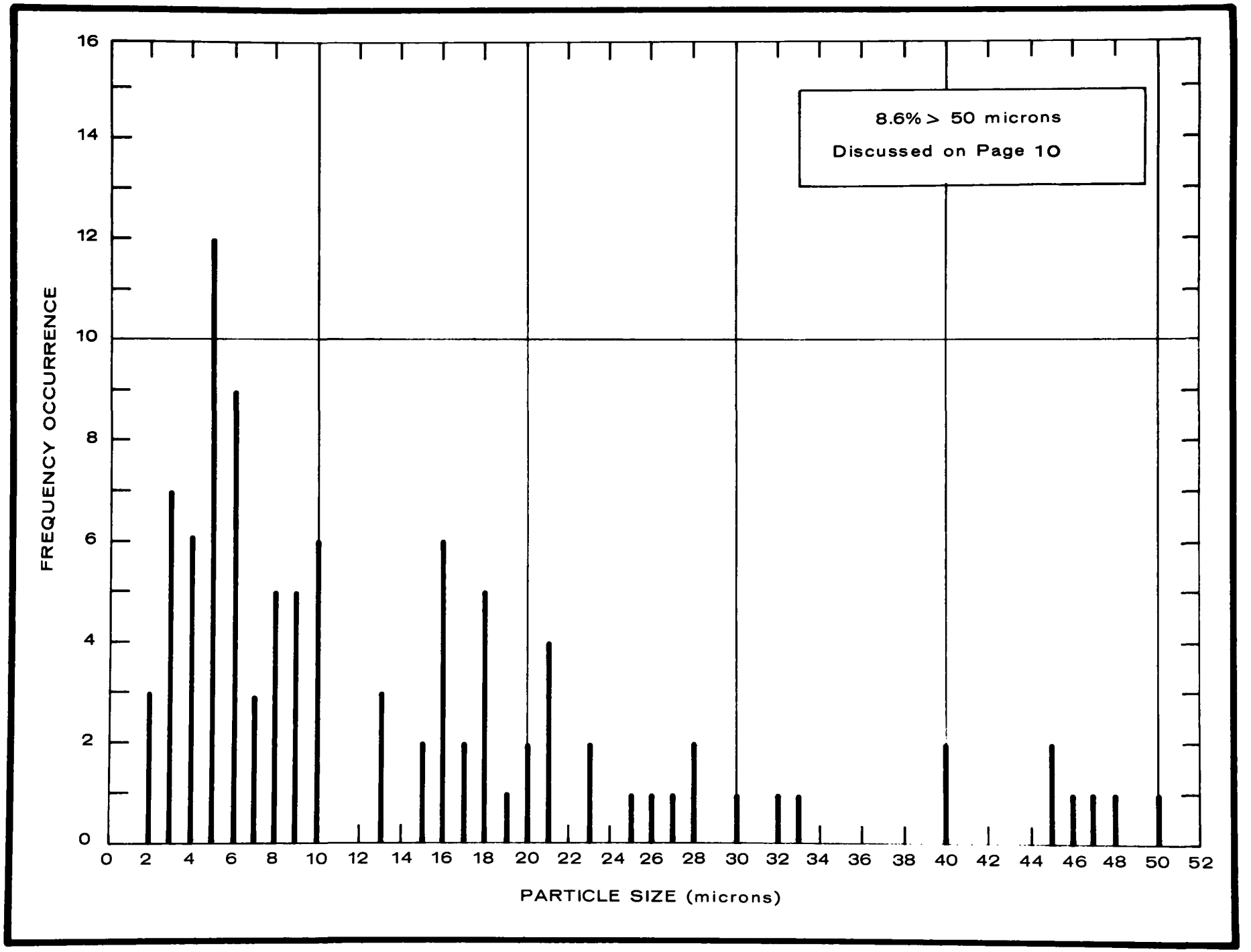

Figure 5. Particle size distribution, Phoebus 1B, EP-IV.

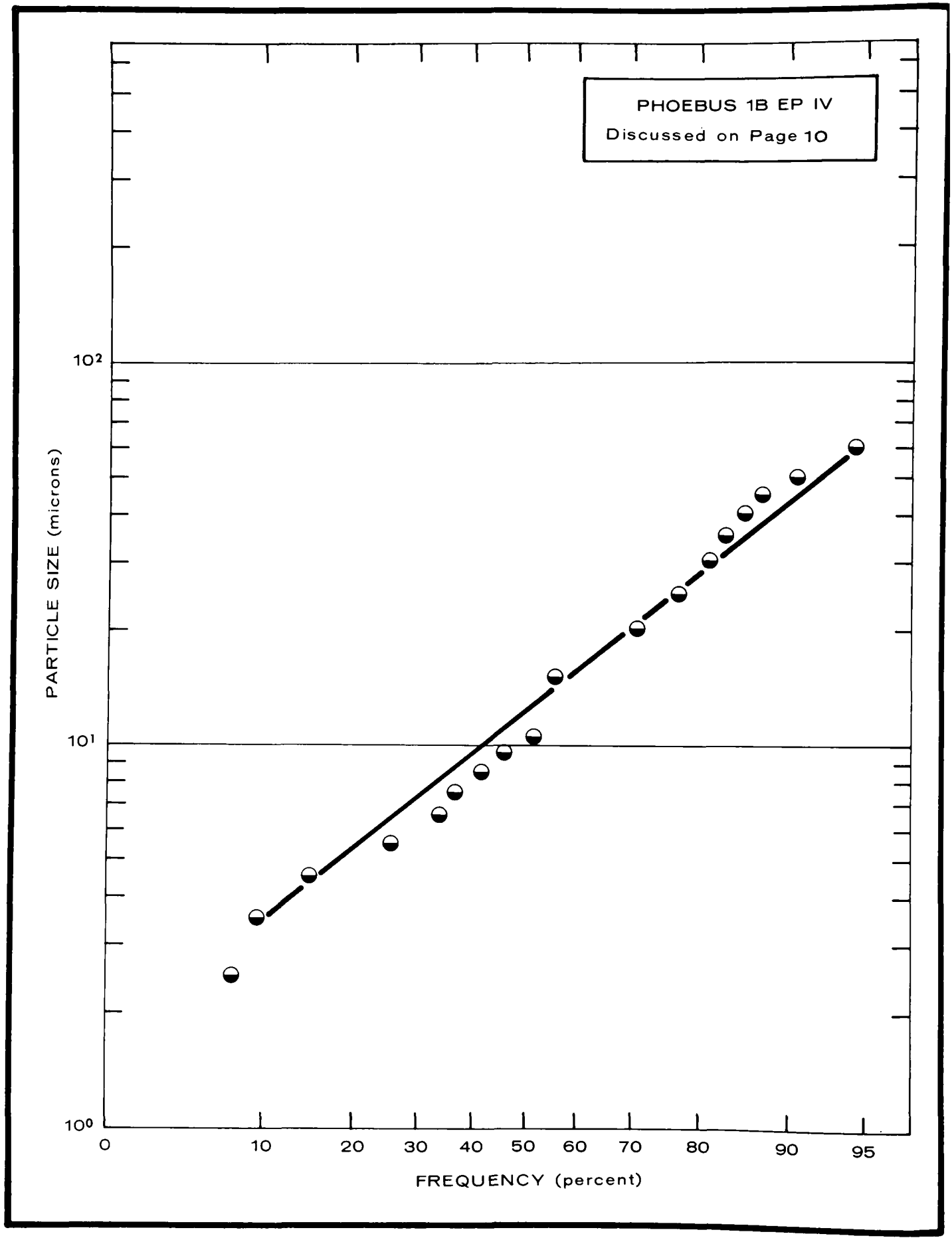

Figure 6. Log probability plot of 109 particles (size in microns).

a small amount of extraneous material and were handled with minimum contact in the laboratory, i.e., sieve techniques, etc., were not used.

It is worth emphasizing the following points:

1. A measured area was surveyed at each location. The survey was performed using a survey meter and traversing the area at one-foot intervals or less with the probe 6 inches or less from the ground. All indications of particles were noted. Of a total of 228 indications, about 180 particles were collected, 170 from monitored locations, 10 close-in where particles per unit area were not determined. Of those picked up, about 15 were used in developing techniques and are thus not reported in the sizing results. Of the remaining 165 particles, 56 were noted to be fractured during sizing and thus not used in analysis of size. It is felt that the reduction from the original 165 to 109 for sizing occurred in a random manner. Where all the particles located in the measured area were not picked up, the choice was for the "hotter" ones.

2. A cut-off in size (such as 10μ) was not used. That is, the actual size of all the particles was determined and reported.

3. Even if the 65 unsized particles were included and were considered to be above the geometric mean determined (12μ), the new geometric mean would be less than 20μ .

Table 1 gives a subdivision of the particles into several categories based on size and the distance at which the particles were collected.

Table 1. Particle size distribution(microns).

Category	No. of Particles in Category	Geometric Mean Diameter	Geometric Standard Deviation
All particles; at all distances	109	12.2	2.7
Particles $\geq$ 10μ at all distances	59	25.7	1.9
All particles; 10 miles	43	14.1	3.2
Particles $\geq$ 10μ < 10 miles	28	34.7	2.0
Particles $\geq$ 10μ 22-39 miles	13	18.7	1.8
All particles; 39-82 miles	17	11.4	---
Particles $\geq$ 10μ 39 miles	9	22	---

This breakdown into various size categories makes the results more analogous to those of other study groups (Pan American and Los Alamos Scientific Laboratory) which were based on analysis of particles greater than 10 - 15μ in diameter.[3,5]

C. Radioactive Constituents of Particles

Radiometric measurements were made on the particles to determine beta plus gamma activity (open window GM probe), alpha activity, and specific nuclides. The various radiometric measurements were not all made on the same particles, therefore, they are not necessarily related.

The beta plus gamma activity (GM probe) of 78 particles, along with their size (Feret diameter), is given in Appendix D. The distance downwind from the reactor, where they were found, is also indicated. This gross activity is only a relative number, and is reported as counts per minute detected by the GM probe at

time of count. The particles were all counted at about the same time (5 days after the event). The probe used has an approximate efficiency of about 10% for beta and less than 1% for gamma activity.

The alpha activity of eleven of the particles was measured on a NMC PC-3B counter and ranged from approximately 0.1 to 28 pCi/particle with an average of 8 pCi/particle.

The specific isotope analysis, based on gamma spectroscopy using a NaI(Tl) 4- by 4-inch crystal, for a number of particles is indicated in Table 2. Several particles were analyzed on a germanium (Li) detector. The germanium detector was not calibrated for quantitative analysis, thus the results were used only to help confirm the "NaI analysis."

The results in Table 2 are based on hand analysis by the Compton subtraction method, including the use of half-life verification, of the gamma spectra. In some of the samples reported in Table 2, more than one active particle was found. In these cases it is not known whether each particle was deposited separately or if they resulted from a single particle which was fractured by handling, but the latter appears more likely. In addition to the results in Table 2, the spectrum from the germanium detector indicated the presence of ^{231}Th (daughter product of ^{235}U). Short half-life isotopes noted to be present, but not quantitated were ^{91}Sr, ^{92}Sr, ^{92}Y and ^{135}I. Other fission products were undoubtedly present, but below the level of detectability. A least squares spectrum stripping computer program was used to analyze 14 of the particles. The results were similar to those in Table 2.

Table 2. Results of isotopic analysis of particles (pCi)*

Sample No.	20528 A	20528 B	20540 A	20540 B	20540 C	20542 A	20542 B	20543	20544	20545 A	20545 B	20545 C	20545 D	20546
^{95}Zr	ND	ND	2,500	15,000	110,000	1,100	2,000	12,000	18,000	3,500	5,300	18,000	23,000	34,000
^{97}Zr	ND	ND	240,000	ND	8,600,000	93,000	180,000	1,900,000	1,100,000	330,000	880,000	2,000,000	2,100,000	2,600,000
^{99}Mo	84,000	110,000	12,000	18,000	540,000	1,700	6,200	280,000	180,000	4,500	11,000	350,000	74,000	88,000
^{103}Ru	ND	ND	ND	5,500	ND	ND	1,100	2,000	ND	6,300	24,000	ND	55,000	ND
^{132}Te	8,200	7,500	3,000	8,000	71,000	1,100	3,000	2,600	12,000	4,400	3,700	8,700	16,000	16,000
^{133}I	ND	ND	ND	ND	ND	ND	ND	1,000	ND	ND	ND	ND	ND	ND
^{140}Ba	150,000	110,000	1,600	15,000	60,000	2,200	4,900	ND	24,000	7,400	20,000	81,000	60,000	64,000
^{141}Ce	78,000	67,000	2,800	210,000	100,000	1,500	3,300	300	240,000	5,500	17,000	27,000	53,000	9,600
^{143}Ce	ND	ND	8,500	7,100	280,000	3,400	8,200	2,500	61,000	14,000	11,000	82,000	92,000	110,000
^{147}Nd	ND	ND	3,000	26,000	210,000	1,800	5,000	ND	47,000	6,500	8,300	31,000	57,000	61,000
^{239}Np	ND	ND	51,000	470,000	1,300,000	10,000	35,000	ND	120,000	50,000	120,000	210,000	220,000	180,000
Total	320,000	290,000	320,000	770,000	11,000,000	120,000	250,000	2,200,000	1,800,000	430,000	1,100,000	2,800,000	2,800,000	3,200,000

Aircraft Sample - Indications of ^{91}Sr, ^{92}Sr, ^{92}Y, ^{97}Zr, ^{99}Mo, ^{132}I, ^{135}I, ^{140}Ba, ^{141}Ce, ^{143}Ce, ^{147}Nd, ^{239}Np

ple #20623-1- Indications of ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{140}Ba, ^{141}Ce, ^{143}Ce, ^{147}Nd, ^{239}Np

ple #20623-2- Indications of ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{140}Ba, ^{141}Ce, ^{143}Ce, ^{147}Nd, ^{239}Np

rapolated to 1515 PST, February 23, 1967

phabetic letters (A, B, etc.) indicate a subdivision of the sample

- Not detectable

Comparison of the results in Table 2 shows that the two portions of sample 20528 have similar inventories of fission products and are dissimilar to all other samples, indicating that they did result from fractionation of a single particle (note page 32, microprobe analysis indicated a possible difference.) The similarity in inventories of 20540-A and C indicate the same; however, 20540-B is unlike A and C, which would tend to show that this was not a part of the original particle or that the original particle may have been a combination of several types of material. This might be explainable by a combination of reactor material (UC_2 and/or graphite) and environmental dust. The gamma spectra from 20542-A and B indicate that these probably originated from one particle. Inspection of 20545 indicates that portions A, B and D are similar, but that portion C is different. Because of the low density of particles on the ground, it is difficult to conceive of more than one particle being picked up in a sample (taken from an area of about one square inch). Thus, the hypothesis of heterogeneous particle composition is presented.

Radioisotope analysis indicated a significant amount of fractionation (discussed in Appendix F).

D. Correlation of Particle Size, Gross Activity and Distance

A number of attempts were made to correlate the parameters of particle size, gross activity and distance.

Both regression analysis and correlation coefficients were used on various transforms of the data (linear, log linear, and log log). The results of the various correlation attempts are given in Appendix E. The correlations best justified by theory are given in Table 3. The premises used were:

1. Regression is generally used where one of the variables is

Table 3. Correlations of size, activity and distance

Data	Type Correlation	Number of Data Points	Slope of Line[4]	Correlation Coefficient
Size and Distance[3]	Log Log	6	-0.2	-0.64[1] (See Fig. 7)
Activity[2] and Distance	Log Log	6	-0.4	-0.71[1] (See Fig. 8)
Activity and Size	Log Log	68		-0.17[1]
	Linear	68		-0.46

1. Not significantly different from zero at the 95% confidence level.
2. Radioactivity as determined by a GM probe.
3. Distance from test cell at which particle was found.
4. Parameter = $\int (\text{Distance})^n$ where n is reported slope.

assumed to vary with the other, i.e., size with distance. Correlation coefficients are applicable for two variables which are both random, but are assumed to vary together, e.g., size and activity. The correlation coefficient may also be used to test the goodness of fit of a regression line. That is, the correlation coefficient squared is the fraction of variance in the data explained by the regression line.

2. The log log correlations are reported because the variables are assumed to be related by power functions, e.g., activity is a function of the surface area (r^2) and the volume (r^3) of the particle and the distance traveled is a function of terminal settling velocity(r^2). Thus a log log plot would be a straight line where graphs of other functions of the data would be expected to be curves.

3. The particle size and activity are described by a distribution rather than a precise value at each distance. Thus, the geometric mean of size and activity for various distance increments was used in the correlation attempts. The

geometric average size and activity were determined for the distance intervals: 7, 9, 22-26, 29-34, 39-44, and 51-82 miles. The separation into intervals was necessary to obtain a reasonable number of particles for each distance. The averages were then plotted at an average distance (weighted by number of particles at the various distances).

The geometric mean for particle size is plotted versus distance in Figure 7. The line is based on a regression analysis. The correlation coefficient for the indicated size with distance relation (mean size = distance to the minus 0.2 power) is -0.64. Due in part to the small number of points used in the correlation, this is not significantly different from zero at the 95% confidence level, but although the precise relation is in doubt, there appears to be a relationship between size and distance.

The correlation coefficient for the regression line (Figure 8) for activity of particle versus distance is -0.71. This indicates a reasonable fit of the data, but due to the limited number of points, it is not significantly different from zero at the 95% confidence level. However, it definitely suggests a decrease in activity with distance.

It is difficult to explain the correlations of activity ($\beta + \gamma$) with size. The activity should be a function of the area or volume of the particle (radius squared or cubed), so a log log plot should give the best straight line. However, the linear relationship is stronger than the log log relationship. Thus, they are both reported in Table 3. This may in part be due to the use of a GM probe for the radioactivity measurement. Beta and gamma efficiency for the probe, resolving time (about 10% or more), and energy dependence were not considered.

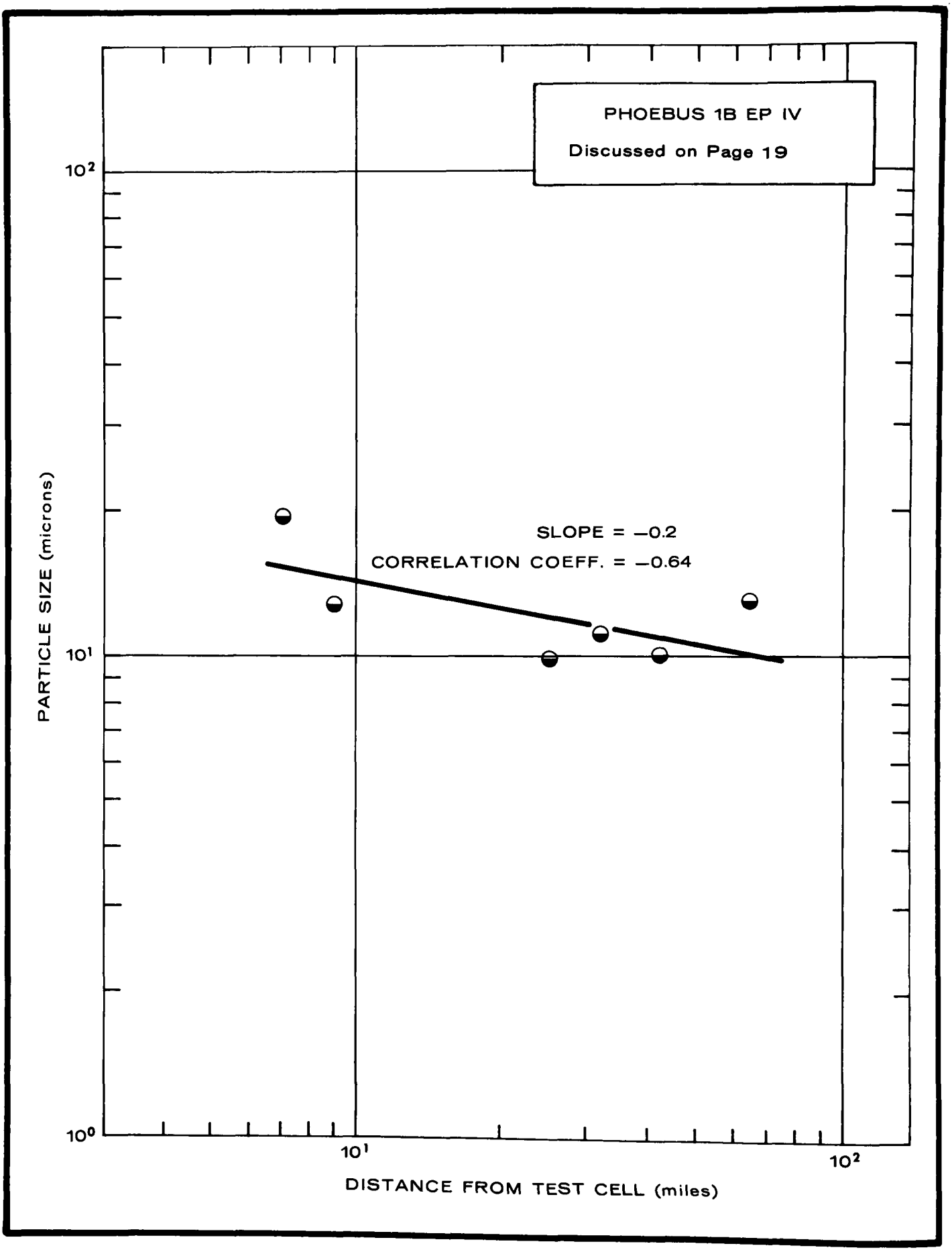

Figure 7. Particle size versus distance from test cell.

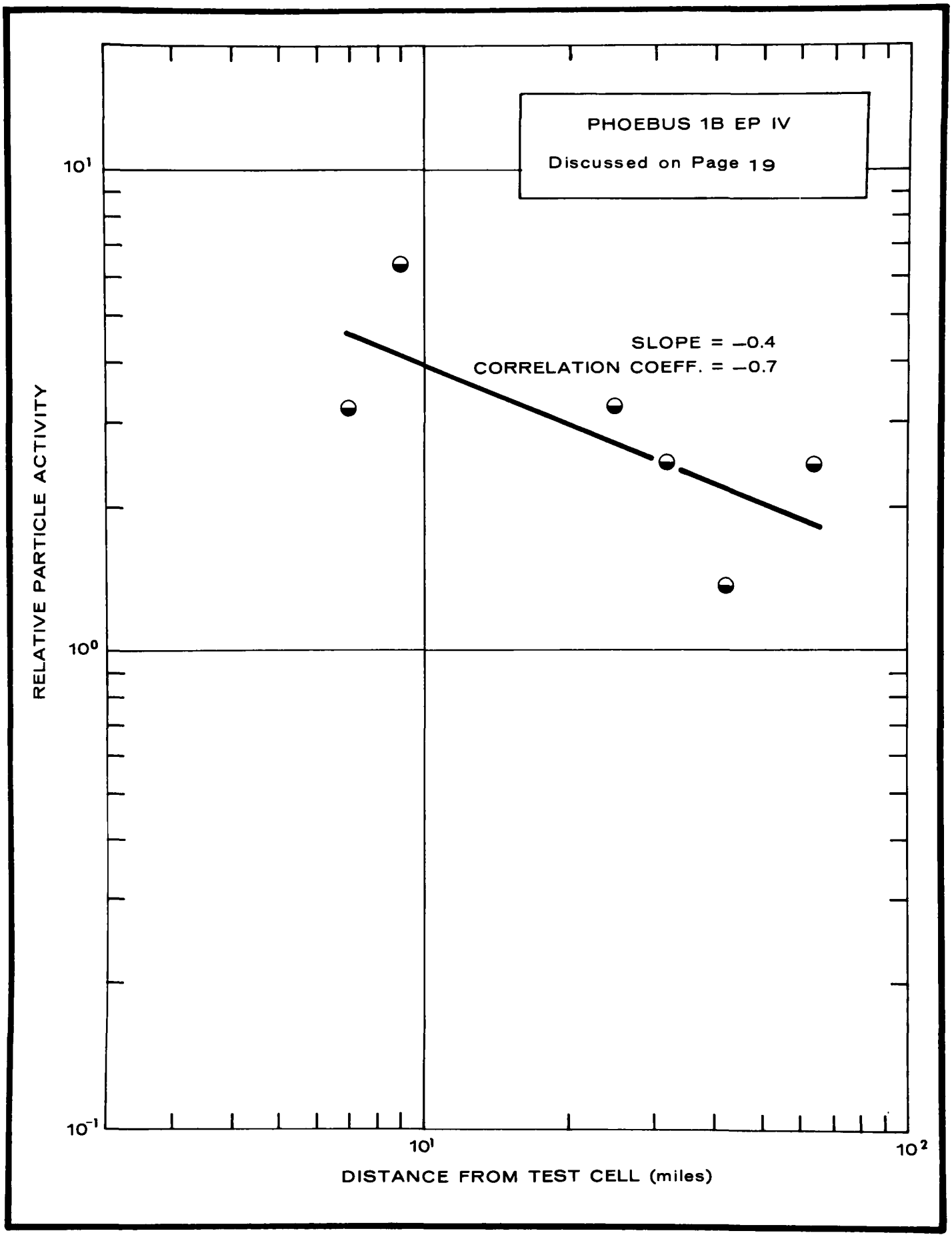

Figure 8. Survey meter readings of particles versus distance from test cell (beta + gamma with G.M. instrument).

Figure 9 gives an indication of the distance downwind various size particles can be transported. The figure is based on:

1. Stokes' Law settling velocity.
2. Release height of one mile, i.e., difference between release height and ground level at point of deposition.
3. Wind or transport speed of 15 mph.

Atmospheric diffusion or turbulence was not considered, but would primarily cause the deposition to be spread (distance plus and minus) around the indicated transport distance.

E. Elemental and Chemical Composition

A number of samples were analyzed with an electron microprobe to determine their elemental and chemical composition. All the particles were not analyzed, but those selected should be generally representative of most of the particles collected. These results are given in Table 4 and the general sample description is given in Table 5.

Three particles were analyzed for quantitative results. Quantitative analyses of one bead yielded the following results:

Element	Percent by Weight
Uranium	91.0
Carbon	8.4
Oxygen	0.7

The other results were similar, i.e., $\pm$ 10% of the given value. These elemental fractions indicate a stoichiometric composition of about 90% of uranium as UC_2 and about 10% or less as UO_2.

One particle was analyzed by X-ray diffraction subsequent to microprobe analysis (U, C, and O). The particle was removed

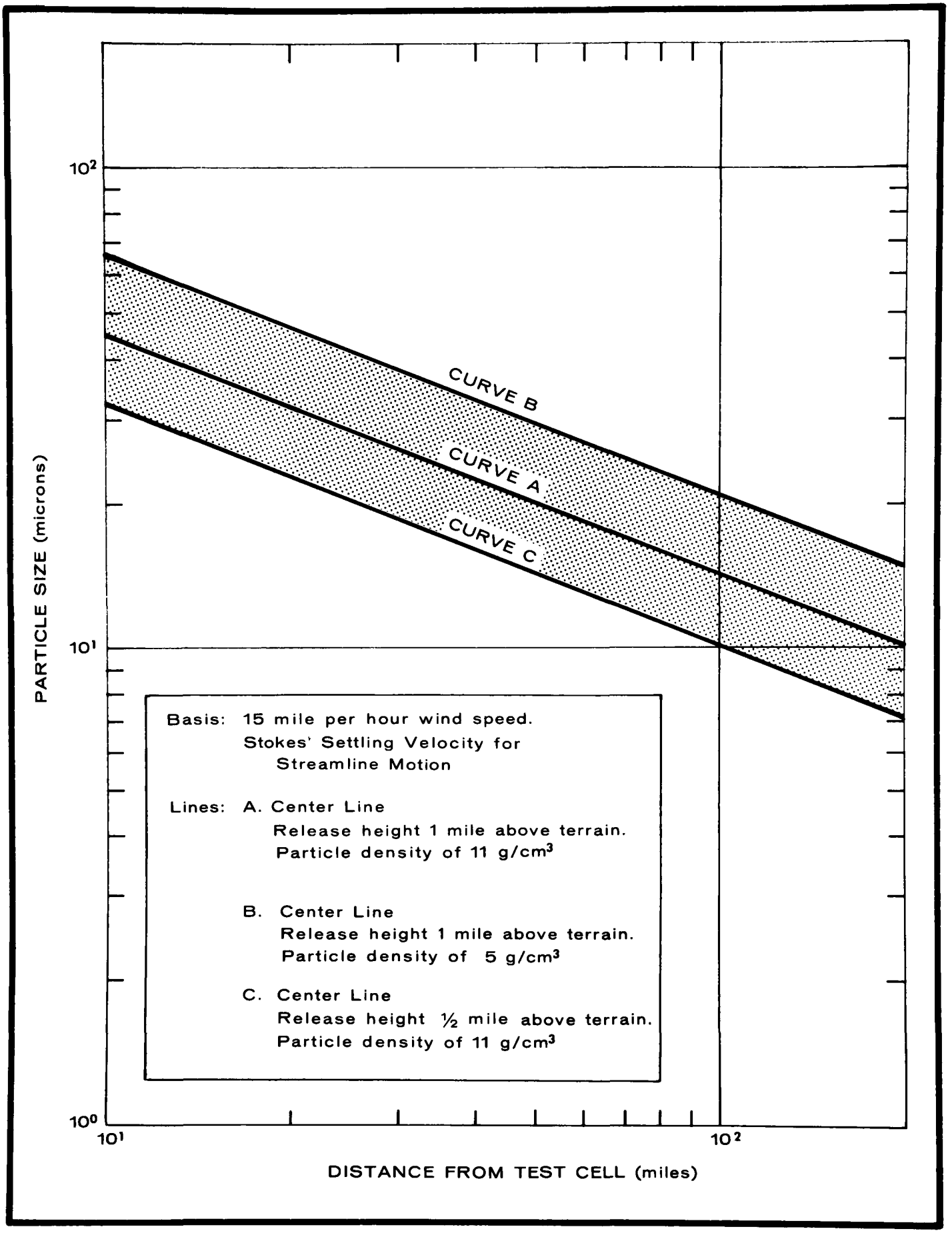

Figure 9. Estimated downwind transport distance for various size particles. Discussed on page 22.

Table 4. Results of electron microprobe analysis

Sample No. *	Microprobe Results	Comments
20528-A	No detectable U, Zr, Mo, Nb	Radionuclide analysis indicated A & B were similar.
20528-B	4 hot spots on this sample (1) Contained U and C No detectable Zr, Mo, Nb or O (2) Contained U and C Indications of traces of Nb and O No detectable Zr or Mo (3) Contained U and C No detectable Zr, Mo, or Nb (4) Contained U and C No detectable Zr, Mo, or Nb	
20540-A	No detectable U, Zr, Mo, or Nb	Radionuclide analysis noted dissimilarity.
20540-B	Silicate matrix, disintegrated under electron beam.	
20542-A	Particle lost in transfer	Radionuclide analysis similar for A & B.
20542-B	No detectable U, Zr, Mo, or Nb	
20543	Contained U and C Indications of trace of Nb	
20544	Contained U and C on silica matrix	
20545-A	Sample could not be analyzed because the activity was on a large grain of sand.	Radionuclide analysis noted A & B to be similar, but C to be different from A & B.

Table 4. Results of electron microprobe analysis (continued)

Sample No.*	Microprobe Results	Comments
20545-B	Contained U, C, and O No detectable Zr, Mo, or Nb	
20545-C	No detectable U, Zr, Mo, or NB	
_____	Contained U, C and O	
_____	Contained U, C and O	
_____	Contained U, C and O	

*See Table 5 for collection location, size and gross activity.

Table 5. Location, size, and total isotopic activity of particles

Sample No.	Azimuth	Distance	Size (μ)	Activity*(pCi)	Comments
20528	357°	6.0 mi	45 x 32**	610,000	
20540	3°	21.5 mi	16 2 x 3	12,000,000	
20542	16°	25.0 mi	20 x 24 19 x 24	370,000	
20543	16°	25.0 mi	9 x 14	2,200,000	
20544	12°	23.5 mi		1,800,000	Many <6μ
20545	13°	23.0 mi		7,100,000	One 25 x 20μ One on a grain of sand Several <10μ
20546	13°	23.0 mi		3,200,000	One on grain of sand
20623	9°	72.0 mi	15		>25 active pieces

*Total identified activity to two significant figures. See Table 2 for specific isotopic activity.

** The two figures indicate the dimensions of the particle as seen in a plane view. A single figure indicates uniform dimensions.

from its mounting with a micro-manipulator causing it to fracture. A fragment of the original particle of approximately eight microns in diameter was mounted on a pyrex glass fiber and placed in a Debye-Scherrer powder camera. Forty-four hours of exposure produced very faint but readable diffraction lines. The relative line intensities could not be measured; however, line positions were determined. The diffraction pattern of the sample was directly compared with those from a reactor bead and spectrographic grade carbon. The diffraction analysis indicated only uranium carbide and free carbon to be present. Uranium oxide would not be detected in this particular analysis because of the low concentration of uranium oxide in the sample and the small sample size.

The free carbon that was indicated in the diffraction analysis is probably due to carbon deposition on the sample during the microprobe analysis. Correlation of these analyses indicated that the particle was composed of UC_2 and a form of uranium oxide. The uranium oxide was probably UO_2.

F. Particle Density

Density determinations were made on 8 particles (see table 6). The arithmetic mean density (also geometric mean) was 11 gm/cm^3. The determination was based on the equivalent diameter (based on two dimensional projection of a spherical particle of equal cross sectional area) and Stokes' settling velocity in hexane. The method is further described in Appendix A.

Table 6. Density of particles

Equivalent Diameter (μ)	Density (gm/cc)
28	9.59
25	13.20
20	9.79
18	11.56
22	11.53
20	12.88
12	11.43
20	8.11

G. Biological Clearance Rates

A study was performed to estimate the clearance rate in rats for particles collected from the reactor run. The objectives were as follows:

Primary - Establish methodology for future studies.

Secondary - Obtain an indication of clearance times and routes and the solubility of the radioactivity associated with the particles.

Due to the limited number of radioactive particulates available for this study, only two rats were injected; one intratracheally and the other intraesophageally. Particulates with a CMD* of less than 10μ were suspended in an aqueous solution and 0.15 ml of the suspension was injected into the trachea (lungs) of one rat and into the esophagus (stomach) of the other rat.

Clearance from the animal injected by the intraesophageal route (stomach) was rapid. Thirty-five percent of the original body burden cleared in the first 24-hour period and an additional

*Count mean diameter.

fifty-eight percent of the original burden cleared in the second 24-hour period. The amount remaining after the fifth day was insignificant.

Clearance was much slower and relatively constant for the animal injected by the intratracheal route. A clearance half-time of approximately 20 days was calculated for this animal with over 10 percent of the original body burden still remaining 66 days following injection of the radioactive material. This animal is still under observation and will be permitted to expire naturally. The results indicate that these particulates were relatively insoluble since essentially all the radioactivity lost in both rats was accounted for in the feces.

III. SUMMARY AND CONCLUSIONS

The objectives of the program were intentionally limited so that a reasonable degree of success could be accomplished with available personnel and financial resources. Thus, although there is reasonable doubt remaining concerning some of particle parameters and many questions to be answered or investigated more fully in the future, it is felt the objectives outlined in Section 1 - B were met.

Post-run inspections of the reactor cores have shown varying types of corrosion both with type of reactor, i.e., Phoebus or NRX (KIWI), and with reactors within a type, i.e., NRX-A4 or A5. Thus, knowledge of the parameters of particles in the effluent from a given reactor test may not be directly applicable to other reactor tests.

It should be reiterated that this report, and therefore the conclusions drawn from it, are based on a limited amount of information. Thus, the following observations should be used with discretion and should be validated with future reactor tests.

Observations

A. A least squares fit of the data indicated that the particle concentration per unit area on the ground decreased with distance to about the 2.5 power (Figure 4, page 13). In addition to indicating the change with distance, an estimate of particle concentration on the hotline is indicated. There was very little wind shear for the Phoebus 1B, EP-IV, and a moderate wind speed. Thus the figure should give a reasonable indication for reactor runs of this type in the future (same power, power integral, and fuel type).

B. The average particle size and activity decreased with distance. The data are insufficient to denote a definite relationship, but indicate the decrease with distance is less than distance to the first power (see Figures 7 and 8).

C. The overall size distribution of the particles collected from 6 to 82 miles (based on sizing 109 particles) gave a reasonable fit to a log normal distribution. This distribution had a geometric mean of 12μ and geometric standard deviation of 2.7. A breakdown of the distribution into those particles equal to or greater than 10μ collected at less than 10 miles gave a geometric mean of 35μ with a geometric standard deviation of 2.

D. Based on the average density for eight particles (11 g/cc - which may not be representative of all particles) and the considerations of aerodynamic diameter, greater than 90% of the particles detected were above an aerodynamic diameter of 10μ which is usually considered the upper size level for penetration into the respiratory system. This statement holds for all downwind distances where particles were detected. Admittedly insufficient particles were detected to determine more than an indication of particle size with distance, especially beyond 50 miles.

E. Based on eleven randomly selected particles analyzed for elemental composition it is concluded that the particles collected were primarily uranium and carbon. The density results also indicate the particles are largely uranium. It is suggested, based on other reports [3, 5], that the particles might fall into two types of distributions, i.e., large particles around 100μ may be UC_2 with associated graphite coat and possibly natural environmental material, and the small particles primarily UC_2. Some of the

particles were noted to be small UC_2 particles associated with silicate material (material from the natural environment).

F. There appears to be a "weathering" effect in detecting and collecting the particles. The particles collected beyond 40 miles from the test cell were primarily collected on the 26th or 2 to 3 days after the near-in collections. From Figure 7 it can be noted that particle size did not decrease as much with distance as might be expected (based on Stokes' law). This could be explained if the small particles were no longer detectable at the time of survey. Also (Figure 4) the observed particle concentration on the ground fell off slightly faster than might be expected. This would follow the previous reasoning in that the small particles were not detected, at least not with the same efficiency as the larger particles, at the later survey times.

G. As can be noted from Figures 1 and 2, particles were deposited in the off-site area (out to 82 miles from the test cell). But, the deposition density was of the magnitude of 1 per $100m^2$ and was in areas of very low population density. The only people in an area of known deposition were at Diablo (3 people). There was no known case where the general population came in contact with this effluent.

H. Due to the variation in distance to the off-site boundary and variation in population density in the off-site area, it is difficult to assess the potential hazards from these particles. This would be true even if the hazard resulting from human interaction with the particles was known; which is not the case. With the unknowns, it becomes impossible to make a definitive statement at this time concerning the hazard in the off-site area. Among

the statements that can be made concerning the potential hazard of these particles are:

1. Particles were transported to 82 miles.
2. The ground density of the particles and thus the probability of human interaction (disregarding population densities) decreases with distance to about the 2.5 power.
3. The particles contain microcurie quantities of fission products.
4. The biological half-life in the lung may be very long.

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APPENDICES

APPENDIX A

METHODS OF COLLECTION AND ANALYSIS

A. Particle Collection

Particles were located by monitoring teams surveying either an area of 300 square feet or 100 square meters (except as noted in Table 1) with an Eberline E-500B Geiger counter with the shield open (beta plus gamma) and/or a Precision 111 "Scintillator." The general area to be surveyed was determined from the results of aerial cloud tracking, gamma exposure measurements during cloud passage, and gross gamma counting of vegetation results from two on-site vegetation arcs and an arc off-site on Highway 25 (Figure 1). Collection took place over a six-day period. The surveys were performed along a downwind sector from about 350° to 30° and between 9 and 115 miles. Fifty-three areas were surveyed (see Figure 1). Particles were collected with as little soil as possible. It is felt that the collection techniques were reasonably good, but by no means absolute. Out of the 228 indications for particles, about 170 particles were picked up (no attempt was made to pick up all of them). Where all the particles were not picked up, the "hotter" ones were selected. Various numbers of these particles were then used in the different analyses.

One particle was collected by impaction on sticky material placed on the leading wing edge of the PHS sampling aircraft. The location of collection of the particle could not be defined because of the method of collection.

The particles were isolated from the sampled material, using microscopy, autoradiography, and collimated β/λ detectors. They were fixed on glass slides with polyvinyl chloride (PVC) or saran film. After fixing on the slide, particles were autoradiographed (except for 5 of them) to determine which was the actual particle. In some samples more than one particle was found. The particles were extremely fragile and so in these cases it is not known whether samples were fractured through handling or were deposited as several particles. When more than one particle was noted in the sample, it was excluded from the sizing results. Less than 1/3 of the particles sized were excluded because of fracturing.

B. Particle Sizing

The diameter was determined using the Feret diameter measurement. A qualitative determination to establish that the third dimension was of the same regularity as the other two was made by focusing alternately on the topmost portion of the particle and on its lowest maximum dimension. The particle dimensions were irregular, but were more spherical than "needle like."

C. Constituent Radioactivity of the Particles

Three types of radiometric measurements were performed. They are as follows:

1. Gross $\beta + \lambda$ - Particles were counted approximately 5 days after collection using a RM-3A GM (open window) detector. The detector was not calibrated, but probably has about 10% and less than 1% overall efficiency respectively for beta and gamma. Results are given in CPM at time of count.

2. Alpha Activity - Alpha activity was measured on an NMC PC-3B counter. Eleven particles were analyzed. Activity is reported in pCi/particle as of time of count. No correction was made for self-absorption. Thus, in reality, the activity reported is probably surface activity rather than total particle activity.

3. Specific Isotope Analysis - Quantitative specific isotope analysis was performed by gamma spectroscopy. Spectra were obtained by using a 4- by 4-inch NaI(Tl) crystal, a 4-inch photo-multiplier, and 200 channels of a 400-channel pulse height analyzer. Analysis was performed from 0-2 MeV at 10 keV per channel. Spectra from a lithium-drifted germanium diode detector in connection with a 1024 channel analyzer were used in analysis but were not quantitated due to lack of qualitative calibration of the instrument.

These results were analyzed by two techniques - Compton Subtraction and weighted least squares computer program.

COMPTON SUBTRACTION

Spectra from several recounts were used with this method, thus utilizing half-life determinations for the various gamma peaks to confirm the analysis.

LEAST SQUARES PROGRAM

A computer program based on a weighted least squares analysis was used. The weighting function was based on the counting error in each channel. The number of isotopes that can be used in the computer program is variable, but it is limited to isotopes where standards are available. Standards for the following isotopes were not available for

the computer program: ^{91}Sr, ^{92}Sr, ^{92}Y, ^{135}I, $^{97}ZrNb$, and ^{143}Ce. Gamma spectra taken about 2 weeks after the reactor run were used for the analysis. The previously mentioned isotopes would have decayed to one-thousandth or less of their original activity by this time.

D. Correlation Coefficients

Linear correlation coefficients were calculated for both the untransformed data and logarithmic transforms (transforms are an attempt to make the data fit a straight line - the correlation coefficient is a measure of the fit to a straight line). Particle size and activity are distributed at each distance. Thus, a point by point correlation, e.g., parameter of particle versus distance at which it was found is misleading. Thus, these correlations are based on the mean of the parameter at each distance. The correlation for particle size and activity was done on a point by point basis.

E. Elemental Chemical Composition

Chemical composition was determined by electron microprobe (Norelco Instrument) analysis. In this process, the characteristic X-rays from electron excitation are passed through an X-ray energy dispersing crystal to a detector. Normally, an emission angle from 30 to 70 degrees is measured with a rotation of 2 degrees per minute. The signal and associated angle are recorded on a strip chart recorder. Due to the large number of particles to be analyzed, in most cases, just the spectral peaks for the various elements were measured. The particles were analyzed for U, Zr, Mo, Nb, C and O. The area of the electron beam used on these samples was approximately one micron. The minimum detectable quantity of

U, Zr, Mo and Nb was approximately 10^{-12} grams regardless of beam size, providing that the beam remained entirely on the particle. The minimum detectable limit is significantly greater for elements of low atomic weight such as O and C.

The small beam size was used on all particles to ensure that the beam remained on the particle being examined. Some larger particles were also examined with a larger beam, approximating the particle size, to determine if detectable amounts of the elements sought were spread over the entire surface.

The most conclusive semi-quantitative analysis of uranium and carbon was accomplished by comparing the samples with a crushed reactor bead mounted by the same procedure that was used on the samples. The relative uranium-carbon count rate ratios were used for comparison with the data from the sample. The oxygen content was quantitated by comparing the uncorrected, net count rate of the oxygen in the sample with the oxygen count rate from ruby (Al_2O_3). The usual mathematical corrections used for quantitative probe analysis could not be used because mass absorption coefficients and fluorescence corrections are not available for carbon and oxygen.

F. Density of Particles

The density of eight particles (12 - 28μ in size) was estimated by use of Stokes' Law and the settling velocity through hexane (viscosity = 0.326 cp). The equivalent diameter was taken to be that of the two dimensional projection of a spherical particle of equal cross sectional area. The determination of equivalent size was done on a Zeiss Particle Size Analyzer. A measure of the accuracy and reproducibility of this procedure was made by using 27.4μ silver spheres in the same experimental arrangement.

Ten of these spheres were allowed to fall 15.24 cm in hexane and their terminal velocities were recorded. The average settling velocity was 1.28 cm/sec with a range of from 1.21 - 1.34 cm/sec. Based on the average measurement, a density of 10.8 gm/cc was calculated for the silver spheres (pure silver has a density of 10.5). Inaccuracies caused by irregularities of particle dimensions would tend to give a low rather than a high density using the settling velocity technique.

G. Biological Clearing Rate of Internally Deposited Particles for Rats

A preliminary biological experiment was performed with rats. Particles with a CMD* of less than 10μ were suspended in an aqueous solution and injected in two rats; one by the intratracheal route and the other intraesophageally. The main objective of the study was to establish methodology, with secondary objectives of obtaining gross indications of clearance times and routes and particulate solubility in biological fluids.

Following the injection, each animal was placed in a restrainer between the 9-inch opposed sodium iodide crystals and whole-body counted. This procedure was repeated daily on each rat until the detectable activity dropped to less than 50 percent of the original body burden. At this point, the counting frequency was changed to a weekly schedule and maintained at this rate until significant counts were no longer obtained. In addition, urine and fecal eliminations were collected from each animal and counted at the same counting frequency as that utilized for the whole-body count.

*Count mean diameter.

APPENDIX B

PARTICLE SURVEY RESULTS

Date of Collection February 1967	Distance from Test Cell "C" in Miles	Azimuth from Test Cell "C" in True Degrees	Number* Particles per 300 ft^2	Number Particles per 100 m^2
23	9	20		225****
	11	30		0
	14	20		15
	17	30		0
	19	20		3
	30	30		0
	50	20		0
24	19	355	15**	54
	21.5	3	16**	57
	23	13	16	57
	23.5	12	16	57
	24	27	0	
	25	16	4	14
	26	7		5
	26	10		12
	29	8		12
	34	7		6
25	65	18	0***	
	66	21	0	
	69	10	0***	
	72	9	1**, ***	4

APPENDIX B

PARTICLE SURVEY RESULTS (continued)

Date of Collection February 1967	Distance from Test Cell "C" in Miles	Azimuth from Test Cell "C" in True Degrees	Number* Particles per 300 ft^2	Number Particles per 100 m^2
26	39	5		4
	44	4		7
	51	5		6
	60	7		4
	67	4		5
	74	0		1
	82	356		1
	82	356		1
27 & 28	92	6	0	
	92	7	0	
	93	9	0	
	93.5	10	0	
	95	11	0	
	87	7	0	
	76.5	8	0	
	74	8	0	
	72	9	0	
	70	10	0	
	112.5	3	0	
	113	2	0	
	113.5	1	0	
	114	0	0	
	114.5	359	0	

APPENDIX B

PARTICLE SURVEY RESULTS (continued)

Date of Collection February 1967	Distance from Test Cell "C" in Miles	Azimuth from Test Cell "C" in True Degrees	Number* Particles per 300 ft^2	Number Particles per 100 m^2
27 & 28 (cont'd)	115.5	358	0	
	115.5	357	0	
	93.5	356	0	
	93.5	357	0	
	93.5	359	0	
	94	0	0	
	94	1	0	
	95	2	0	
	91	5	0	

*The area surveyed was 300 ft^2 for indicated results in this column and 100 m^2 for other results. Results were then normalized to 100 m^2 and included in the next column.

**$\beta + \gamma$ contact readings for E-500 B GM survey instrument at time of collection.

Particles at 19 miles	1 particle	> 200 mR/hr
	2 particles	100 - 200 mR/hr
	3 particles	50 - 100 mR/hr
	9 particles	< 50 mR/hr
Particles at 21.5 miles	4 particles	> 100 - 200 mR/hr
	1 particle	50 - 100 mR/hr
	11 particles	< 50 mR/hr
Particle at 72 miles	1 particle	10 - 50 mR/hr

***Area surveyed was approximate.

****An area of about 35m^2 was surveyed and 78 particles detected and picked up.

APPENDIX C

FREQUENCY OF PARTICLE SIZE

Size (μ)	Frequency	% Frequency	Cumulative % <Stated Size
2	3	2.8	2.75
3	7	6.4	9.18
4	6	5.5	14.7
5	12	11.0	25.6
6	9	8.3	34.0
7	3	2.8	36.7
8	5	4.6	41.3
9	5	4.6	45.9
10	6	5.4	51.3
13	3	2.8	54.1
15	2	1.8	56.0
16	6	5.5	61.5
17	2	1.8	63.3
18	5	4.6	68.0
19	1	0.9	68.9
20	2	1.8	70.6
21	4	3.7	74.4
23	2	1.8	76.1
25	1	0.9	77.1
26	1	0.9	78.0
27	1	0.9	79.0
28	2	1.8	80.8
30	1	0.9	81.7

APPENDIX C

FREQUENCY OF PARTICLE SIZE (continued)

Size (μ)	Frequency	% Frequency	Cumulative % < Stated Size
32	1	0.9	82.6
33	1	0.9	83.5
40	2	1.8	85.4
45	2	1.8	87.2
46	1	0.9	88.1
47	1	0.9	89.0
48	1	0.9	90.0
50	1	0.9	90.8
51	1	0.9	91.9
52	1	0.9	92.6
58	1	0.9	93.6
60	1	0.9	94.5
78	2	1.8	96.4
81	1	0.9	97.3
83	1	0.9	97.2
115	1	0.9	99.1
144	1	0.9	100.0

APPENDIX D

SURVEY METER READINGS OF PARTICLES

Distance (Miles)	Size (μ)	β + γ Activity* (cpm)	Distance (Miles)	Size (μ)	β + γ Activity* (cpm)
6**	40	NM	9	5	3,800
7**	6	14,000	9	78	19,000
7	19	25,000	9	6	11,500
7	13	3,100	9	9	5,000
7	40	600	9	10	2,000
7	32	3,100	9	3	13,000
7	7	1,400	9	78	16,000
7	26	1,400	9	28	6,000
7	30	3,800	9	144	26,000
7	18	2,500	9	5	30,000
9	23	2,900	9	4	1,400
9	7	9,000	9	3	2,100
9	33	20,000	22.5	16	NM
9	20	700	25	13	NM
9	3	5,500	26	6	2,500
9	9	1,900	26	5	3,400
9	3	8,500	26	18	1,800
9	60	14,000	26	3	7,000
9	7	3,200	26	47	9,500
9	45	10,000	26	10	4,800
9	5	2,700	26	9	2,900
9	83	24,000	26	10	2,200

APPENDIX D

SURVEY METER READINGS OF PARTICLES (continued)

Distance (Miles)	Size (μ)	β + γ Activity* (cpm)	Distance (Miles)	Size (μ)	β + γ Activity* (cpm)
9	2	28,000	26	6	1,200
9	81	21,000	29	6	7,500
9	16	8,000	29	13	1,350
9	16	5,500	29	50	10,000
9	52	8,500	29	21	7,000
9	2	6,000	34	17	6,500
9	5	1,100	34	8	1,800
9	4	7,500	34	10	1,500
9	58	2,100	34	18	1,100
34	3	650	44	10	170
34	5	650	51	8	8,000
39	3	2,500	51	18	7,500
39	5	1,100	51	21	350
39	6	3,700	60	15	2,100
39	48	4,400	67	51	350
44	9	350	67	21	3,000
44	45	2,800	74	4	4,500
44	10	430	82	5	4,700
44	6	4,600			

NM - Not Measured

*As measured by an RM-3A GM monitor.

**The particles from 6 and 7 miles were not located by surveying a defined area. They are not reported in Table 1 and Figure 1.

APPENDIX E

PARTICLE SIZE, ACTIVITY AND DISTANCE CORRELATION COEFFICIENTS

Relationship	Activity (A) Size (S)	Coefficient (r) (68 Data Points)	Size (S)** Distance (D)	Coefficient (r) (6 Data Points)	Activity (A)** Distance (D)	Coefficient (6 Data Points)
Linear	A vs S	0.46	S vs D	-0.44*	A vs D	-0.64*
Exponential	A vs log S	0.25*	S vs log D	-0.66*	A vs log D	-0.69*
Exponential	S vs log A	0.34	log S vs D	-0.41*	log A vs D	-0.65*
Power Function	log S vs log A	0.17*	log S vs log D	-0.64*	log A vs log D	-0.71*

*Correlation coefficient not significantly different from zero at the 95% confidence level.

**Correlation performed on the average size or activity for a distance interval, rather than each particle.

APPENDIX F

ISOTOPIC FRACTIONATION

Relative abundances of ^{235}U fission products at one hour after fission relative to ^{99}Mo, were obtained from Bolles and Ballou.[4] The ratios of fission products identified for each sample were then related to the amount of ^{99}Mo found. These ratios, based on data from Table 4 in the report are presented in Table F-1. Each ratio calculated for each sample was divided by the corresponding ratio calculated from Bolles and Ballou to determine an "enrichment factor" for the isotope relative to the amount expected on the basis of ^{99}Mo. These enrichment factors are presented in Table F-2.

From Table F-2 it can be seen that the zirconiums, ^{103}Ru, ^{140}Ba, ^{141}Ce, and ^{147}Nd are enriched relative to ^{99}Mo, while ^{132}Te and ^{143}Ce are depleted. This might also be interpreted to mean that ^{99}Mo is depleted relative to most of the other isotopes quantitated. It is also observed that the enrichment factors for ^{95}Zr and ^{97}Zr are generally the same within each sample, although they may vary between samples. In general, the enrichment factors for the ceriums differ by about an order of magnitude within each sample. The activity values are plotted in Figures F-1 and F-2 for the ceriums and zirconiums. Linear correlation coefficients were determined to be 0.54 for the $^{141}Ce/^{143}Ce$ ratios and 0.97 for the $^{95}Zr/^{97}Zr$ ratios. Similar correlations may be determined for any other pair of isotopes.

Table F-1. Ratios* of fission product activities based on ^{99}Mo

Isotope	Bolles & Ballou**	20528		20540			20542		20543	20544	20545				20546
		A	B	A	B	C	A	B			A	B	C	D	
^{95}Zr	0.046	ND	ND	0.21	0.85	0.21	0.68	0.32	0.04	0.1	0.78	0.51	0.05	0.31	0.42
^{97}Zr	4.05	ND	ND	21	ND	16	55	29.0	6.6	6.2	74	84	5.6	28	30
^{99}Mo	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
^{103}Ru	0.041	ND	ND	ND	0.31	ND	ND	0.17	0.007	ND	1.4	2.3	ND	0.75	ND
^{132}Te	0.58	0.097	0.07	0.25	0.46	0.13	0.68	0.48	0.009	0.07	0.98	0.35	0.02	0.21	0.18
^{140}Ba	0.23	1.7	1.0	0.13	0.87	0.11	1.3	0.78	ND	0.13	1.6	1.9	0.23	0.82	0.73
^{141}Ce	0.093	0.92	0.63	0.23	12	0.19	0.88	0.53	0.001	1.3	1.2	1.6	0.08	0.71	0.11
^{143}Ce	1.92	ND	ND	0.71	0.41	0.51	2.0	1.3	0.009	0.34	3.0	1.0	0.23	1.2	1.2
^{147}Nd	0.12	ND	ND	0.25	1.5	0.38	1.0	0.79	ND	0.26	1.4	0.79	0.09	0.77	0.70

*These ratios are based on calculated activities before being rounded off to two significant figures for Table 2.

**Values obtained from USNRDL-456. Ratio of isotope activity to ^{99}Mo activity at H+1 hour.

Table F-2. Isotopic fractionation factor based on ^{99}Mo

Isotope	20528		20540			20542		20543	20544	20545				20546
	A	B	A	B	C	A	B			A	B	C	D	
^{95}Zr	---	---	4.6	19	4.6	15	7	0.87	2.2	17	11	1.1	6.7	9.1
^{97}Zr	---	---	5.1	---	3.9	14	7.2	1.6	1.5	18	21	1.4	6.9	7.4
^{99}Mo	1	1	1	1	1	1	1	1	1	1	1	1	1	1
^{103}Ru	---	---	---	7.6	---	---	4.1	0.17	---	34	56	---	18	---
^{132}Te	0.17	0.12	0.43	0.79	0.22	1.2	0.83	0.015	0.12	1.7	0.60	0.034	0.36	0.31
^{140}Ba	7.6	4.5	0.56	3.8	0.48	5.6	3.4	---	0.56	7.0	8.3	1.0	3.6	3.2
^{141}Ce	9.9	6.8	2.5	130	2.0	9.5	5.7	0.01	14	13	17	0.86	7.6	1.2
^{143}Ce	---	---	0.37	0.21	0.26	1.0	0.68	0.005	0.18	1.6	0.54	0.12	0.62	0.62
^{147}Nd	---	---	2.1	13	3.2	8.3	6.6	---	2.2	12	6.6	0.75	6.4	5.8

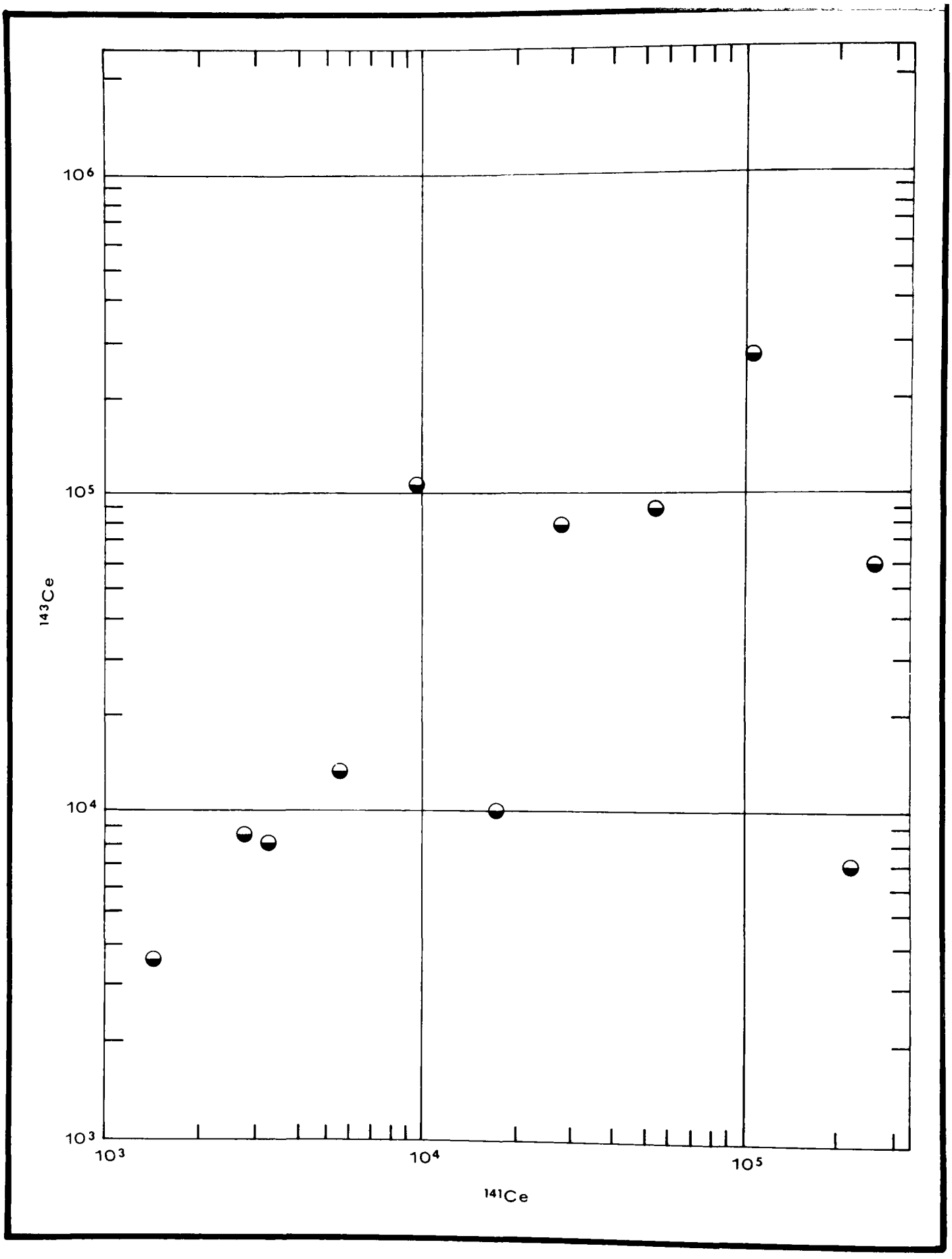

Figure F-1. ^{141}Ce versus ^{143}Ce activity for selected Phoebus 1B, EP-IV particles.

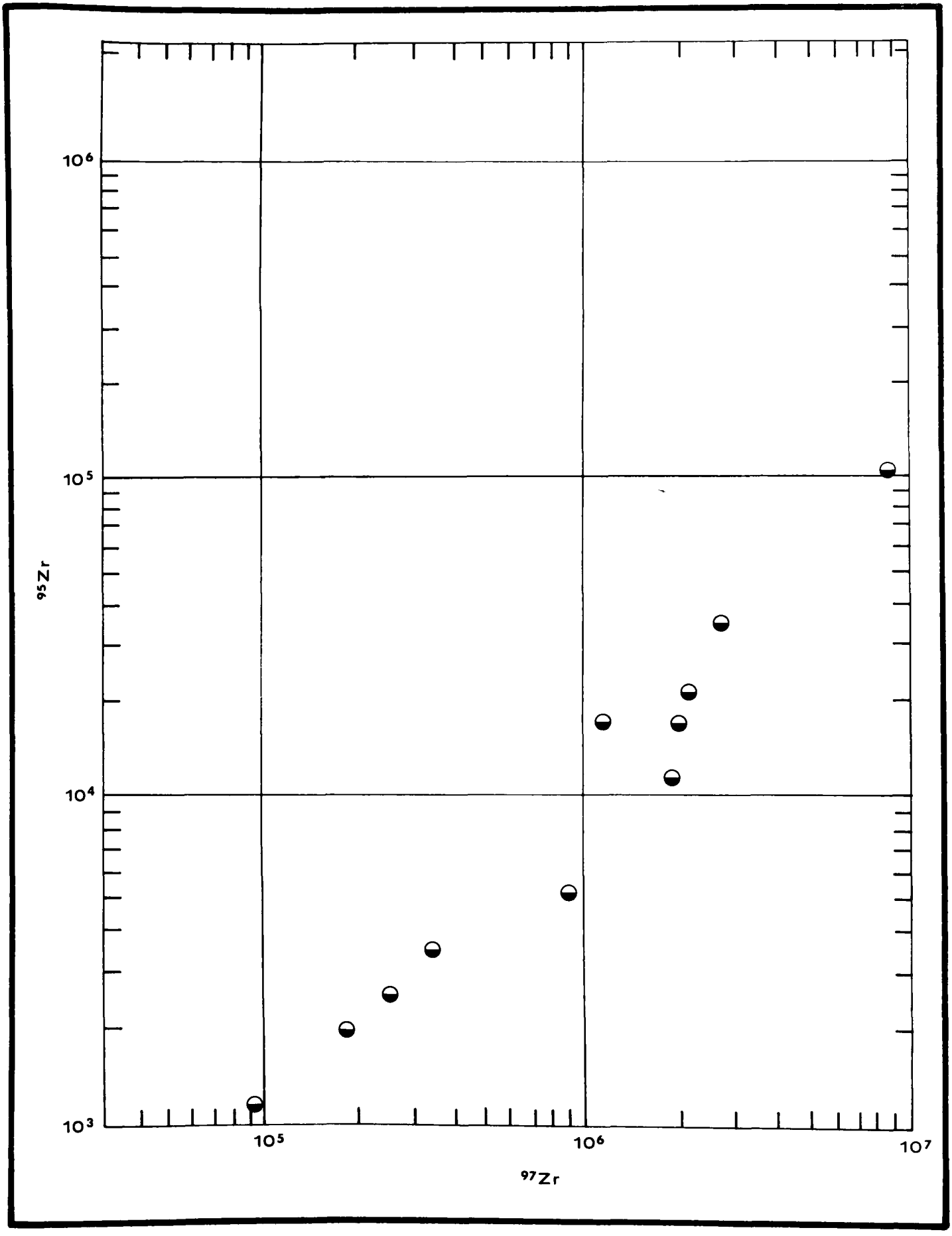

Figure F-2. ^{95}Zr versus ^{97}Zr activity for selected Phoebus 1B, EP-IV particles.

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