UNITED STATES DEPARTMENT OF COMMERCE
WEATHER BUREAU
WASHINGTON

Analysis of Stratospheric Strontium-90 Measurements

L. Machta and R. J. List
U. S. Weather Bureau
March, 1959

Research sponsored by the Division of Biology and Medicine
U. S. Atomic Energy Commission

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I. Introduction

Since it was first proposed by Dr. W. F. Libby[1] in 1953, the concept of stratospheric storage of long-lived fission products resulting from the detonation of high-yield nuclear devices has become generally accepted. As of the end of 1958, Libby[2] has estimated that about 65 megatons of fission products had been injected into the stratosphere. Knowledge of the fate of this debris, which may remain in the stratosphere for a period of years, is vital to our evaluation of the problem of long-lived fission products, such as **Strontium-90 and Cesium-137.**

The atmospheric processes which control the movement of debris in the stratosphere, its eventual removal into the troposphere and its deposition in the biosphere are determining factors in being able to predict future levels of contamination from debris already in the atmosphere and from debris which may possibly be injected in the future. This understanding will also permit the design of an optimum monitoring and sampling program to keep track of the stratospheric Sr-90. A corollary of the study will be a better understanding of the meteorology of the stratosphere, since these fission products represent one of the few tracers available to study stratospheric motions.

In this paper we should like to review the estimates of what is expected to be found in the stratosphere, the various hypotheses advanced concerning the distribution of radioactivity in the stratosphere and its subsequent removal, and how this fits the observed stratospheric concentrations, along with an estimate of the quality of the data obtained.

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II. Estimated Stratospheric Content

It has been customary to divide the radioactive debris resulting from the detonation of nuclear devices into three categories: (1) close-in fallout, (2) tropospheric debris and (3) stratospheric debris. The apportionment in any individual case is a function of the energy of the burst and the conditions of firing (e.g., surface, tower, air, underground, etc.). For bursts

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fired at or near the surface of the ground, a large fraction of the fission-product activity is deposited within the first day in the general area of the test site, due to condensation and scavenging by the large particles sucked into the rising fireball. Those particles which are too small to have appreciable fall rates are carried away from the burst site by the winds. For detonations with yields of less than 1 megaton, all of these small particles are left in the troposphere, where they are eventually removed from the air by precipitation scavenging and impaction at the ground. There is general agreement (Libby,[3] Stewart et al,[4] National Academy of Sciences[5] and many others) that the mean lifetime of a particle in the troposphere, before it is removed, is about a month.

Here, however, we are concerned primarily with the effects of very large yield bursts, those powerful enough to inject large quantities of debris into the stratosphere, i.e., yields greater than one megaton, and in particular with the fate of the Sr-90 resulting from these bursts.

Strontium-90 is singled out, of course, because it is thought to represent the principal long-term potential biological hazard from nuclear testing. However, this isotope is also of particular interest in studying stratospheric motions because of apparent very small particle size. It is produced as a daughter product of Krypton-90 as follows:

$$\text{Kr-90} \xrightarrow{33\ \text{sec}} \text{Rb-90} \xrightarrow{2.7\ \text{min}} \text{Sr-90} \xrightarrow{28\ \text{yr}} \text{Y-90} \xrightarrow{64.6\ \text{hr}} \text{Zr-90}\,(\text{stable})$$

This means that the Sr-90 will tend to condense later than the bulk of the fission products, resulting in the formation of sub-micron particles which are easily carried into the stratosphere by the ascending cloud, and which may have negligible fall rates.

Stratospheric injections of Sr-90, as estimated by Libby,[2] are shown in Table 1. These estimates are based on a series of assumptions and speculations concerning the nature of the fallout. The stratospheric content of Sr-90 is obtained by the subtraction of local and tropospheric fallout from the total Sr-90 radioactivity produced by nuclear explosions whose clouds enter the stratosphere. Local fallout is assumed to be 80 percent for land surface shots, 20 percent for surface water shots and 10 percent for air shots. Shots of more than a megaton yield are assumed to put 1 percent of the debris which did not fall out locally into the troposphere and the remainder into the stratosphere. Shots in the kiloton range are assigned to the troposphere.

There are many uncertainties in these estimates. For example, the 80 percent local fallout from surface bursts is primarily based on very incomplete monitoring data for the Castle Bravo shot, and even if the distribution of total fallout of gross fission products were known, the question of fractionation of the Sr-90 remains unsolved. There is uncertainty concerning the fraction of debris from large shots which remainsin the troposphere, with evidence that it is as much as 5 percent. Another uncertainty is the yield and conditions of firing of the USSR tests. With all these uncertainties, it is obvious that direct sampling is needed to determine the actual Sr-90 content of the stratosphere.

Table 1

Estimated Stratospheric Injections
(after Libby[2])

		MT
1952	Fall	1.0
1954	Spring	19.5
1955	Fall	1.2
1956	Summer	5.5
	Fall	3.0
1957	Spring	3.0
	Fall	4.5
	Fall	1.5
1958	Winter	3.3
	Spring	4.0
	Fall	20.0

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III. Other measurements

The reservoir of radioactive debris in the stratosphere has been established by direct measurement, at least in the lower stratosphere, by several investigators prior to the establishment of the AEC balloon sampling program. However, these earlier techniques were confined to total activity measurements rather than to the determination of specific isotopes. In 1954 and early

1955 so much of the debris originated from the Spring 1954 test series (Castle) that one could reasonably assign an origin to the fission products and deal with the gross activity, if fractionation could be neglected. After 1955, the number of tests were so numerous that almost all samples contained fission products from many tests. The method of estimating individual isotopes from the gross fission product activity and burst assignment was no longer as useful.

Aircraft filtering flights reaching into the lower stratosphere have been reported by Stewart et al,[4] by Aler, Bjornerstedt, Edvarson and Low[6] in Sweden, and by Hvinden[7] in Norway. All showed significant increases in the long-lived radioactivity of the air with increasing altitude, and a relatively sharp increase at or near the tropopause. The results are shown in Figure 1.

Ishii[8], in Japan, employed a technique utilizing balloon-borne Geiger counters to attain altitudes of 50,000 feet, compared to the 40,000 feet attained by the aircraft. However, this technique had the drawback of measuring the total gamma activity in situ, and it was necessary to subtract the cosmic ray contribution based on a knowledge of the mean distribution of cosmic-ray activity with altitude. The two contributions were of about the same order of magnitude in the lower stratosphere. While these flights showed higher concentrations of fission products at the tropopause level, they did not show the sharp gradient at the tropopause or the large differences between the troposphere and stratosphere.

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IV. Models of Stratospheric Storage and Fallout

Utilizing the estimates of stratospheric injection, the scanty upper-air measurements available and the observations of surface deposition, several estimates of the probable residence time of Sr-90 particles in the stratosphere have been made. Libby[3] estimates about 10 $\pm$ 5 years, Stewart et al[4], 5-10 years, Machta[9], about 5 years, and the U.N. Scientific Committee[10] a value of about 8 years with a range of 5-10 years. All of these estimates imply a stratospheric structure at variance with known meteorological principles (this has been recognized by the last two references cited). The concept of an unvarying mean residence time for stratospheric debris would imply uniform mixing in the stratosphere with a semi-permeable membrane at the tropopause and would be independent of the actual altitude

of injection of the debris. Such conditions do not, of course, prevail in the stratosphere.

Proposed models of stratospheric transport and removal have been developed by several investigations to explain the observed Sr-90 patterns at the ground. Three such models will be mentioned here.

The first, due to Dr. Libby, assumes that in the main, debris is uniformly distributed throughout the stratosphere relatively quickly, that a fixed fraction per year of the stratospheric debris enters the troposphere more or less uniformly over the world, and that deposition is principally in precipitation (this last assumption is common to all models). Irregularities in the observed distribution such as the large Sr-90 deposition observed in the north temperate latitudes and apparent changes in the rate of deposition at certain stations are assumed to be the result of tropospheric fallout from kiloton-yield tests in Nevada and the USSR.

Martell has modified this simple model to allow for a difference between the behavior of debris from the large U. S. Pacific shots and the USSR thermonuclear bursts which inject their debris into the polar or temperate latitude stratosphere. It is his contention that the U. S. Pacific bursts contribute to a relatively uniform world-wide stratospheric fallout, while the stratospheric debris from the USSR thermonuclear bursts has a shorter residence time in the stratosphere, of the order of a few months. He concludes that the increased fallout in the north temperate latitudes is due to stratospheric fallout from Russian debris which was injected into the lowest layers of the stratosphere.

Machta and List[12] in this country and Stewart and others in England have developed a model originally proposed by Brewer and Dobson based in part on independent meteorological evidence from water vapor and ozone distribution, which calls for a principal source of stratospheric air in the ascending currents of the tropics, with a slow poleward drift, particularly in the winter hemisphere, and a sink over the temperate and polar regions. This circulation, together with the known precipitation patterns, is used to explain the observed fallout of Sr-90 and the apparent increase in the rate of fallout during the spring season.

Each of these models would predict a somewhat different distribution of Sr-90 in the stratosphere. Dr. Libby's would, of course, call for a uniform

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the atmosphere sampled on the several flights which sampled through appreciable vertical thickness. A few flights made in Arizona are shown on the Texas figure in italics. The arrows at the top of Fig. 2 (Minneapolis) indicate the dates of the large Russian nuclear detonations which presumably injected debris into the temperate or polar stratosphere. Similarly, the arrows at the top of Fig. 4 (Panama) indicate the megaton range devices detonated by the United States and the United Kingdom in tropical latitudes.

An inspection of the data shows that there is a large variability. This variability may be real, and a measure of the true state of affairs in the stratosphere, or it may be a function of the collection or analysis techniques. Some of the variability must be a reflection of recent injections into the stratosphere. For example, the high values over Minneapolis in late April and May of 1957 may be a result of the April Russian thermonuclear tests, similarly the high value over South America in July, 1957, may reflect injections from the U. K. tests of the preceding month.

A major difficulty in making a quantitative estimate of the Sr-90 content of the stratosphere results from the lack of precise knowledge of the efficiency of the filter material at the low pressures and flow rates encountered, together with a lack of knowledge of the particle sizes involved. Although theoretical and laboratory studies on the characteristics of the filter system have yielded information for particles as small as 0.088 μ, a comparison of collections made by aircraft at 40,000 feet simultaneously with balloon collections indicate that the collection efficiency curves must be extrapolated to 0.02μ or smaller particles to explain the Sr-90 values. Using this semi-empirical approach, a series of filter efficiency factors for Sr-90 particles of 0.02μ were obtained (Table 2).

Table 2 NAS

Filter efficiency for 0.02μ particles

Altitude, feet, MSL	Filter Efficiency
50,000	0.30
65,000	0.38
80,000	0.60
90,000	0.82

The sparsity of stations and the variability of the data make it impracticable to draw an instantaneous picture of the world-wide distribution of Sr-90. However, it is possible to make a rough estimate of the average concentration and distribution of debris in the stratosphere over an extended period of the order of six months. Figure 6 shows the average concentration of debris in the stratosphere as a function of altitude and latitude for two six-month periods, January - June, 1957, and January-June, 1958. The data at each sampling altitude and station were averaged over the period and corrected for the filter efficiencies given in Table 2.

It is evident that the sixteen data points, each based on a mean of only a few observations with large variability cannot serve to define the stratospheric Sr-90 content of the world. However, some information as to the distribution can be extracted. A series of isolines showing what is felt to be the most probable distribution based on the available observations are also shown, although there are admittedly many other logical patterns which may be drawn.

A comparison of the two cross-sections shows that between the first half of 1957 and the first half of 1958, there appeared to be a general decrease in the intensity of the gradients observed, principally due to a decrease in the concentration of Sr-90 at the two northernmost stations at 65,000 feet. Another evident change is the increase at all levels over the Canal Zone, possibly as a result of the British thermonuclear tests in the Christmas Island area which occurred in May, June, and November of 1957. These tests may also be responsible for the increases at 90,000 feet over Texas and Minnesota.

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The patterns shown in Figure 6 may be integrated to estimate the total stratospheric burden of Sr-90. This computation gives the equivalent of 7.8 MT of fission products in 1957 and 5.5 MT in 1958. (A computation based on 0.25 filter efficiency factor at all altitudes gives 12 MT in 1957 and 8 MT in 1958). These figures can be compared with estimates of the stratospheric content made by budgeting the debris. Table 1 shows that about 39 MT had been injected into the stratosphere as of the end of 1957. However, much of this debris has been removed from the stratosphere. Libby estimates the mean removal rate to be about 10% per year, Machta and List about 20% per year. These estimates would result in a stratospheric content of 20 to 24 MT

in early 1957 and 26 to 34 MT in early 1958. In either case, it is apparent that the uncertain estimate of the stratospheric content from the balloon data is appreciably smaller than that estimated from the budgeting of bomb releases and the change from 1957 to 1958 is in the opposite direction.

The discrepancy in the two calculations of the stratospheric Sr-90 content can result from a variety of causes. The uncertainties of the estimate based on the amount injected have been discussed. An equal or possibly larger uncertainty exists in the estimate derived from the balloon measurements. Among the problems are:

A. The representativeness of the limited number of measurements.

B. Knowledge of the actual amount of air filtered on any given flight.

C. Efficiency of the filter for the particle sizes involved.

D. Radiochemical analysis.

A. Representativeness of the data. If the high atmosphere is indeed as variable as the stratospheric balloon flights indicate, then the few samples, one flight a month at a total of 16 places in the atmosphere (4 stations at 4 altitudes), cannot be considered adequate to define the world-wide stratospheric content with the desired accuracy. Although there is no conclusive evidence to preclude the possibility that this variability is real, a consideration of diffusion rates suggests that the fission products are not as patchy as the balloon data indicate.

B. Volume of air filtered. The question of the reliability of the fan law used in calculating the volume of air passing through the filter is under investigation by General Mills. In an attempt to see if there is an obvious bias in the results from this cause, the Sr-90 content of the samples was plotted against the reported volume at the various stations and altitudes. If a consistent error in the fan law were present, this would appear as a relationship between the apparent volume of air filtered and the reported air concentration. No such relationship was found. This, of course, does not rule out the possibility of other non-systematic errors in the determination of the volume filtered.

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C. Filter efficiency. Many studies have already been made on the problem of the efficiency of air filters for small particles, but the basic problem - the size of the particles with which we are dealing - has not been solved. The extrapolation of laboratory determinations of collection efficiency based on

larger particles, to particles as small as 0.02μ is certainly unreliable, as is the use of the airplane comparison in indirectly determining the particle size. Also adding to the uncertainty in the filter efficiency problem, is the question of flow-rate. It has been assumed that a linear velocity of 100 feet per minute prevails at all times and altitudes. It may perhaps be necessary to determine the flow rate for individual collections, again based on the fan law, once the particle size and efficiency curves are established. Such refinement is probably not warranted at present.

D. Radiochemical Analysis. The stratospheric filters are routinely analyzed for the following fission products:

Table 3

Isotopes Analyzed

Isotope	Half-life
Ba-140	12.8 days
Zr-95	65 days
Ce-144	275 days
Cs-137	28.8 years
Sr-89	50.5 days
Sr-90	27.7 years

Sr-90 is, of course, the isotope of principal concern in evaluating the long-term hazard of delayed fallout. Cs-137, with a similar half-life, fission yield and mode of formation serves as a check on the data and is also of interest as a long-term fallout hazard.

Information on the observed Cs-137/Sr-90 ratio in fallout is available from other sources and can be compared with the balloon filter results. Baus et al[13] of the Naval Research Laboratory have determined the ratio in air filtered at the ground at a series of stations along the 80°W meridian extending from Thule, Greenland, to Punta Arenas, Chile, using monthly collections. Stewart et al[14] in England have made measurements using a world-wide precipitation network making monthly collections. Storebø[15] in Norway, used Stewart's data, together with some additional measurements in Norway, to conclude that there is a difference in behavior of the two isotopes, and suggests that the Sr-90 is more easily captured by rain. If this is a result of some physical parameter, like particle size, a systematic distribution of the Cs-137/Sr-90 ratio with altitude should be seen in the balloon data.

Figure 7 shows the balloon Cs-137/Sr-90 data as a function of altitude. Only those samples with more Sr-90 than 10 dpm per 1000 scf were used to avoid spurious values. The several laboratories involved in the radiochemical analysis program are differentiated by symbols. The results show that there is a tendency for the ratio to increase with altitude, although the variability is large. The data are summarized in Table 4, together with the data from the other investigators. (Certain doubtful values have been excluded in the U.K. rainfall results.)

Table 4

Summary of Cs-137/Sr-90 Data

	No. Cases	Mean	Standard Deviation
Balloon filters:			
50,000 feet	13	2.11	1.11
65,000 feet	48	2.80	1.31
80,000 feet	26	3.42	1.45
90,000 feet	15	3.53	1.99
NRL surface air filters	59	2.20	0.73
U. K. rainfall	122	1.63	0.63

As can be seen from the last column of Table 4, the standard deviation of the Cs-137/Sr-90 ratio from the balloon filters is two to three times greater than found by other investigators. This may be a result of radiochemical procedures and is undoubtedly a function of the extremely minute amounts of material collected during the flights.

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The relatively short-lived isotopes, Ba-140, Zr-95 and Sr-89, can reveal information on the age and origin of the activity sampled, particularly if the debris is fresh, but the great variability found in these isotopes has made interpretation difficult. This variability may be real, reflecting the patchy nature of fresh debris.

Cerium-144 tended to show more meaningful patterns than the shorter-lived isotopes. It has a half-life of 275 days (0.75 yr) and a fission yield about 25% greater than that of Sr-90. If it can be assumed that fractionation is unimportant and the collection efficiency of the filter for the two isotopes is about equal, the Ce-144/Sr-90 ratio can be useful in distinguishing between sources of debris of different ages. In the first half of 1957, the ratio for Castle debris should be about 3 or 4, while Redwing debris would have a value

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of about 25. For very fresh debris, less than 2 months old, the ratio would be over 40. The proviso concerning fractionation and collection efficiency is an important one, since unlike Sr-90, Ce-144 has no gaseous or volatile precursor, which may indicate that larger particle sizes are involved having greater fall rates. These larger particles, if present, may also be more readily captured by the filter. Absolute values of the ratio may be meaningless, but relative values may be useful in comparing ages of debris at the same altitude.

Figures 8-11 show the Ce-144/Sr-90 ratio (large figures) superimposed on the Sr-90 data. As with the other balloon filter results, there appear to be random fluctuations in the data. However, some patterns can be seen. For example, increases in the Ce-144/Sr-90 ratio at 50,000 feet over Minneapolis and at 50,000 and 65,000 feet over Texas in late 1957 and early 1958 could be the result of Russian injection during the autumn of 1957. In general, the values at 65,000 feet over the two northern stations indicate much younger debris (higher Ce-144/Sr-90 ratio) than over Brazil at the same altitude. At Brazil, increases in the ratio can be seen as a result of the British tests in the spring of 1957 and of the U. S. Pacific test series in 1958.

The foregoing discussion of the radiochemical data would seem to indicate that some qualitative reliance may be placed on the results, but that the variability seems to be larger than expected and may partially be a result of analysis procedures. The very minute quantities of debris captured by the sampling equipment make the radiochemical procedures extremely difficult.

VI. Conclusions

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In evaluating the possible hazard from Sr-90 and other long-lived fission products, it has been established that stratospheric storage plays an important role. It then becomes necessary to know the content, distribution and rate of removal of debris from the stratosphere. Indirect determinations of the content, based on assumed initial injections, are too uncertain and meteorological knowledge of stratospheric motions is too poor to satisfy questions on these subjects. We must rely on direct measurement of the stratosphere.

It is evident that there are many uncertainties in the data collected in stratospheric sampling programs, but that a potential exists for a valuable contribution, by good sampling and analysis, both to the Sr-90 problem

and to the meteorological problem of stratospheric transport, diffusion and removal. The basic problems of particle size and filter efficiency have to be solved. It may be that an indirect method, such as comparison with aircraft or whole air collection, will be the only way to provide a solution to the calibration of the balloon sampling. In that event, a number of comparisons at various altitudes will be necessary to establish this calibration. It will also be necessary to perform these comparisons for each of the isotopes of interest.

If the mechanical problems related to the collection of debris can be solved, the question of the quality of the radiochemical analyses becomes even more important. The large unexplained fluctuations in the ratios of the various isotopes suggests that much of the variability arises from this source. If it turns out that the variations observed in the past data are indeed real, then the four-station network sampling each altitude once a month now in existence would be inadequate. However, information presently available concerning the stratosphere suggests that the variations are improbable, especially when viewed in light of the estimate that most of the debris injected into the stratosphere before 1958, came from the Castle series in the Spring of 1954, three to four years prior to the measurements disuussed here.

It is also possible that the relatively low stratospheric content indicated by the balloon data, and the decrease between 1957 and 1958, represents the best estimate, and that the models of injection and stratospheric-tropospheric exchange that have been employed are grossly in error. This would imply a much faster exchange than has been postulated or smaller initial injections or both.

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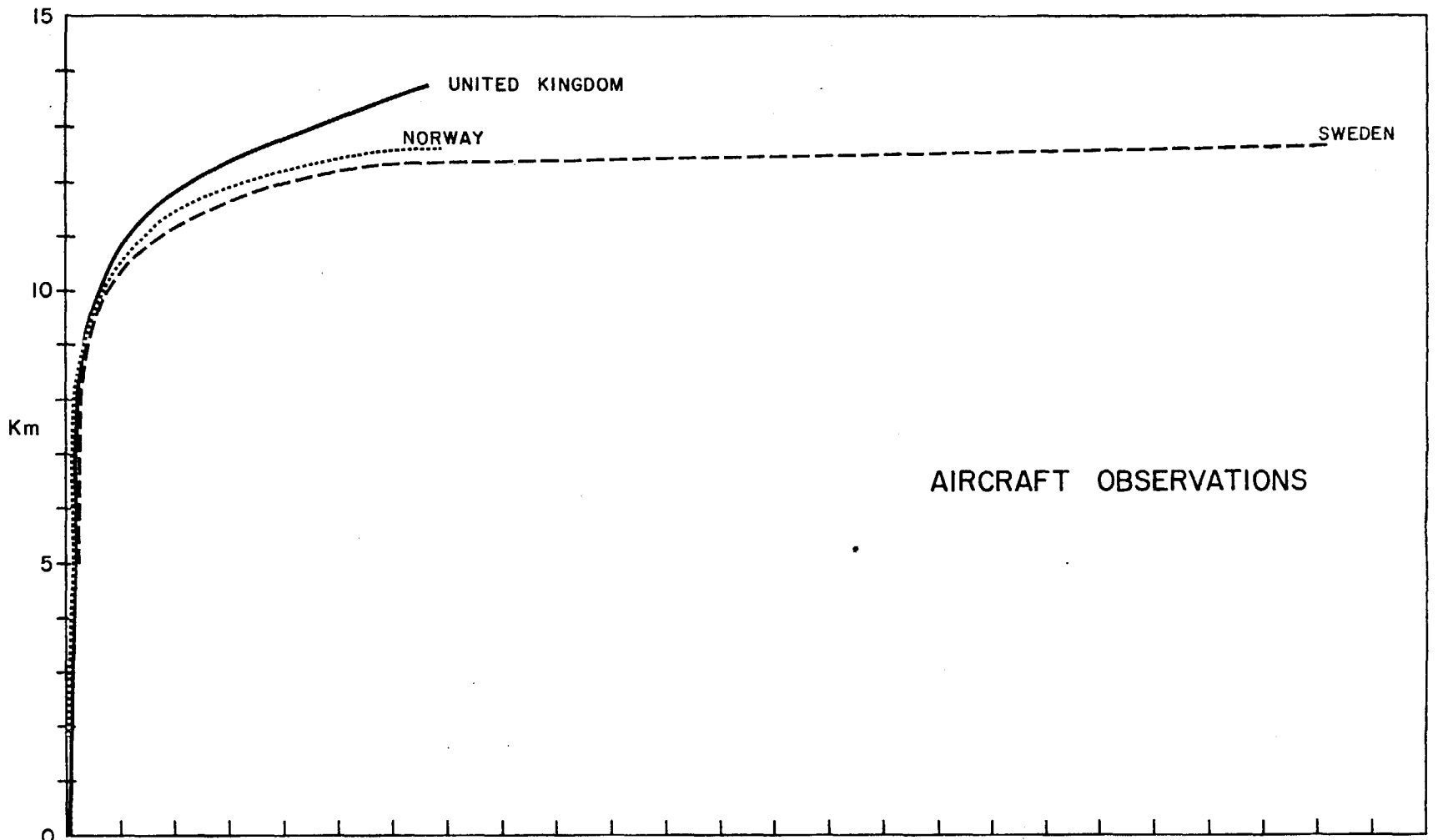
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RELATIVE CONCENTRATION

FIGURE 1

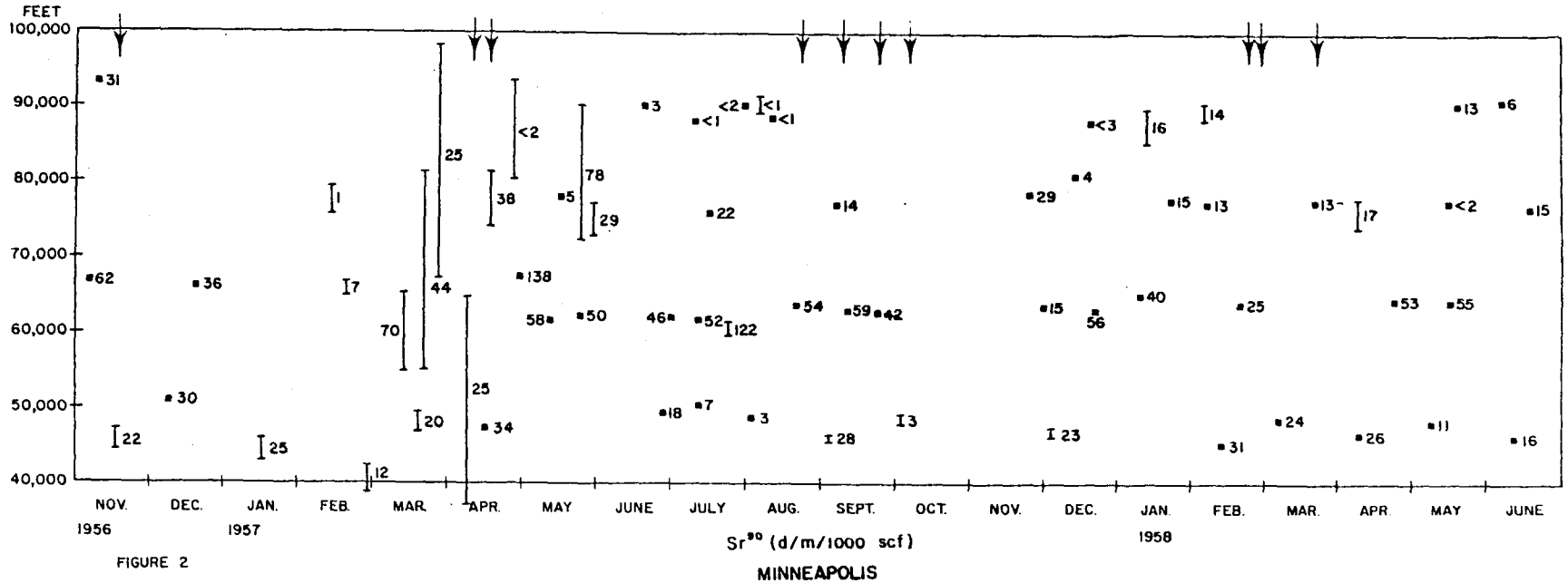


FIGURE 2

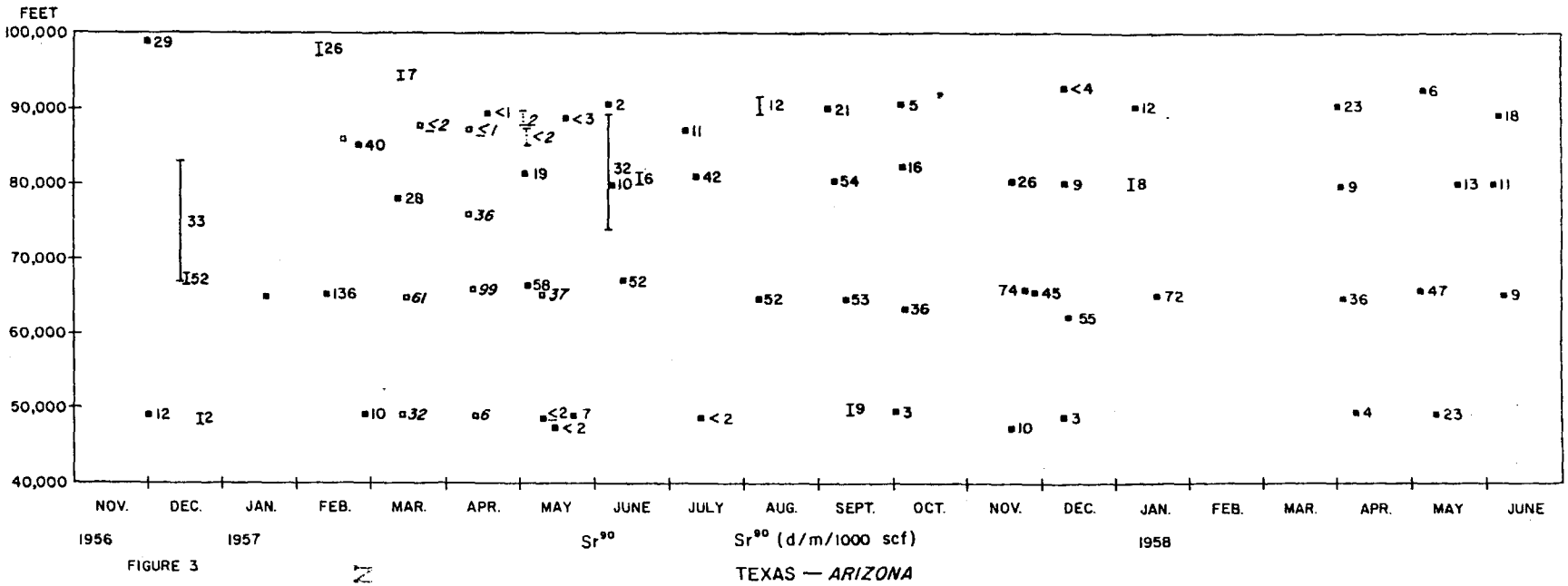


FIGURE 3

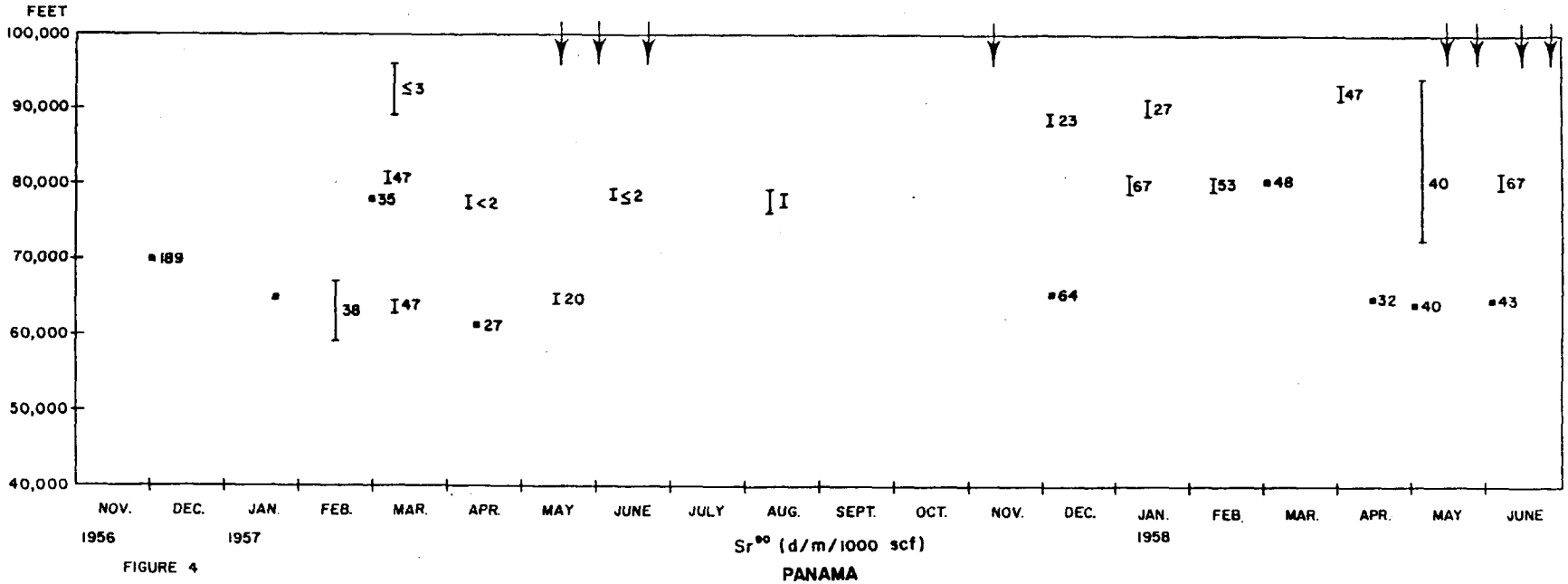


FIGURE 4

Sr^{90} (d/m/1000 scf)

PANAMA

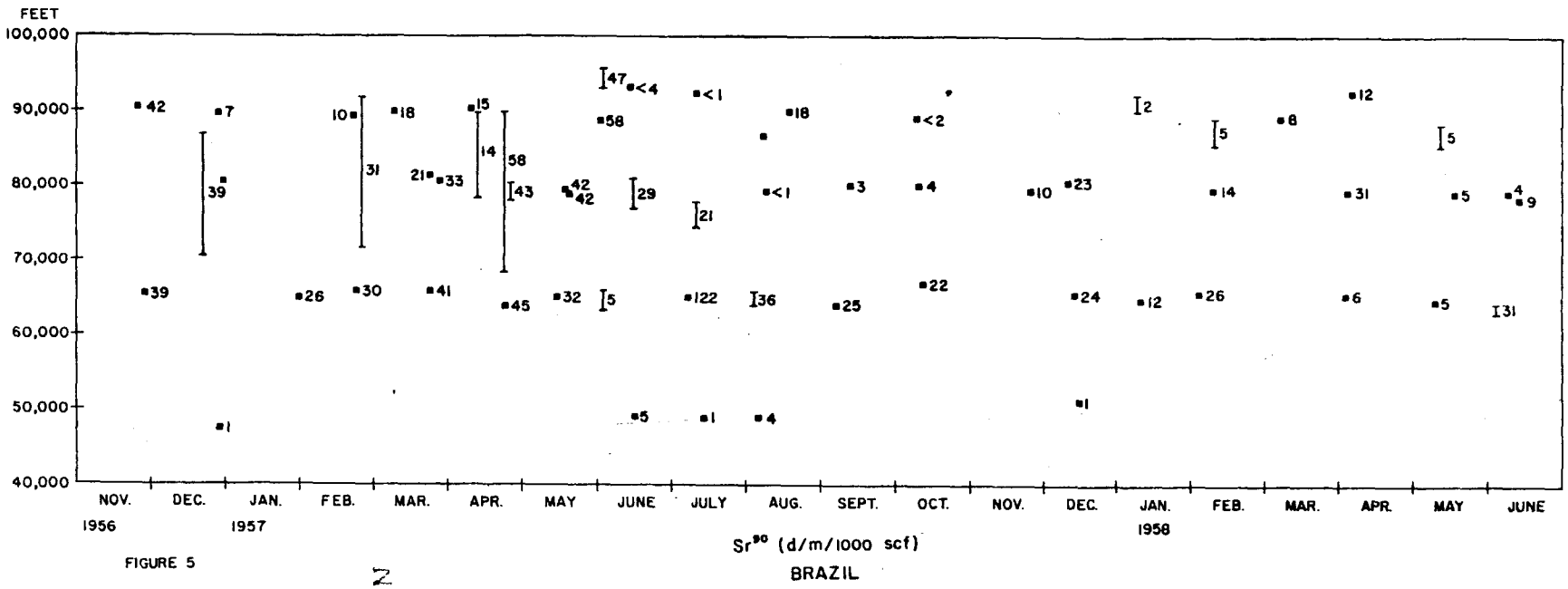
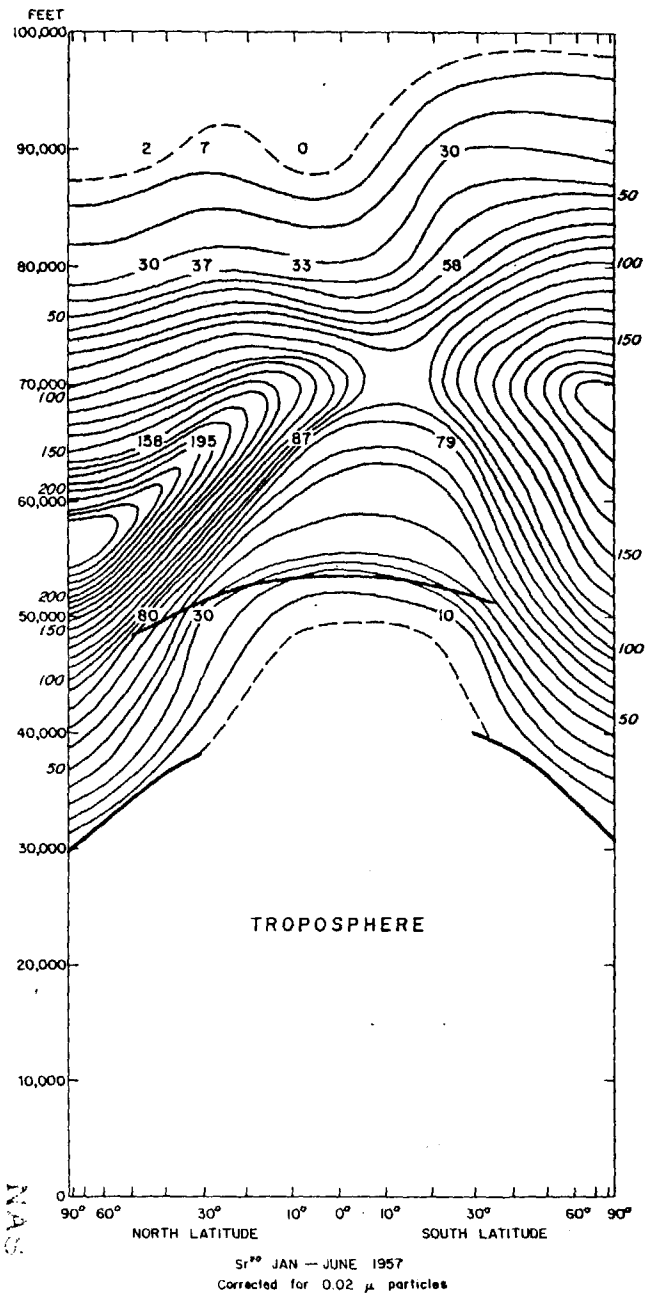


FIGURE 5

Sr^{90} (d/m/1000 scf)

BRAZIL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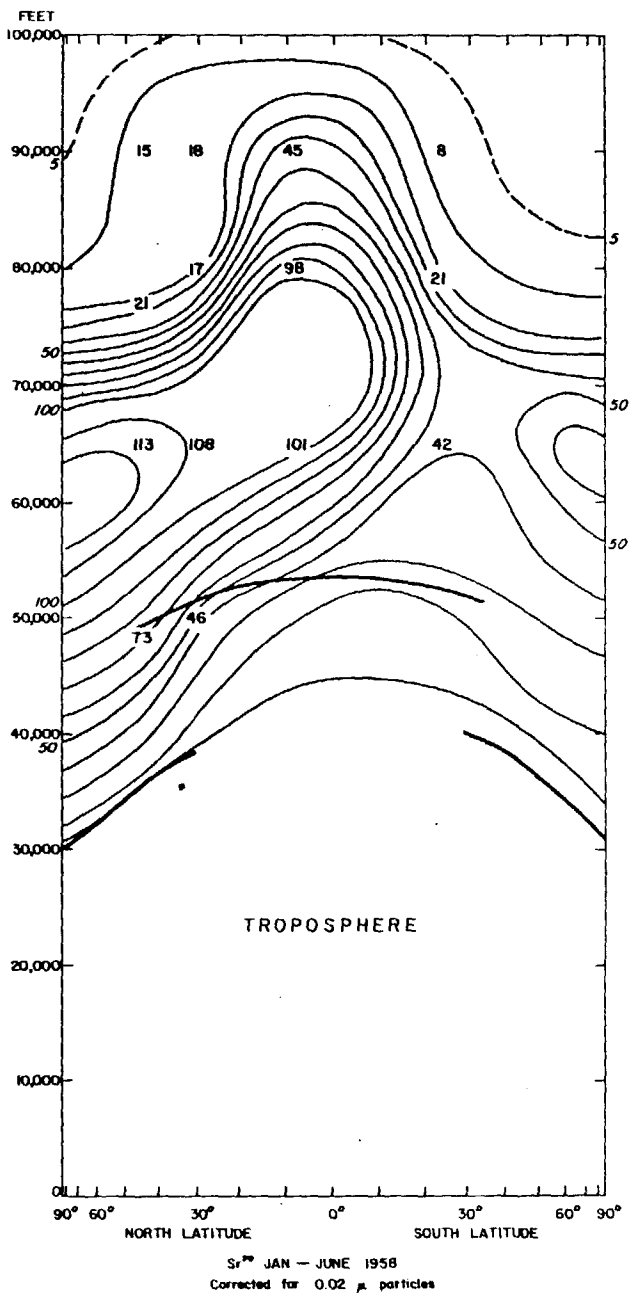
NORTH LATITUDE
SOUTH LATITUDE
Sr⁹⁰ JAN — JUNE 1957
Corrected for 0.02 μ particles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Corrected for 0.02 μ particles

FIGURE 6

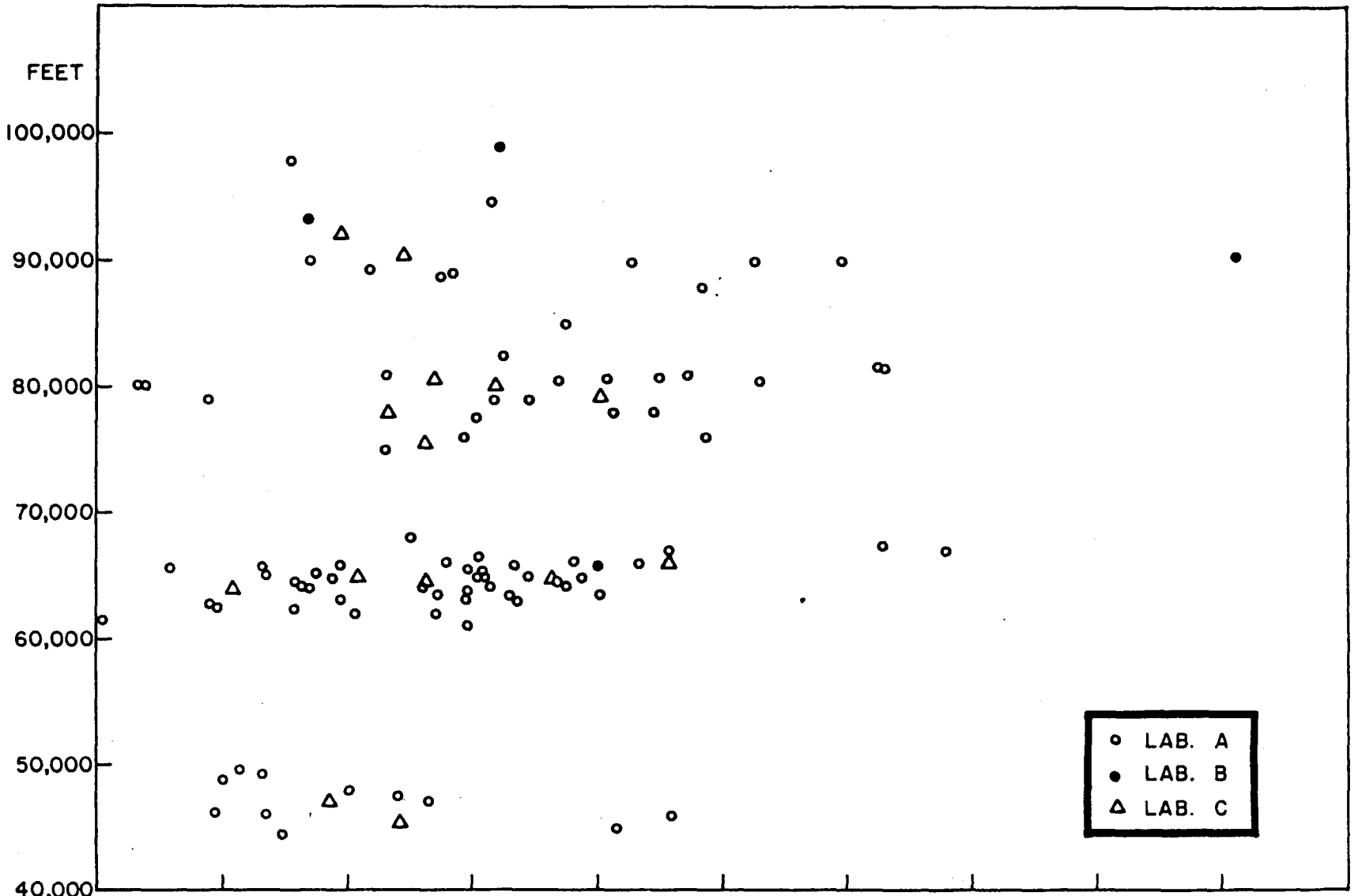

FIGURE 7

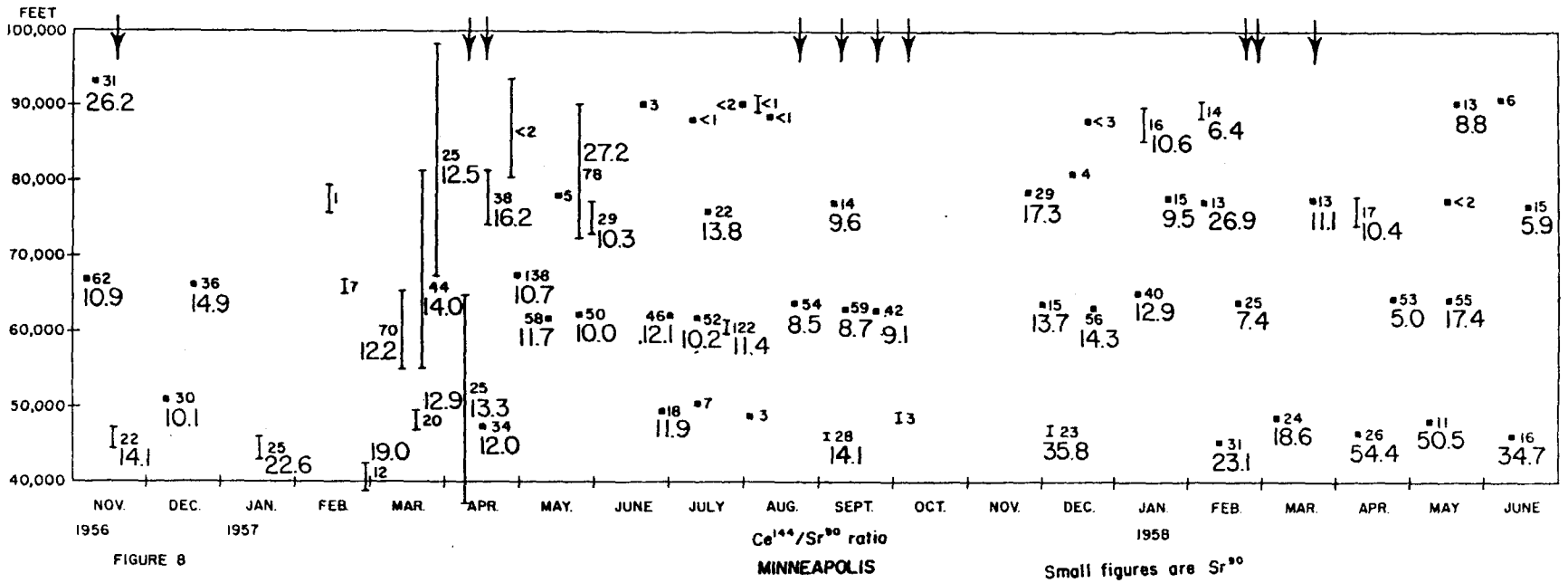

FIGURE 8

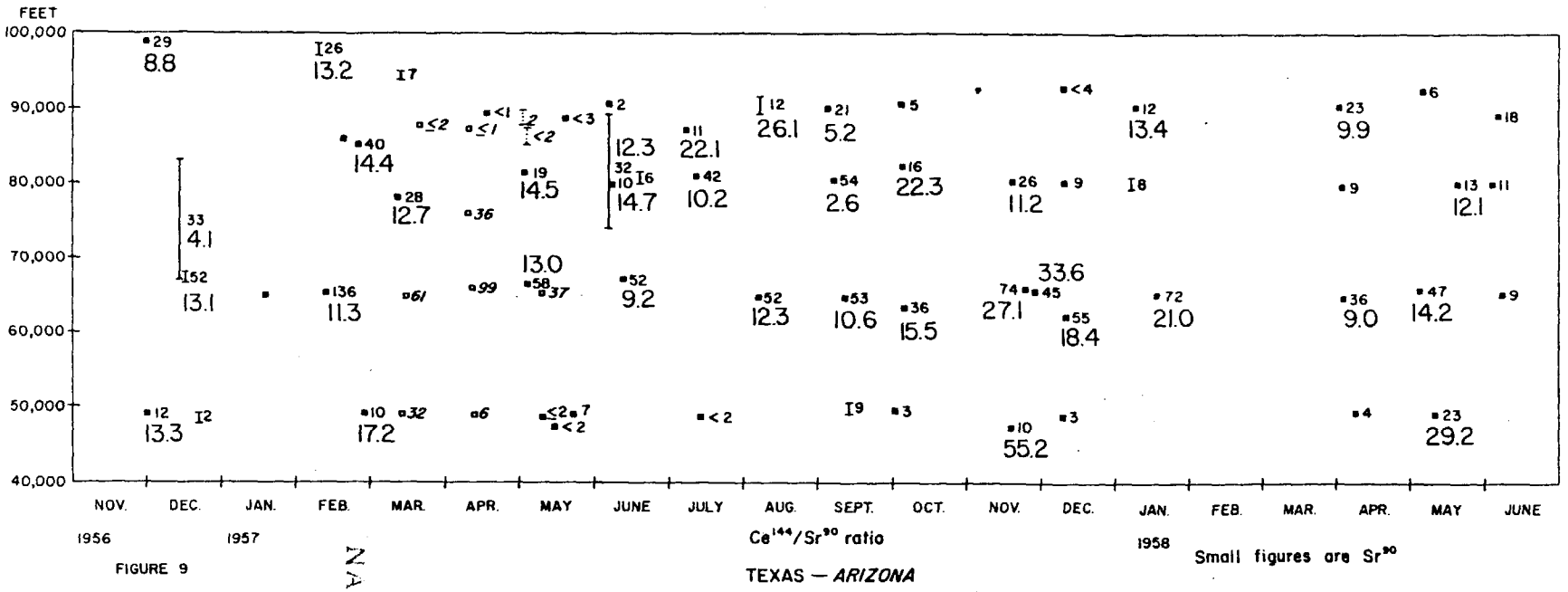

FIGURE 9

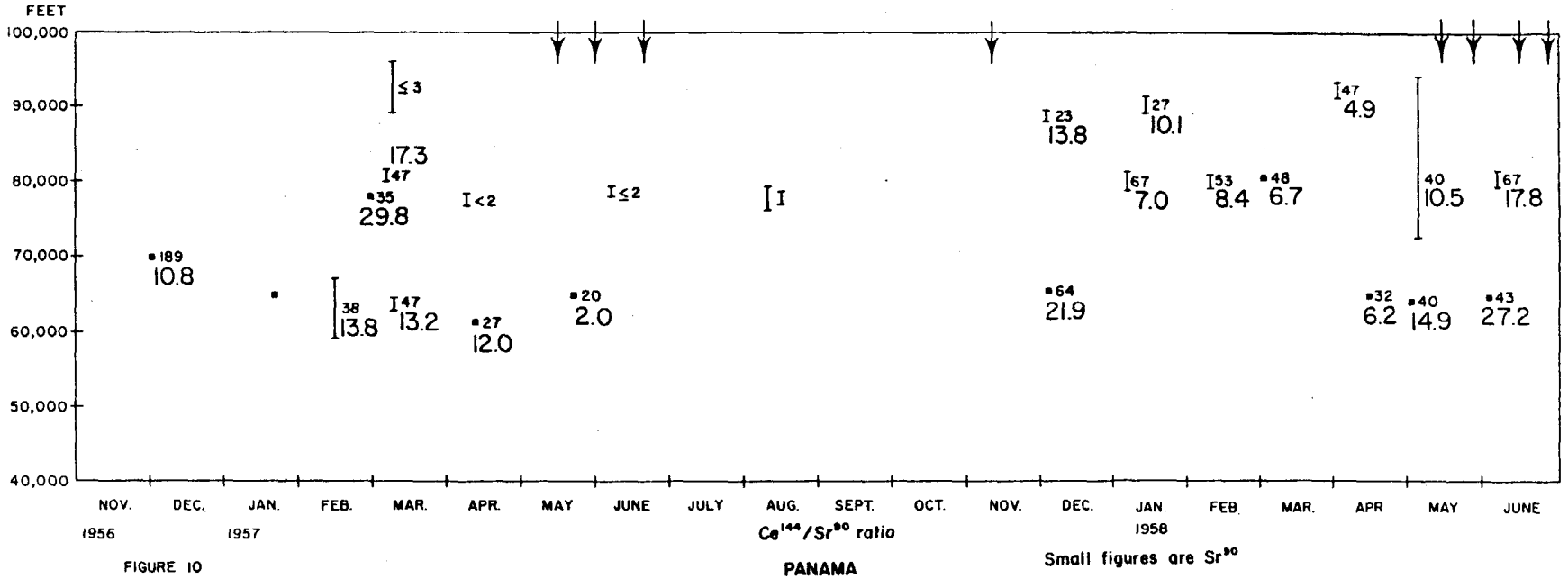

FIGURE 10

PANAMA

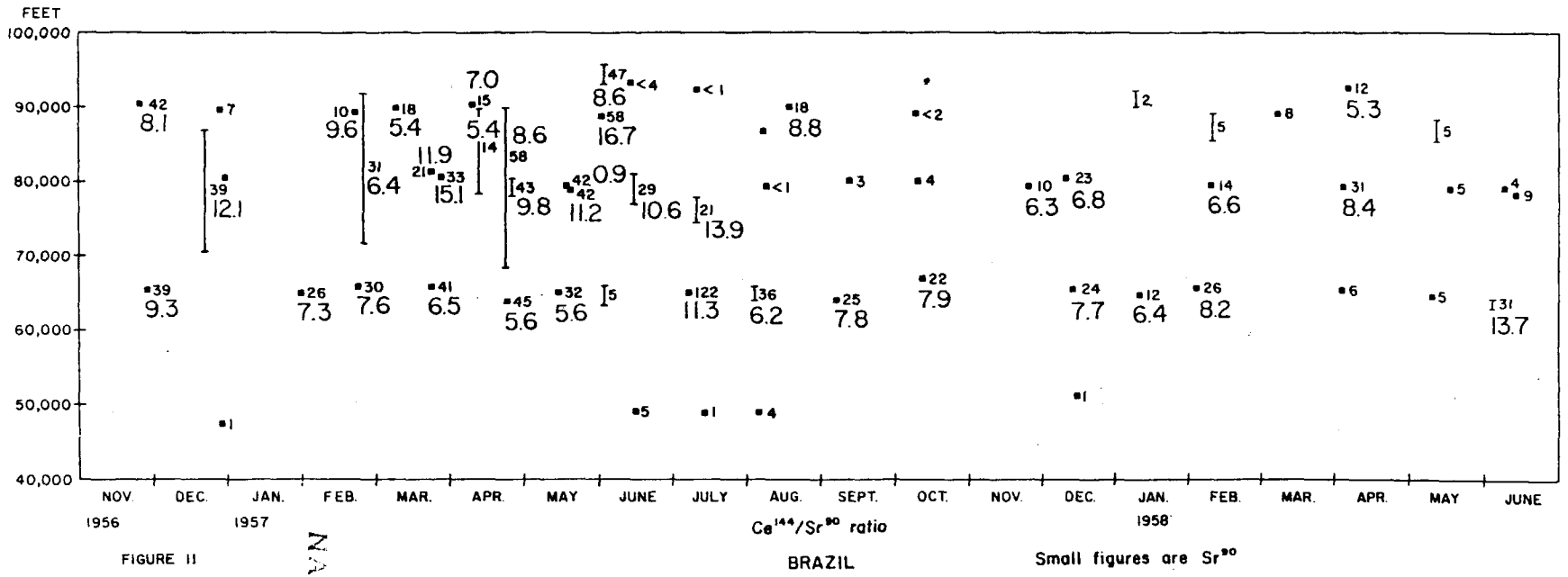

FIGURE 11

BRAZIL