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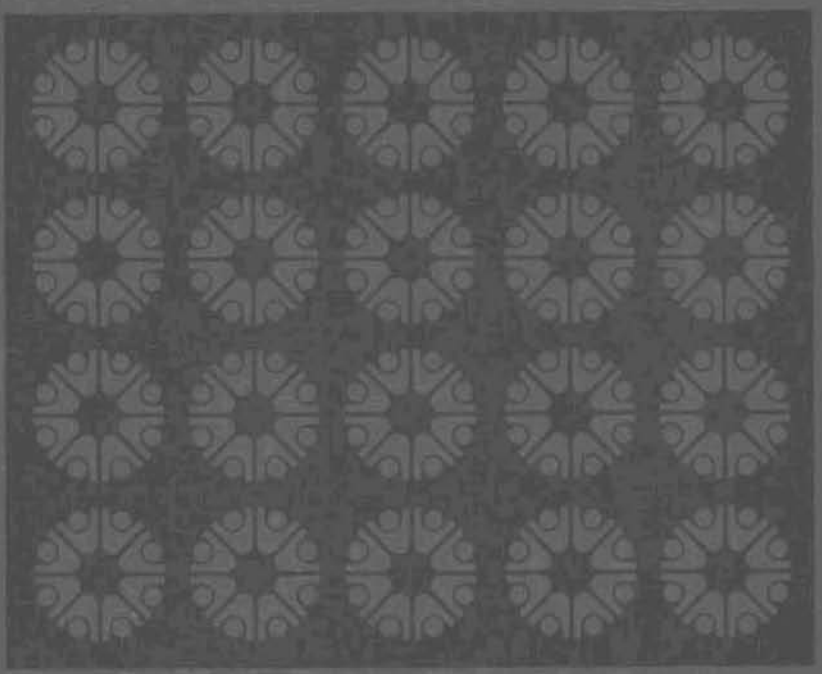
AEC Research and Development Report

SOME CHARACTERISTICS OF POTENTIAL RADIOACTIVE RELEASES TO THE ATMOSPHERE FOLLOWING HYPOTHETICAL REACTOR ACCIDENTS

JUNE 1972



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SOME CHARACTERISTICS OF POTENTIAL RADIOACTIVE RELEASES TO THE ATMOSPHERE FOLLOWING HYPOTHETICAL REACTOR ACCIDENTS

E. C. Watson and D. L. Strenge

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SOME CHARACTERISTICS OF POTENTIAL RADIOACTIVE RELEASES TO THE ATMOSPHERE FOLLOWING HYPOTHETICAL REACTOR ACCIDENTS

E. C. Watson and D. L. Strenge

More than half of the approximately 450 nuclides produced in nuclear fission are unstable. These radionuclides decay with half-lives which vary from less than a second up to more than a million years. Beta particles and gamma radiation are the most common types of emissions associated with their decay.

The relative composition of a radioactive fission product inventory produced in reactor fuel depends on, among other things, the duration of irradiation of the fuel in the reactor core. The quantity of fission products generated, however, depends on the power level. This is to say: the relative composition of fission products in fuel irradiated to 10,000 MWd per ton at 40 MW per ton will differ substantially from that irradiated at 20 MW per ton. The effect of irradiation time on fission product inventories becomes apparent when the fission product activities versus decay time are examined. The computer code RIBD[1] was used to calculate fission product inventories for several arbitrarily selected irradiation times and for decay times out to two years. The results are summarized in Figure 1.

The relative composition of nuclides which may reach the containment vessel spaces following a fuel failure accident would differ considerably from that of the fission product inventory in the fuel. This difference results from complex fractionation processes which occur as these nuclides travel along tortuous paths from the interstitual spaces in the fuel through the primary containment system to the containment vessel's atmosphere. It is common practice to simplify these fractionation processes by classifying fission products into groups and then assign assumed release fractions from failed fuel to each group. A commonly used

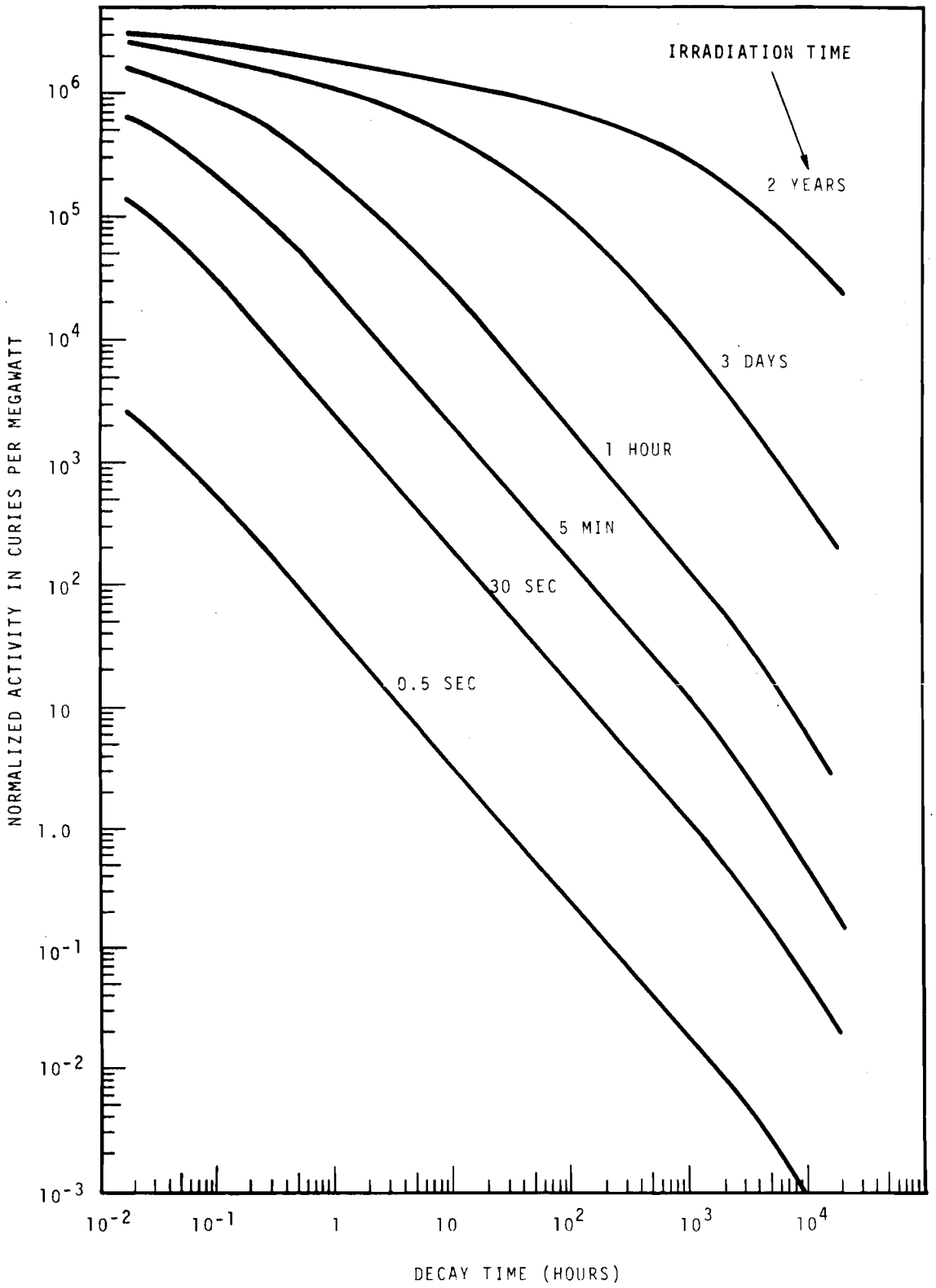

FIGURE 1. Effect of Irradiation Time on Activity - Decay Relationship - All Fission Products

classification is that listed by DiNunno, et al.[2] These groups and their assumed release fractions are:

Noble Gases (Kr, Xe)	1
Halogens (Br, I)	0.5
All Remaining Fission Products (Including nuclear fuel)	0.01

Different classifications of fission products and other release fractions have been assumed in other studies;[3] however, they all have in common, noble gases as a classification with a failed fuel release fraction of one.

Although irradiation time has an effect on the isotopic composition of fission products, it does not drastically alter the relative group composition. For example, from a few minutes to one day after shutdown the noble gas inventory is approximately 10% of the total inventory, irrespective of the fuel irradiation time. The activities of several groups at twenty minutes after shutdown are illustrated in Figure 2.

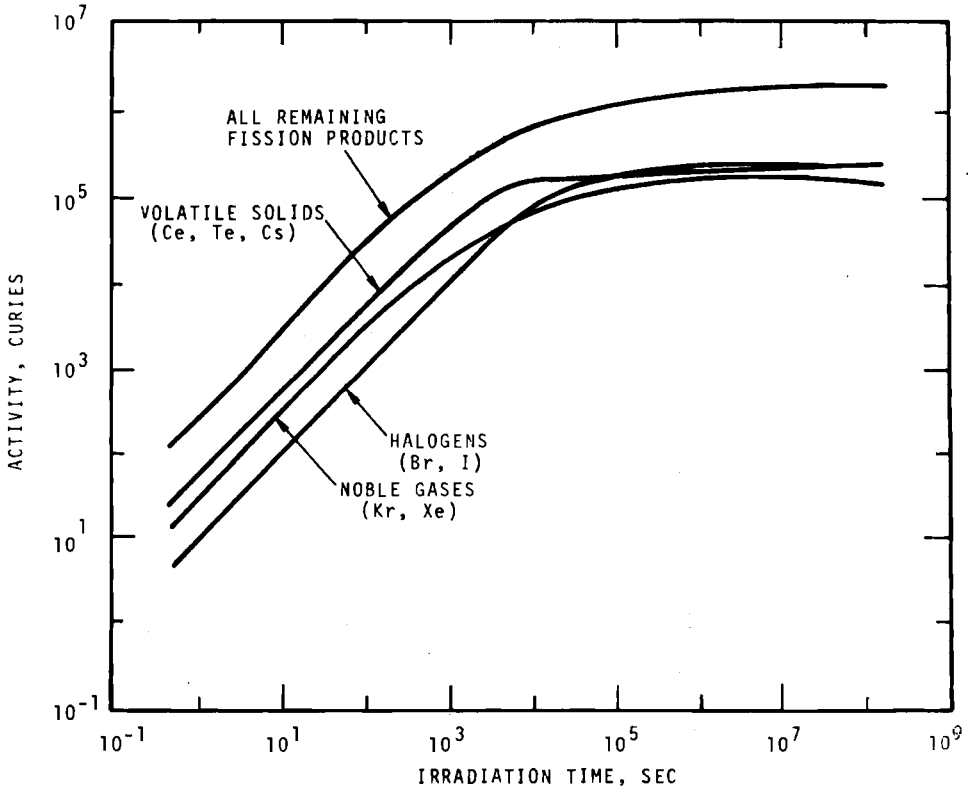

FIGURE 2. Distribution of Fission Products as a Function of Irradiation Time at 20 Minutes After Shutdown

The additional group, volatile solids (Se, Te, Cs) in this figure is a classification which is suggested by some experimental data available.[4] The relative activities of the noble gases and halogens are illustrated in Figures 3 and 4. Another phenomenon pertaining only to the noble gases, is that further depletion due to other removal mechanisms is unlikely. For these reasons then, a study was initiated to determine if the characteristics of radioactive noble gas inventories produced in nuclear fuels may be sufficiently unique to measure; thereby estimating atmospheric releases to the environment in the first few hours following fuel failure accidents.

NOBLE GAS INVENTORIES

The gross activity and isotopic composition of the noble gas fission products varies with power level and irradiation time. Table 1 summarizes the noble gas inventory estimates for selected irradiation times at 1 MW(th) as a function of decay time.

The inventory actually produced in nuclear fuel depends on the type of reactor and its proposed use. For this analysis, however, the differences between pressurized and boiling water reactors are ignored. Reactors are categorized here into one of three types, namely research, test and power reactors.

The noble gas isotopic composition in all three types would be very nearly the same after identical irradiation times, even though the quantity may be quite different. A fuel failure accident can occur at any time after start-up. Therefore, it is conceivable that an accidental release from a power reactor could be identical to that from a test or research reactor. In such an accident, a major difference in the noble gas composition could result when the release occurs in coincidence with a criticality excursion. The additional inventory produced in the excursion would be insignificant in the case of a power reactor operating at a power level of more than a few MW(th) and after more than a few hours of operation. The excursion inventory could be a significant fraction of the release, however, in the case of either a start-up accident in a power reactor with new fuel or an accident involving a research reactor. Maximum inventories estimated from Table 1 for each reactor class are listed in Table 2.

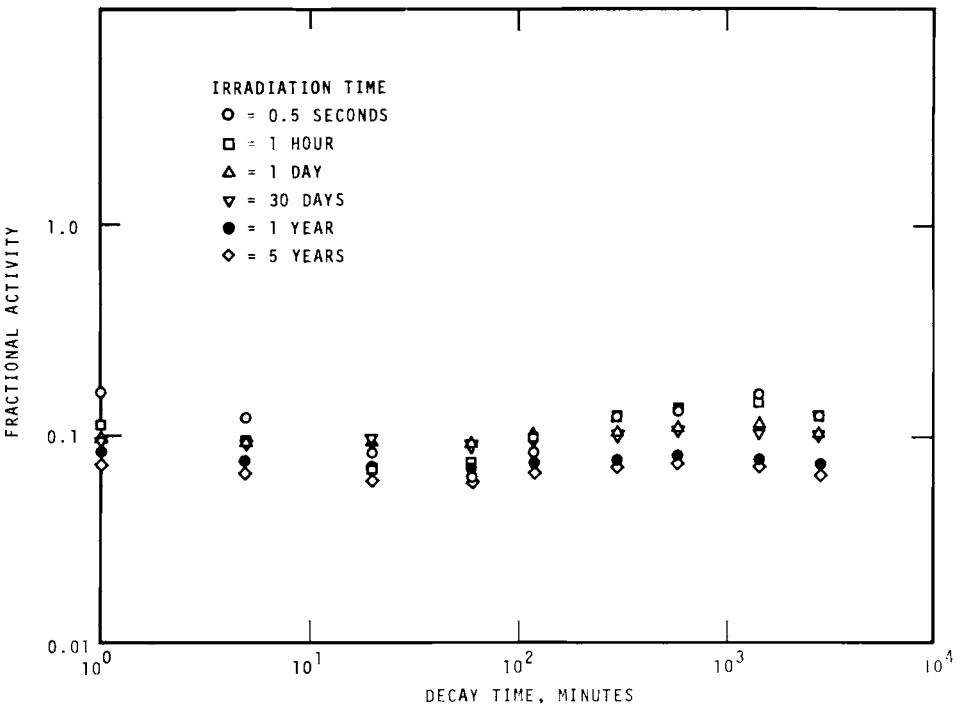

FIGURE 3. Effect of Irradiation Time on Fractional Activity of Noble Gases at Selected Decay Times

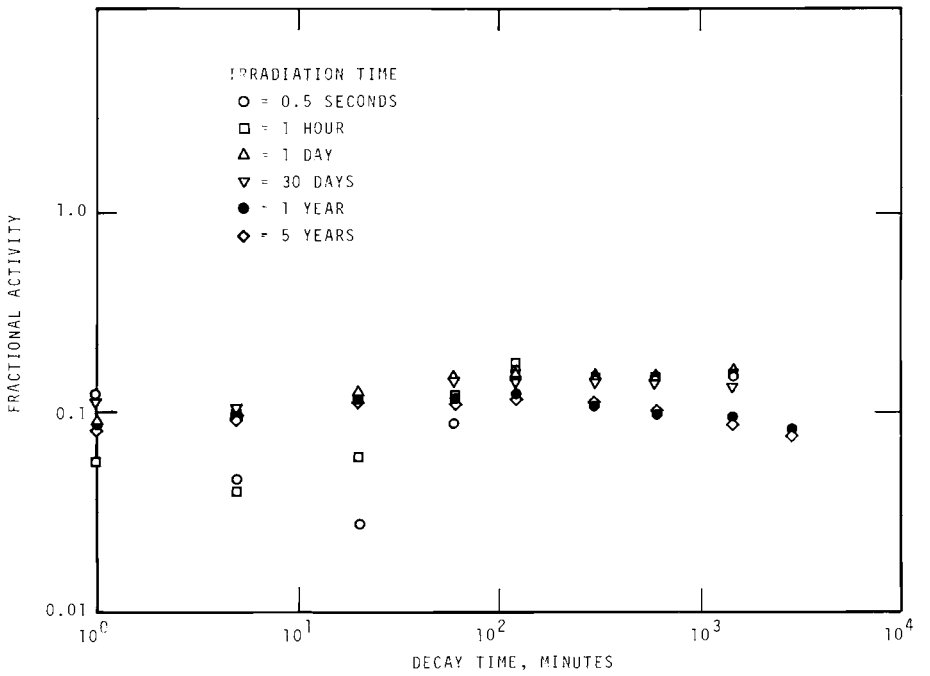

FIGURE 4. Effect of Irradiation Time on Fractional Activity of Halogens at Selected Decay Times

TABLE 1. Noble Gas Fission Products Inventory, Ci Fuel Irradiated at 1 MW for Time Indicated

Irradiation Time	Decay Time						
	2 min	10 min	1 hr	2 hr	5 hr	10 hr	1 day
0.5 s	2.1(2[a]	3.7(1	3.3(0	1.8(0	9.2(-1	5.1(-1	2.2-(1
30 s	1.1(4	2.1(3	2.0(2	1.1(2	5.5(1	3.0(1	1.3(1
5 m	5.9(4	1.6(4	1.9(3	1.0(3	5.5(2	3.0(2	1.3(2
1 h	1.3(5	5.9(4	1.7(4	1.1(4	6.1(3	3.5(3	1.5(3
10 h	1.9(5	1.2(5	6.6(4	5.3(4	3.5(4	2.3(4	1.1(4
1 d	2.1(5	1.3(5	8.1(4	6.7(4	4.8(4	3.4(4	1.8(4
10 d	2.5(5	1.7(5	1.2(5	1.1(5	8.7(4	7.2(4	5.3(4
100 d	2.6(5	1.9(5	1.4(5	1.2(5	1.0(5	9.0(4	7.0(4
1 y	2.5(5	1.8(5	1.3(5	1.2(5	1.0(5	9.0(4	7.1(4
2 y	2.4(5	1.7(5	1.2(5	1.1(5	1.0(5	9.1(4	7.2(4
5 y	2.3(5	1.6(5	1.2(5	1.1(5	1.0(5	9.3(4	7.4(4

(a) $2.1(2 \equiv 2.1 \times 10^2$

TABLE 2. Maximum Reactor Inventories of Noble Gases

Noble Gas Inventory in Ci, by Reactor Class:

Decay Time	Critical[a] Facility	Research[b]		Test[c]	Power[d]
		1[e]	2[e]		
2 min	1.3(4[f]	2.4(4	3.7(4	1.1(7	6.8(8
10 min	2.4(3	1.6(4	1.8(4	8.2(6	4.9(8
1 h	2.1(2	1.2(4	1.2(4	5.9(6	3.5(8
2 h	1.1(2	1.1(4	1.1(4	5.6(6	3.3(8
5 h	5.9(1	1.0(4	1.0(4	5.1(6	3.1(8
10 h	3.2(1	9.3(3	9.3(3	4.6(6	2.8(8
1 d	1.4(1	7.4(3	7.4(3	3.7(6	2.2(8

(a) 10^{18} fissions in 0.5 seconds
(b) Power level of 0.1 MW(th) for 2 years
(c) Power level of 50 MW(th) for 2 years
(d) Power level of 3000 MW(th) for 2 years
(e) Col 1 No Excursion
Col 2 Includes Excursion of 10^{18} fissions -0.5 sec
(f) $1.3(4 \equiv 1.3 \times 10^4$

NOBLE GAS GAMMA ENERGY SPECTRA

Individual gamma energies reported by Lederer et al.[5] for each fission product, have been incorporated in a library of the computer program ISOSHLD-III.[6,7] This library has been used to generate histograms of gamma energies from noble gas fission product inventories produced during each of eight arbitrarily selected irradiation times at eleven arbitrarily selected decay times. Photons emitted by the daughter products formed after shutdown, were included in the histograms. The irradiation times ranged from 0.5 second to five years, and the decay times ranged from zero out to one day. Figure 5 is illustrative of these histograms.

Although the energy spectra show no unique characteristics, a tabulation of peak values indicates that certain energy intervals may be useful for estimating the noble gas activity in a passing cloud. The frequency distribution of the energy at which the peak number of photons occurs is shown in Table 3.

SOURCE TERM DETERMINATION

There does not appear to be any unique characteristic which will permit direct measurement of the quantity of noble gas released to the atmosphere following a reactor accident. An indirect method of estimating the quantity of noble gases released has been investigated. The proposed method assumes that the noble gas mixture is a known fraction of the total fission products released. It is also necessary to assume that measurement of the number of photons within a specified energy interval, can be related to noble gas activity released. The proposed method is described in Appendix A.

CONCLUSIONS

No unique characteristics of reactor-produced fission products were found which could be used to determine the quantity of radionuclides released to the atmosphere following a postulated reactor accident. Although no unique characteristics are apparent, it may be feasible to measure the atmospheric release of fission products by gamma energy

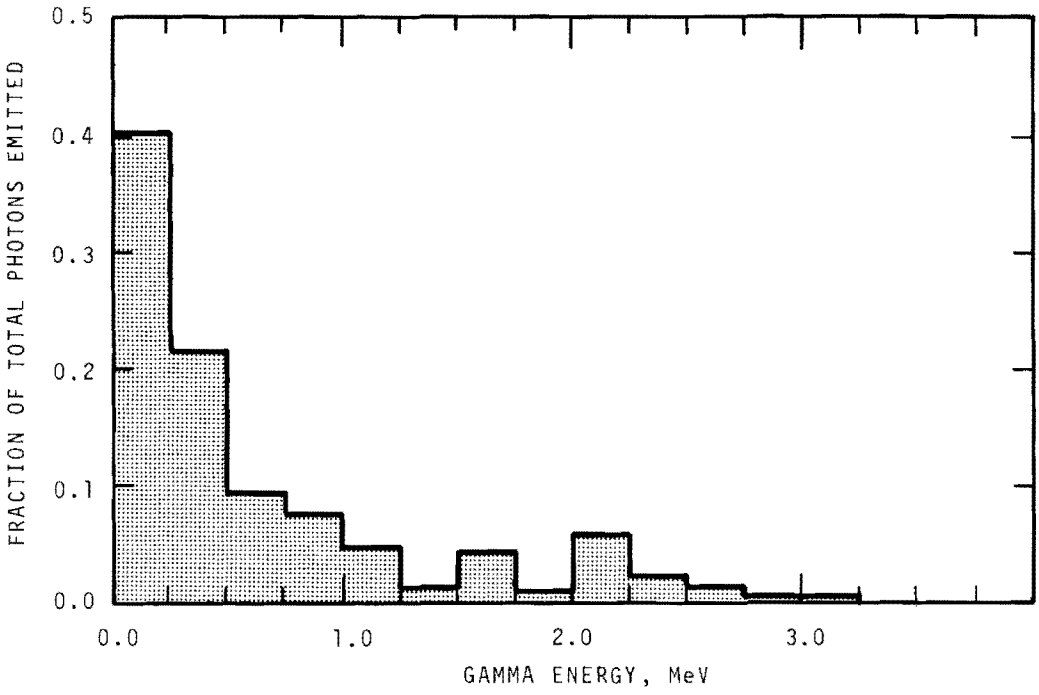

FIGURE 5. Gamma Photon Energy Distribution for Noble Gases Plus Daughters - at Shutdown After Two Years Irradiation

TABLE 3. Frequency of Peak Number of Photons Versus Energy

Irradiation Time	Energy in MeV of Peak Number										
	0.125	0.375	0.625	0.875	1.125	1.375	1.625	1.875	2.125	2.375	2.625
0.5s	-	-	-	2	2	6	-	1	-	-	-
30 s	-	-	2	1	2	3	1	-	1	1	-
5 m	-	-	1	-	-	2	3	-	3	2	-
1 h	1	-	-	-	-	-	-	-	7	2	1
10 h	1	-	-	-	-	-	-	-	6	4	-
3 d	2	-	-	-	-	-	-	-	6	3	-
2 y	3	-	-	-	-	-	-	-	6	2	-
5 y	3	-	-	-	-	-	-	-	7	1	-

analysis and measurement. One method of doing this is discussed in the appendix to this report. This method relies heavily on mathematical modeling and presumes a knowledge of the gamma energy spectrum and relative activity of the noble gases as a group. The method appears feasible; however, experimental verification is prerequisite to its further consideration.

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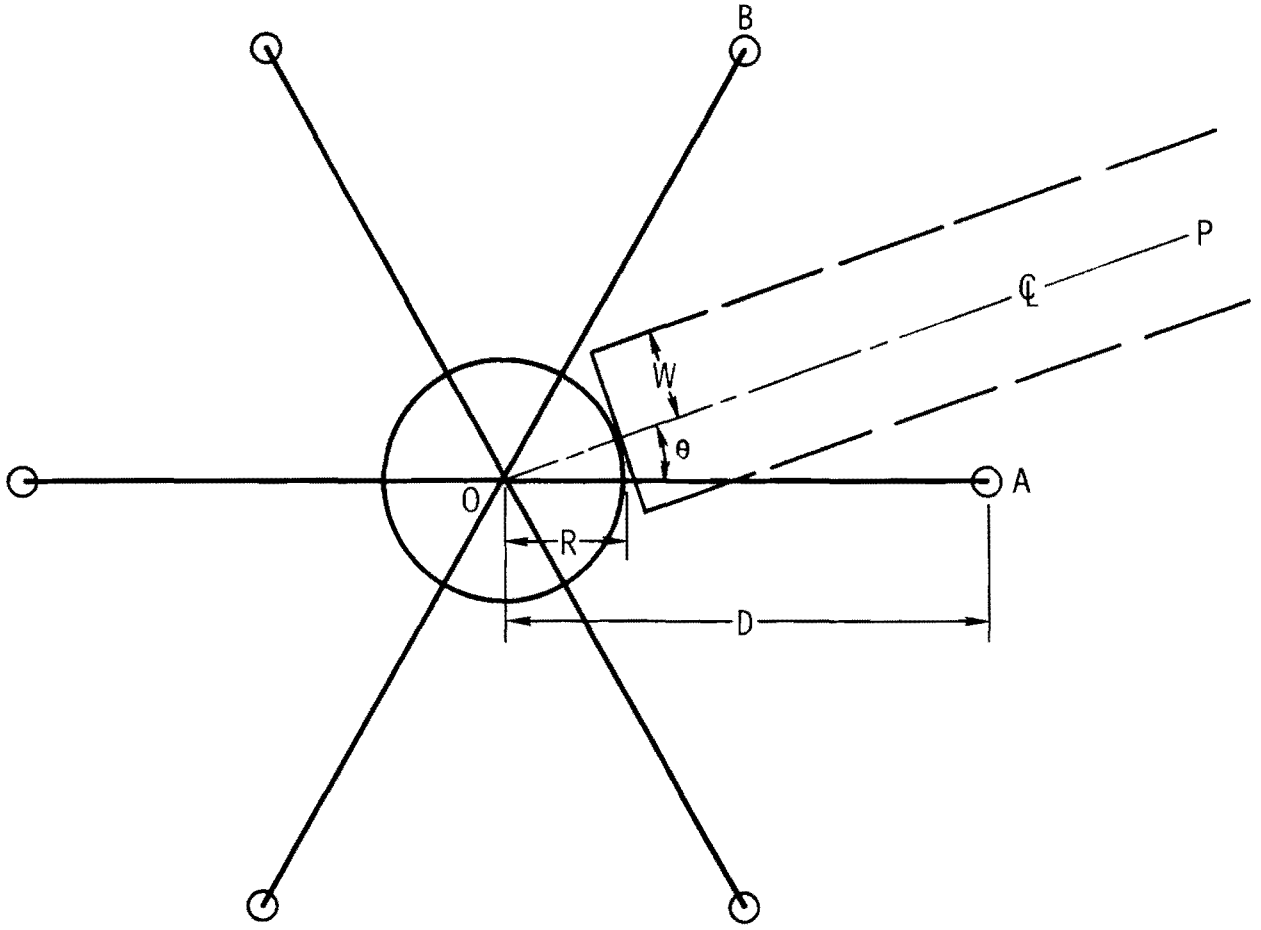
APPENDIX

This appendix describes a method for estimating the atmospheric release rate of radionuclides during the first few hours following a nuclear reactor accident. As indicated in the text, the majority of gamma radiation produced by released materials may be expected to come from noble gases and daughters of noble gases. Hence, the atmospheric release rate is determined as curies of noble gases, released per unit time.

The proposed method uses a system of gamma photon detectors to measure the photon flux at selected points. The photon flux is coupled with information on wind speed and direction and the operating history of the reactor to arrive at an estimate of noble gas release rate.

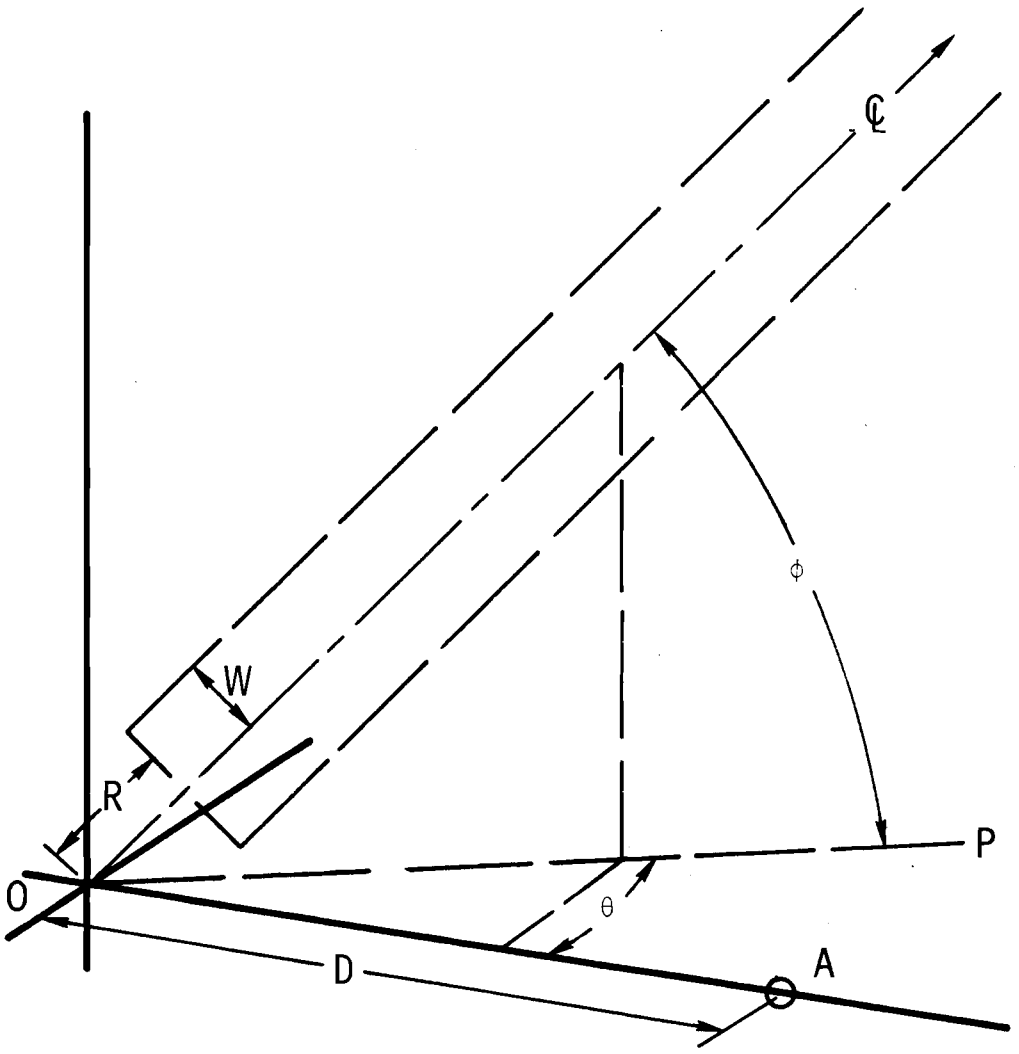
A sample system is described which serves to illustrate the general procedure. This system consists of six photon detectors at about ground level and 100 meters from the reactor. The detectors are spaced at 60° angles, as illustrated in Figure A-1. The path of the plume of released nuclides is indicated by line $\overline{OP}$ and the plume width is 2W. The angle θ is subtended by the plume centerline and the line to the nearest detector, $\overline{OA}$.

Under some conditions the plume from the reactor building may rise as it travels downwind, making an angle ϕ with the horizon. See Figure A-2 for a description of the plume rise geometry. The photon detectors are electronically biased in order to measure the flux of gamma radiation above 2 MeV. The 2 MeV minimum energy value is suggested by the data in Table 3 of the text. Estimates of the flux photons with energies greater than 1.99 MeV at the detector have been made with the computer code ISOSHLD. For these calculations the plume was represented as a cylinder of radius 20 meters and length 400 meters. The plume length was cut at 400 meters since additional length does not significantly increase the calculated value of flux at the detector. The plume source strength was set to 1 photon per second per meter of plume at an energy of 2.4 MeV. The plume was assumed to be radioactive noble gas fission products uniformly distributed in the radial direction. This is a



○ = DETECTOR
$\overline{OP}$ = CLOUD CENTER LINE
R = SOURCE RADIUS
D = DISTANCE TO DETECTOR
W = RADIUS OF PLUME
θ = ANGLE OF PLUME CENTERLINE WITH DETECTOR DIRECTION

FIGURE A-1. Source-Detector Geometry



R = SOURCE RADIUS
D = DISTANCE TO DETECTOR
W = RADIUS OF PLUME
θ = ANGLE OF PLUME CENTERLINE WITH DETECTOR DIRECTION
ϕ = VERTICAL ANGLE OF PLUME CENTERLINE WITH HORIZONTAL PLANE

FIGURE A-2. Plume Rise Geometry

reasonable assumption in light of the building wake mixing effect and the short distance to the detectors (100 meters). The resulting fluxes are presented as a function of θ and ϕ in Figure A-3. The plume width W does not have a great effect on flux at angles greater than 10° as indicated by Figure A-4.

The ratio of the fluxes from two detectors on either side of the plume can be used to help determine the plume position. The flux ratio of the near detector (at angle θ with the plume) to the far detector (at 60 - θ) is illustrated in Figure A-5. The ratio is presented at a function of θ and ϕ. At an angle of 30° the flux ratio is 1 since the plume is midway between the detectors. To estimate the plume rise angle ϕ for θ near 30°, the flux ratio for the near detector (at θ) to the next farthest detector (at 120 - θ) may be used. These ratios are shown in Figure A-6.

From Figures A-3, A-5 and A-6, and measured values of wind direction, the expected normalized photon flux at the detector, ψ, may be determined. The units of normalized flux are photons/cm^2/sec per photon/sec per meter of plume in the downwind direction. The normalized flux may be used to estimate the actual photon emmission rate from nuclides in the cloud from the relation:

$$P_\gamma = \frac{D}{\psi} \overline{U}$$

where P_γ is the high energy photon emmission rate (i.e. for photons of energy >2 MeV) in photons/sec per second of release time, D is the measured high energy photon flux at the near detector and $\overline{U}$ is the average wind speed.

The conversion from photon emmission rate to activity, in curies for noble gases depends on the operating history of the reactor. Figure A-7 is a plot of conversion factors, I, as a function of operating time and decay time after shutdown. The release rate Q', is then determined by:

$$Q' \text{ (N.G.)} = IP_\gamma$$

In summary, the proposed method estimates the release rate in curies per second, of noble gases using measured values of wind speed, wind direction, gamma photon flux and a knowledge of fuel irradiation history.

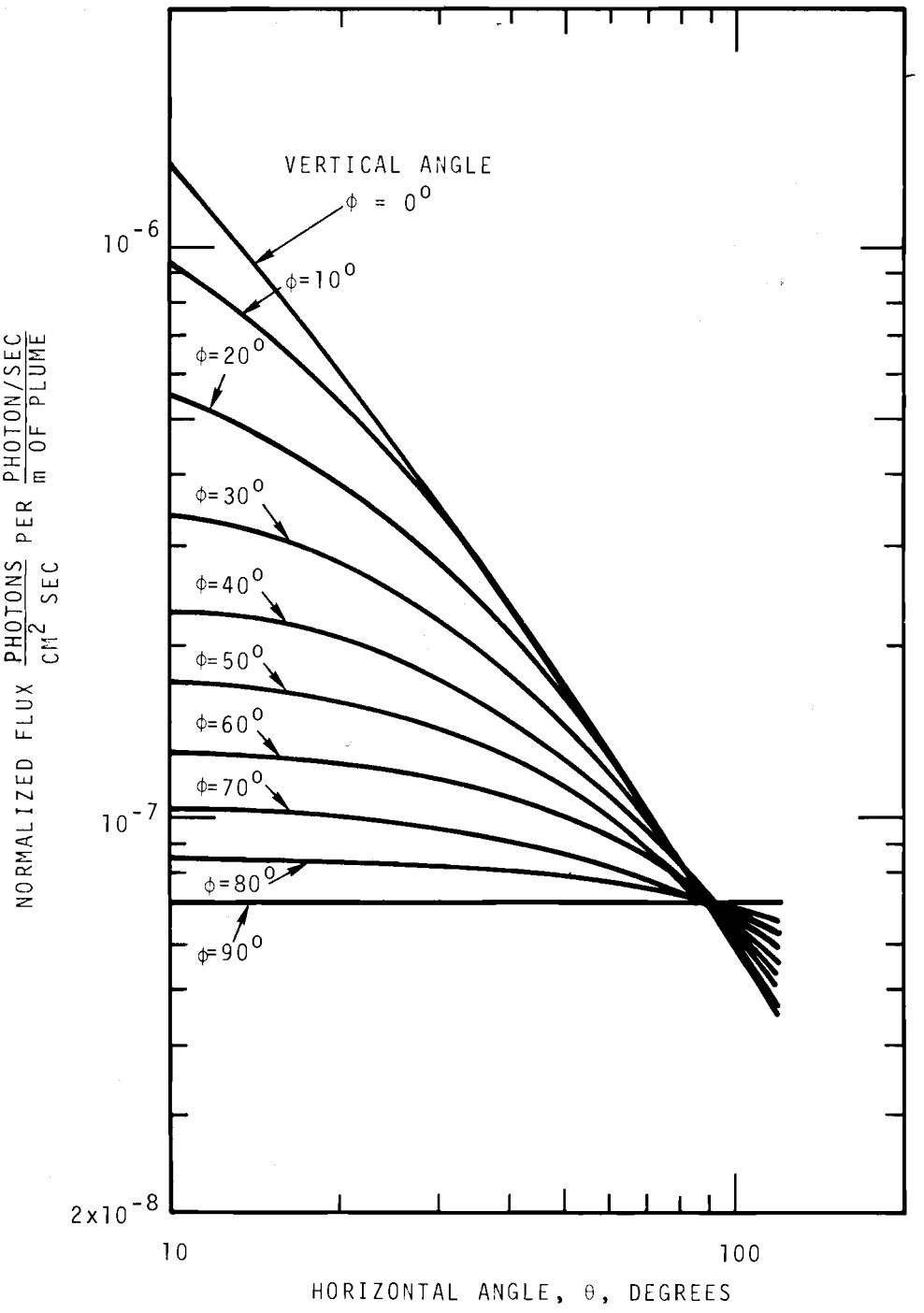


FIGURE A-3. Normalized High Energy Photon Flux as a Function of θ and ϕ.

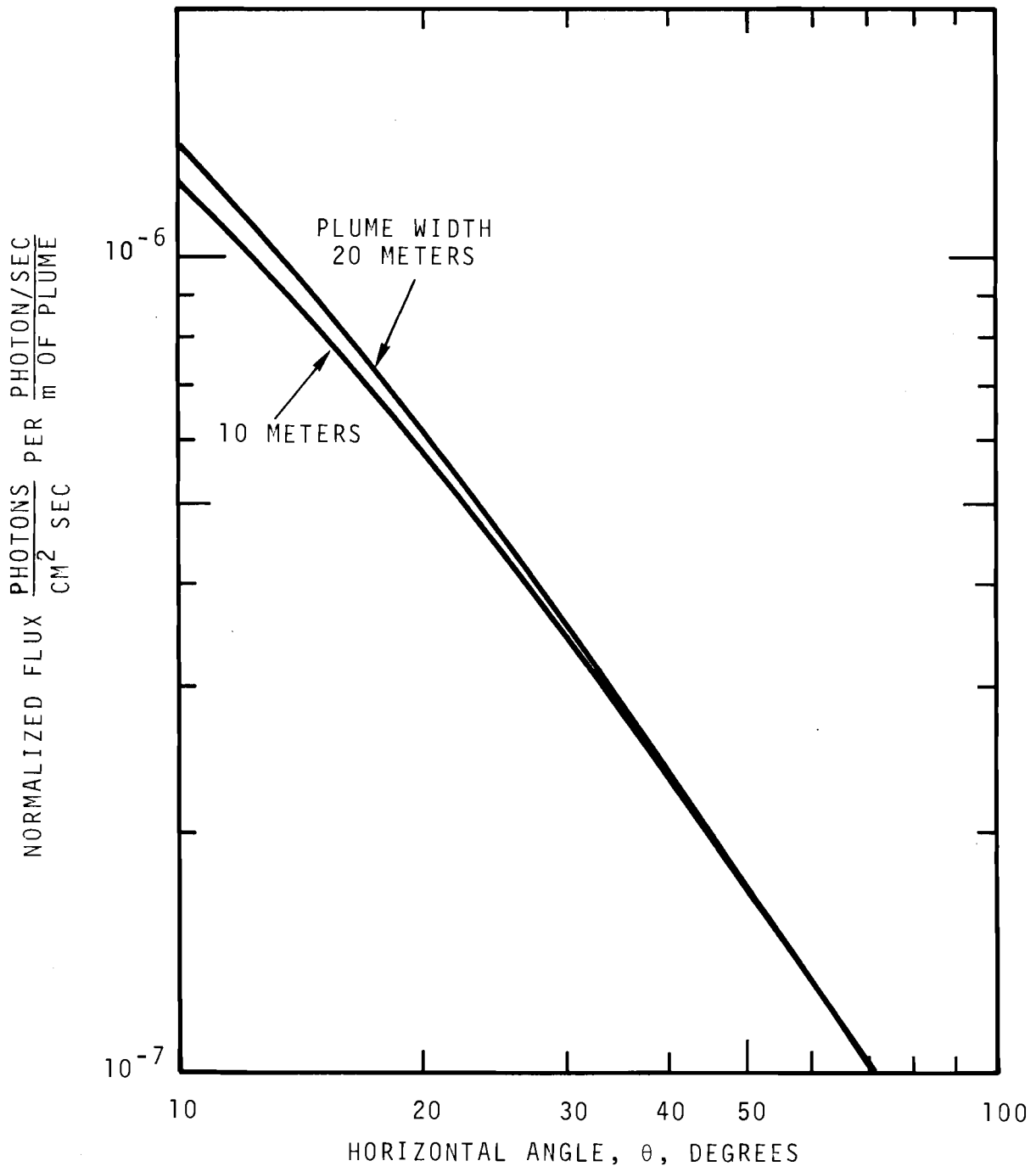


FIGURE A-4. Effect of Plume Width on Normalized High Energy Photon Flux

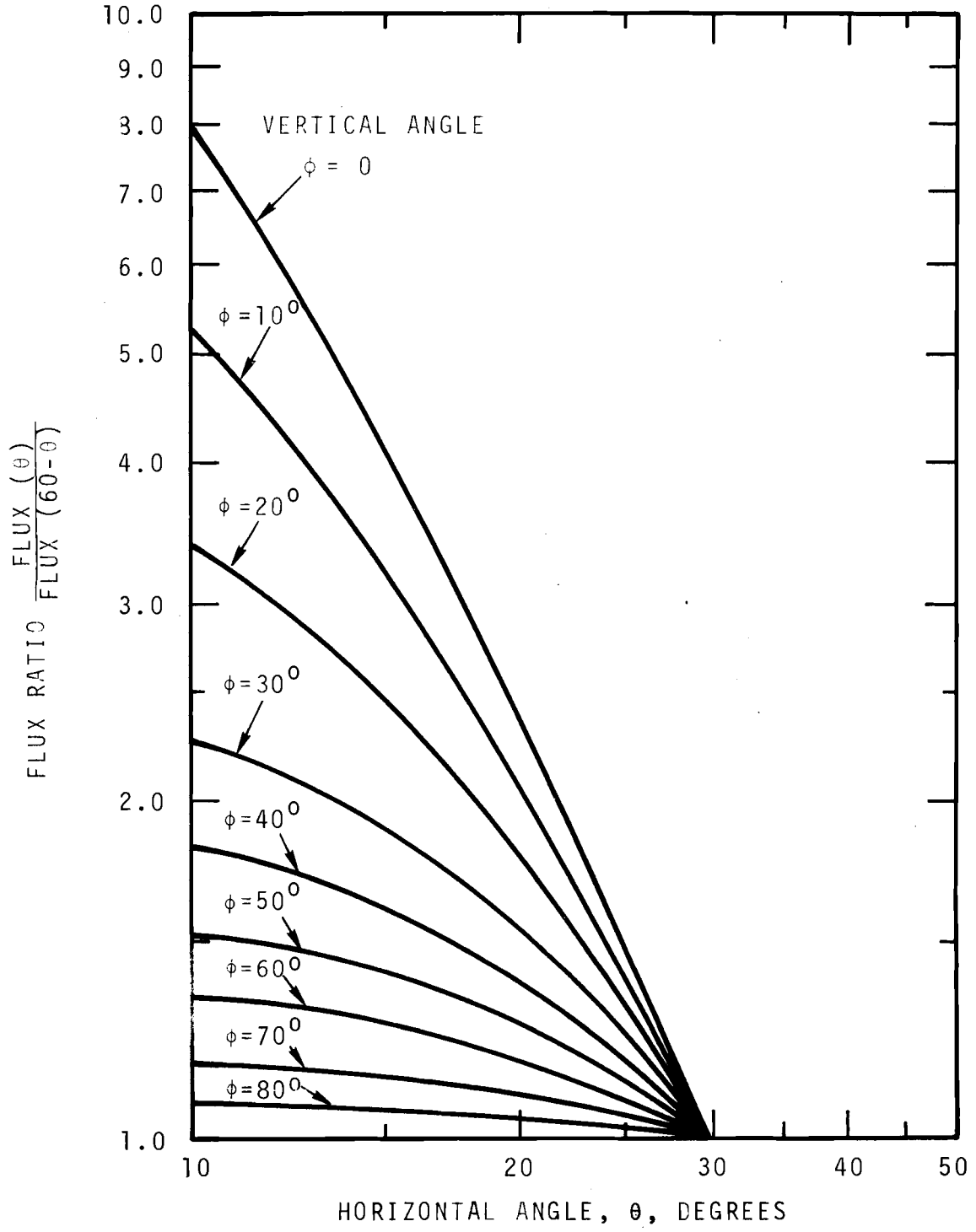


FIGURE A-5. High Energy Photon Flux Ratio as a Function of Plume Direction for Detectors Separated by 60°

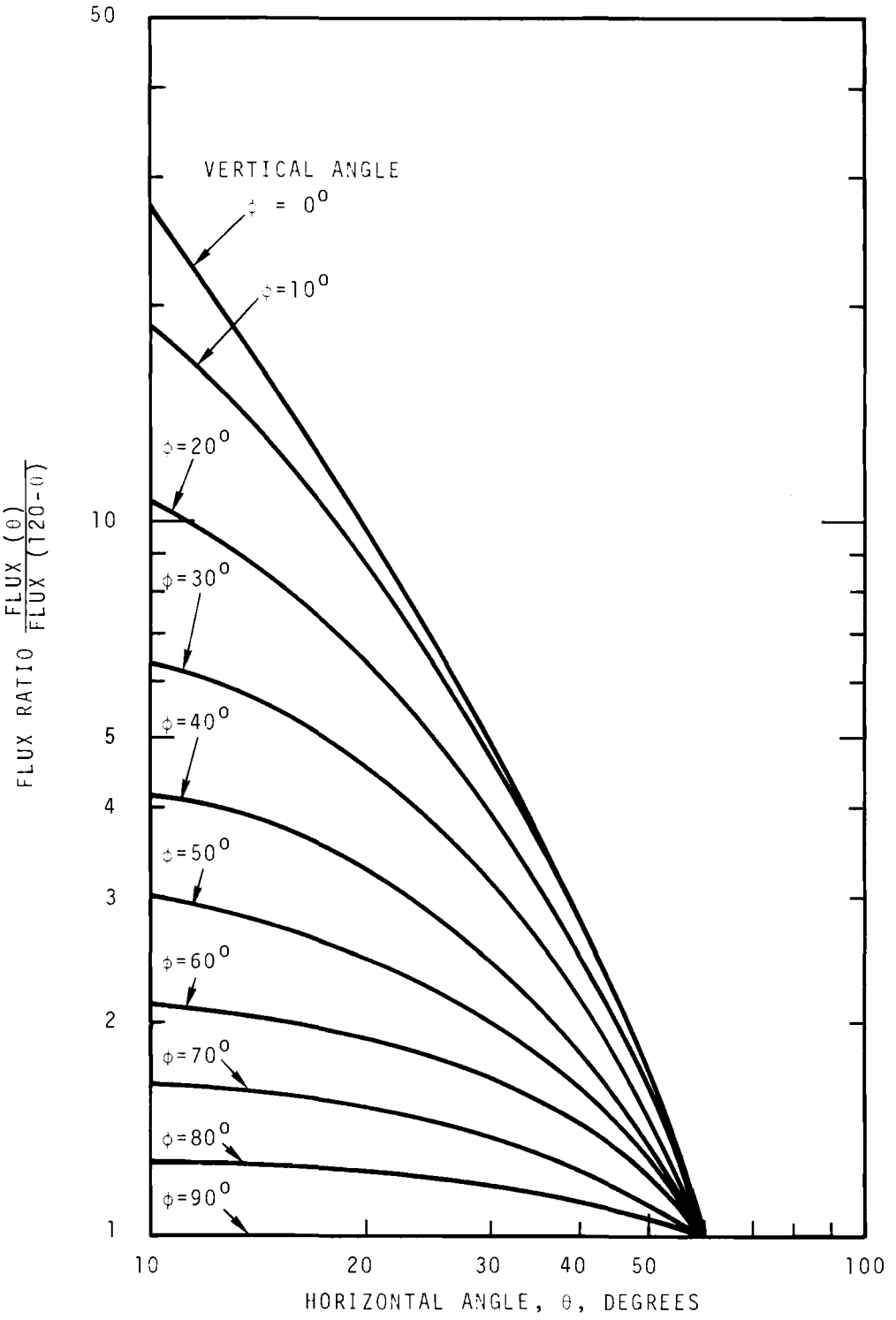


FIGURE A-6. High Energy Photon Flux Ratio as a Function of Plume Direction for Detectors Separated by 120°

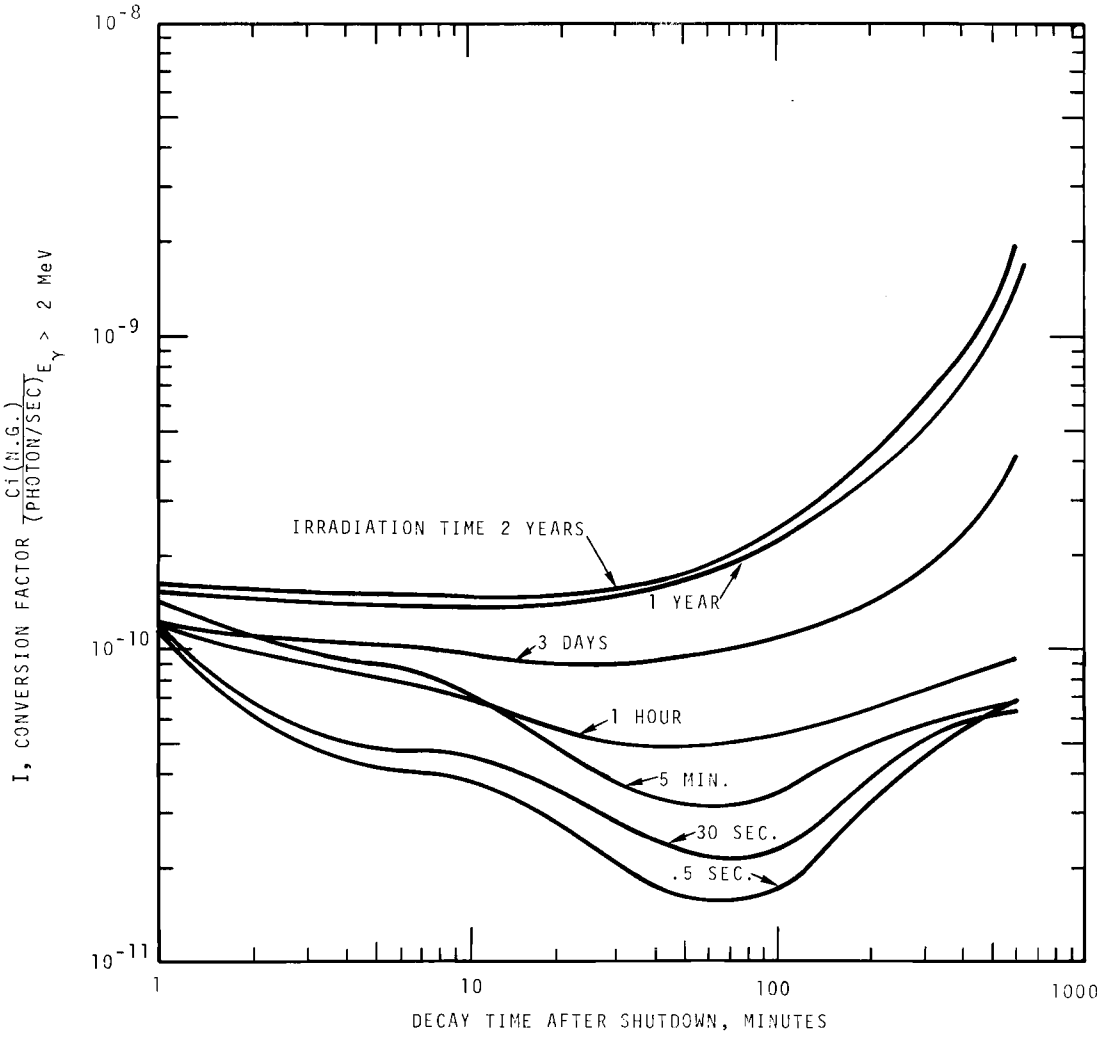


FIGURE A-7. High Energy Photon Emmission Rate from Noble Gases as a Function of Fuel Irradiation Time

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