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**Analysis of Underground**  
**Weapon Test Effluent Samples**

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**Analysis of Underground  
Weapon Test Effluent Samples**

by

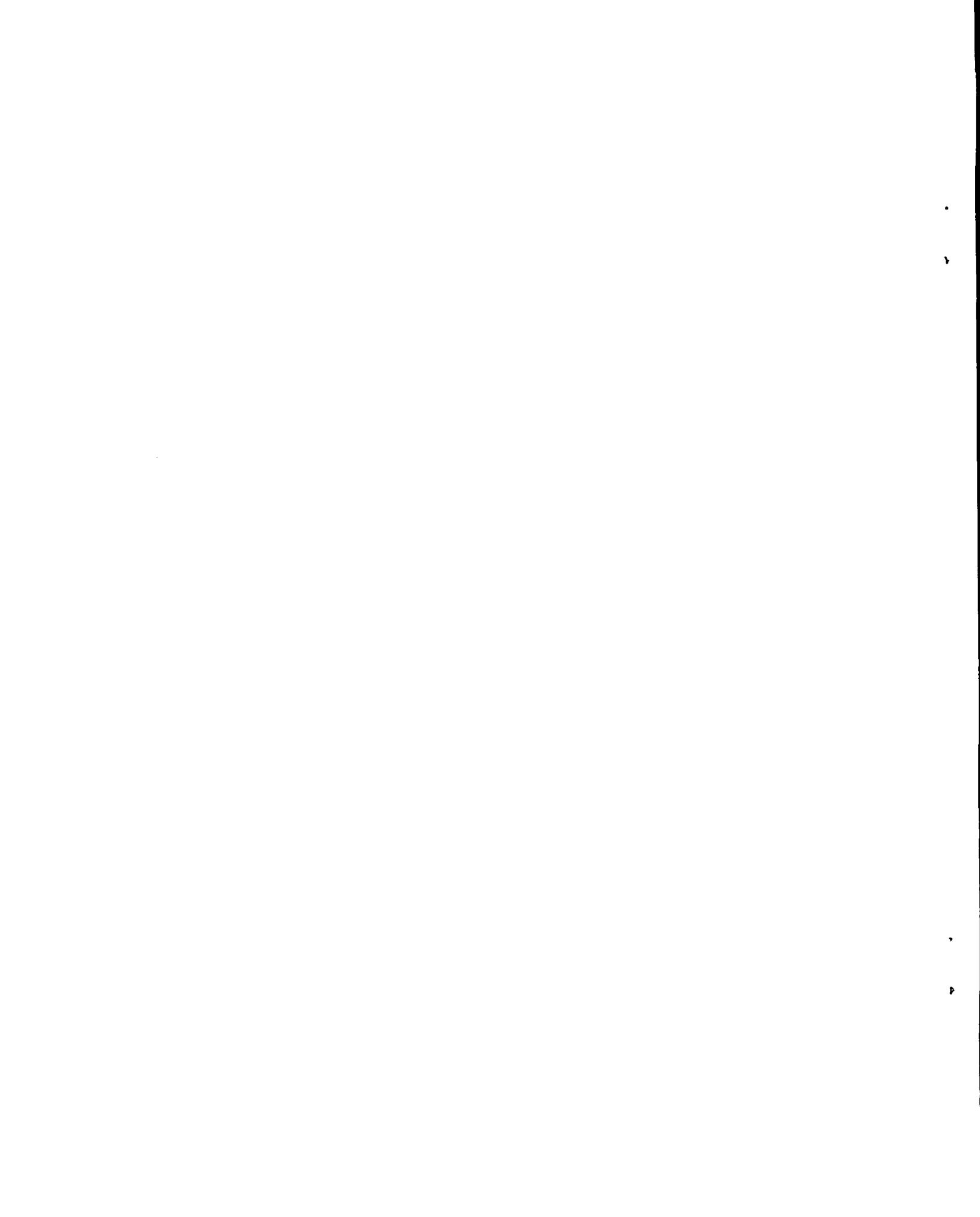
R. W. Henderson





### ABSTRACT

In the course of underground weapons testing inadvertent releases of material to the atmosphere may occur. The composition of this material varies depending on the age of the fission products at the time of the release and the status of the operation. This report gives the methods and procedures used by Group H-8 to derive the isotopic composition of the released material.



## INTRODUCTION

In the course of nuclear weapon underground testing an occasional accidental release of radioactivity is experienced either at the time of detonation or during the drillback operations to obtain samples for radiochemical analyses. For purposes of documentation, it is necessary for the Laboratory to establish an estimate of the total activity and the major constituents of the effluent.

The procedure that has been evolved by LASL Field Studies Group (H-8) for deriving the estimate of total activity released is detailed in LA-3094, "Source Measurement: Determining the Release from a Point Source by Remotely Located Samplers," by R. W. Henderson.

This report outlines the method used to establish the composition of the effluent.

## SAMPLING

To date the most feasible method of obtaining samples of effluent from any possible release over the entire time period from detonation to the sealing of the drillback holes has been to circle the point of possible release, i. e., ground zero or the drilling point, with an array of air samplers. Ideally the air sampler would collect an unfractionated sample of the effluent, and the analysis of the sample would be completed immediately after collection.

In practice this is not possible without an undue expenditure of effort and money. The compromise effected has been to sample the effluent with a Whatman 41 particulate prefilter and activated charcoal cartridge (MSA BM 2306 canister) at ambient temperature. Samples are evaluated

as soon as possible after a known release or after a normal shift change. In either case a minimum of 4 to 6 hr is required to obtain a first count on the samples.

### SAMPLE EVALUATION

In general the isotopic composition of the material collected by the air samplers is determined by a stripping procedure on a gamma activity decay curve, using gamma ray spectral data obtained from a 400 channel analyzer.

In addition studies of weapon test effluent collected on filter paper and charcoal traps at dry ice temperature have established quantitative relationships, applicable in the majority of cases, between the iodine and xenon in the effluent. Xenon is collected with essentially 100% efficiency by charcoal at dry ice temperature but is collected with an unknown but low and variable efficiency at ambient temperatures. These same studies established that the radioactive iodines normally present in released effluents are collected with an efficiency of 99+% at ambient temperatures.

Counting of the ambient temperature samples is, however, complicated by the presence of xenon. Heating and air washing of the charcoal cartridges are two methods that have been developed for removing the xenon without unduly influencing the iodine contents. Air washing the cartridge with about  $0.5 \text{ M}^3/\text{min}$  of clean air at temperatures greater than  $80^\circ\text{F}$  for an hour or simply maintaining the cartridge at  $90^\circ\text{F}$  for 45 min has proven to be effective in reducing the xenon content to below its interference level.

Thus, the analysis of the ambient air samples, together with the information generated by more elaborate sampling procedures on selected events, makes it possible to establish a reasonable quantitative estimate of the effluent composition.

Our experience, to date, has been that the majority of the releases are characterized by effluents that are very similar in composition. The exceptions to this typical effluent have resulted from (1) significant venting or (2) a slow release at early time.

In the first case, the composition is essentially the theoretical gross fission product mixture. All extrapolations and composition estimates are based on this assumption. In the second case, a small release at early times can produce an effluent containing only noble gases and their daughters. In this case all that can be done is to infer the quantity of the noble gas released by the measured amount of a particulate daughter. In general this involves inferring quantities of  $^{138}\text{Xe}$  from measured  $^{138}\text{Cs}$  and, if samples can be collected quickly enough,  $^{88}\text{Kr}$  from measured  $^{88}\text{Rb}$ . For this case it is assumed that only the noble gases are released and that the production of the daughter is initiated at time of release. If this time of release is known, either by visual observation or readings from gamma probes surrounding ground zero, it is used as the reference time for calculations. In the absence of such data, the release is assumed to be an instantaneous release at the midpoint of the sampling period.

Calculations leading to an estimate of the quantity of noble gas that would have been collected by a perfect sampler are based on the amount of particulate daughter collected, decay since time of release, and the ratio of particulate daughter to noble gas at the sampling point assuming production of the daughter occurred in the period required for the cloud to travel from the release point to the sampling point. The results of these calculations for the individual samples are used as detailed in LA-3094 to estimate the total release.

## RESULTS OF DATA EVALUATION

### a. Iodine-Xenon Releases

Ambient temperature charcoal trap samples have been analyzed from at least six events involving approximately 20 sets of samples when radioiodine was released to the atmosphere. Samples were collected as early as 142 to 532 min post event to a few weeks post event. These data show that the radioiodine composition, on a gamma activity basis, may be expressed as follows when related to zero time:

#### Iodine fraction of cloud

$^{131}\text{I}$	1
$^{133}\text{I}$	105
$^{135}\text{I}$	1360

The analysis of samples collected by "cold traps" (charcoal at dry ice temperature) on approximately six events produced a set of consistent data when the radio xenons were measured, extrapolated to zero time exponentially and related to the  $^{131}\text{I}$  in the sample. These samples were collected from the vent line during drilling operations. These samples could be collected only as early as one day post event and extrapolations to earlier times give only an upper limit for the xenon fraction of the effluent. The ratios found are as follows:

#### Xenon to $^{131}\text{I}$ ratios expressed at zero time

$^{135}\text{Xe}/^{131}\text{I}$	$5 \times 10^4$
$^{133}\text{Xe}/^{131}\text{I}$	$1 \times 10^4$

In the data above no correction has been made for gamma yield, and the ratios given are for gamma curies ( $3.7 \times 10^{10}$  gammas per second).

This simplification in data reduction appears to do no worse than slightly overestimate the contribution of  $^{135}\text{I}$  on a beta activity basis.

The calculations and following figures are designed to aid in the evaluation of releases which may be classed as slow leaks (material seeping from the ground at some time following zero time) or a failure of the drillback containment system.

Figures 1 through 4 are generated by the data given above. Figure 1 shows the total gamma activity of a xenon-iodine cloud as a function of time and the radioiodine fraction. Figure 2 shows the iodine fraction only. This curve represents the decay of the ambient charcoal traps as well as the iodine composition of the cloud at any time, for release times beyond 300 min. Figure 3 shows the relative abundance of the various isotopes as a function of time, and Figure 4 presents the ratios of the xenons to  $^{131}\text{I}$ . These curves are used to derive an estimate of the release of xenon and iodine from an event from the data generated by the ambient charcoal traps.

An attempt was made by this group to do whole-air sampling using evacuated flasks, but the concentrations encountered were too low for evaluation by existing equipment. A flow-through system was also devised to try to determine the composition of the effluent, but the predominance of the xenons made this system unworkable using existing equipment.

It would be desirable to obtain data pertinent to the noble gas fraction of the effluent material at early times (inside one day) and to devise a simple system for checking the xenon to iodine ratios on each event.

#### b. Early Time Noble Gas Releases

Low level early time releases have shown themselves to be composed largely of the noble gases  $^{138}\text{Xe}$  and  $^{88}\text{Kr}$ . These two isotopes can not

be measured directly by the existing equipment. However, the produced  $^{138}\text{Cs}$  and  $^{88}\text{Rb}$  can be sampled and measured under proper conditions. In these cases it is assumed that the gaseous parent is released free of the particulate daughter and that the daughter grows in as the cloud passes from the release point to the sampler. This growth time may be calculated from the wind speed and the distance from the release point to the sampler. Using this time and the assumption above, it is possible to calculate the amount of the gaseous parent released from the amount of particulate daughter measured.

The calculations used for the reduction of measured  $^{138}\text{Cs}$  illustrate those used for the calculation of both types of noble gas releases.

Assuming that no  $^{138}\text{Cs}$  is released and  $T_1$  is the time between release and sampling we have:

$$^{138}\text{Cs}(T_1) = ^{138}\text{Xe}(0) \frac{\lambda_{\text{Xe}}}{\lambda_{\text{Cs}} - \lambda_{\text{Xe}}} \left[ \exp(-\lambda_{\text{Xe}} T_1) - \exp(-\lambda_{\text{Cs}} T_1) \right]$$

It is now useful to calculate an amount of  $^{138}\text{Cs}$  at release time which could produce the same amount of  $^{138}\text{Cs}$  at sampling time. This is given by:

$$\begin{aligned} ^{138}\text{Cs}_A(0) &= ^{138}\text{Cs}(T_1) \exp(+\lambda_{\text{Cs}} T_1) \\ &= ^{138}\text{Xe}(0) \frac{\lambda_{\text{Xe}}}{\lambda_{\text{Cs}} - \lambda_{\text{Xe}}} \left[ \exp(\lambda_{\text{Cs}} - \lambda_{\text{Xe}}) T_1 - 1 \right] \end{aligned}$$

We may now calculate the ratio of  $^{138}\text{Xe}$  at release time to the apparent amount of  $^{138}\text{Cs}$  at release time as a function of  $T_1$ :

$$\frac{{}^{138}\text{Xe}(0)}{{}^{138}\text{Cs}_A(0)} = \frac{{}^{138}\text{Xe}(0)}{{}^{138}\text{Xe}(0) \frac{\lambda_{\text{Xe}}}{\lambda_{\text{Cs}} - \lambda_{\text{Xe}}} \left[ \exp(\lambda_{\text{Cs}} - \lambda_{\text{Xe}}) T_1 - 1 \right]}$$

$$\frac{{}^{138}\text{Xe}(0)}{{}^{138}\text{Cs}_A(0)} = \frac{\lambda_{\text{Cs}} - \lambda_{\text{Xe}}}{\lambda_{\text{Xe}} \left[ \exp(\lambda_{\text{Cs}} - \lambda_{\text{Xe}}) T_1 - 1 \right]}$$

For ease of use  $T_1$  can be expressed in terms of sample distance over wind speed. This has been done in the curves shown in Figures 5 through 8. Further, it is most convenient to use activity and we have:

$$\frac{A_{\text{Xe}}(0)}{A_{\text{Cs}}(0)} = \frac{{}^{138}\text{Xe}(0) \lambda_{\text{Xe}}}{{}^{138}\text{Cs}_A(0) \lambda_{\text{Cs}}} = \frac{\lambda_{\text{Cs}} - \lambda_{\text{Xe}}}{\lambda_{\text{Cs}} \left[ \exp(\lambda_{\text{Cs}} - \lambda_{\text{Xe}}) T_1 - 1 \right]}$$

Figures 5 through 8 show the results of these calculations. These curves show the ratio of the parent to the extrapolated apparent quantity of the daughter as a function of sampler distance over wind speed. Using these curves the amount of the daughter is measured and extrapolated to release time exponentially. The value from the curve then yields the amount of the parent released. Figures 5 and 6 are used for the calculation of  ${}^{138}\text{Xe}$  and Figures 7 and 8 are for  ${}^{88}\text{Kr}$ . The results of these calculations are then used in conjunction with LA-3094 to estimate the total release.

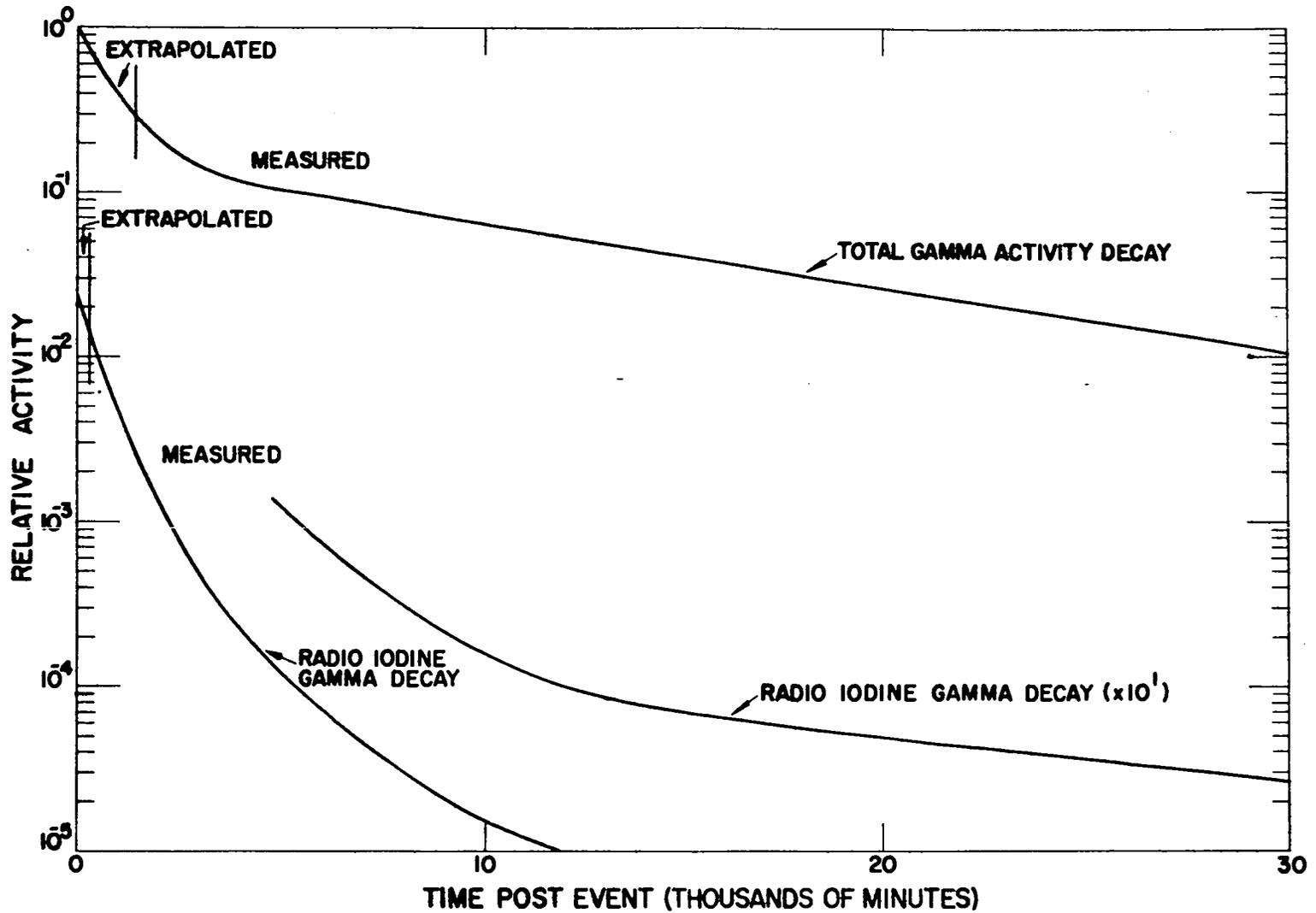


Fig. 1. Total gamma activity as a function of time.

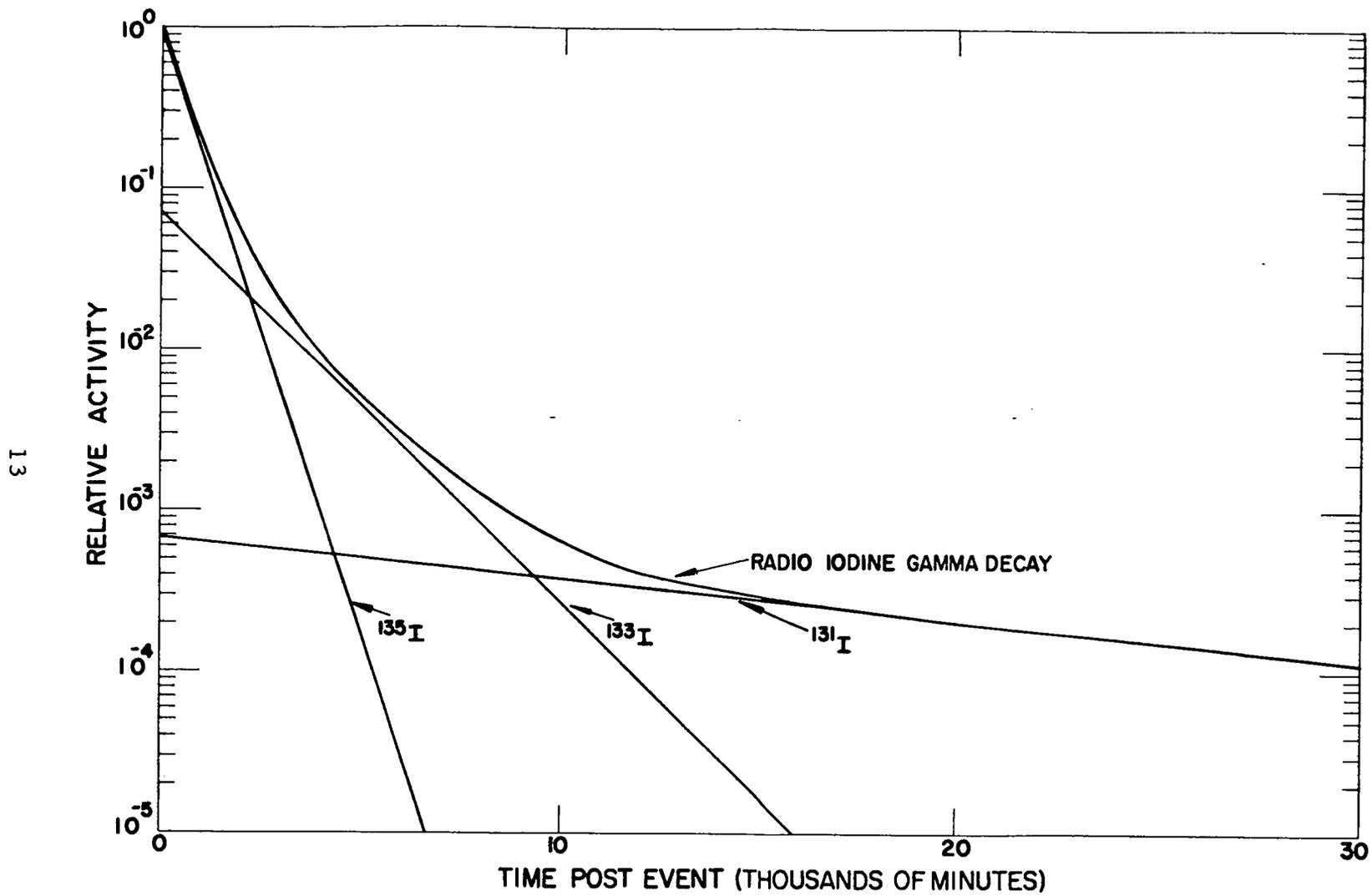


Fig. 2. Radioiodine fraction as a function of time.

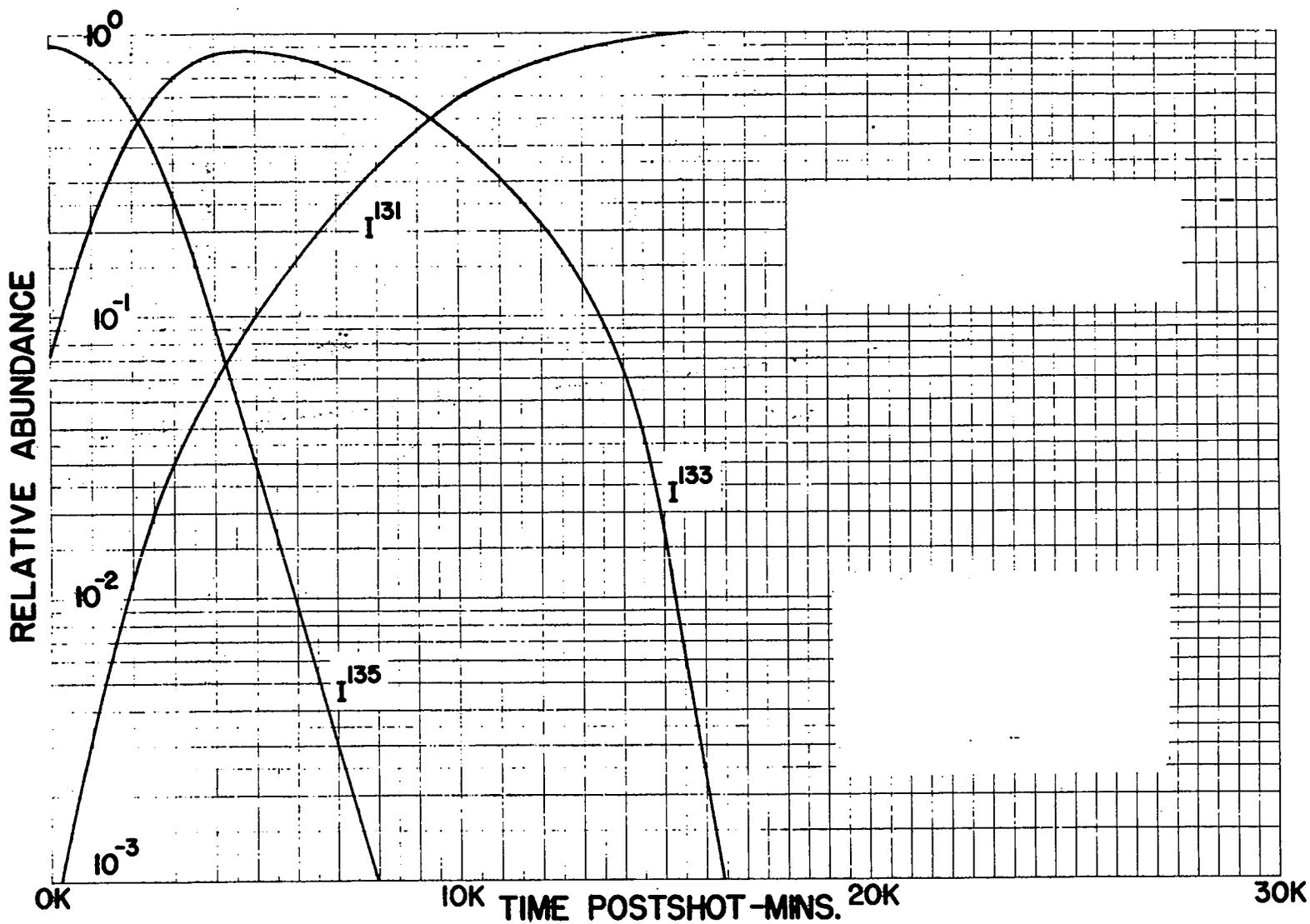


Fig. 3. Relative abundance of the iodine isotopes as a function of time.

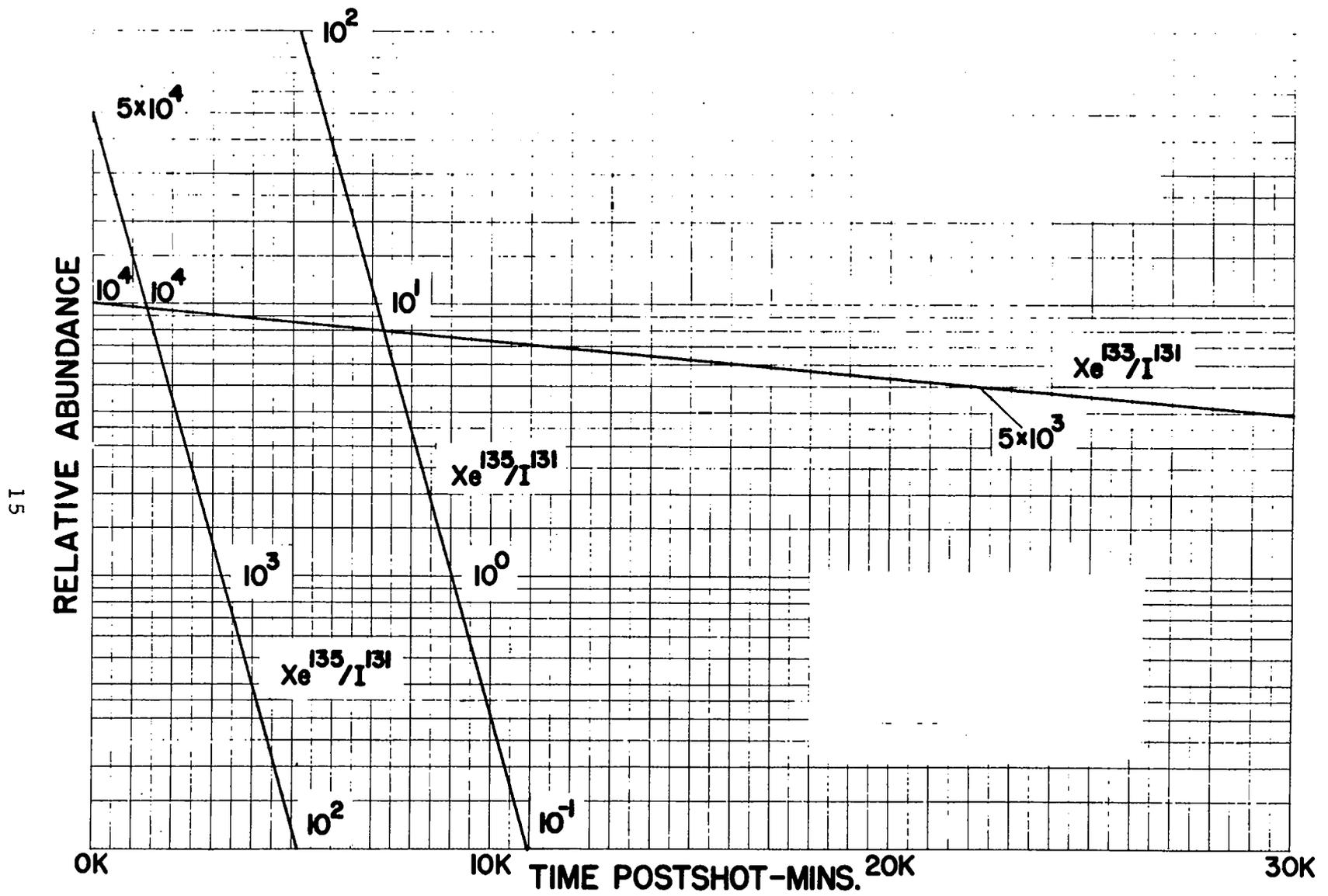


Fig. 4. Ratio of the xenons to  $^{131}\text{I}$ .

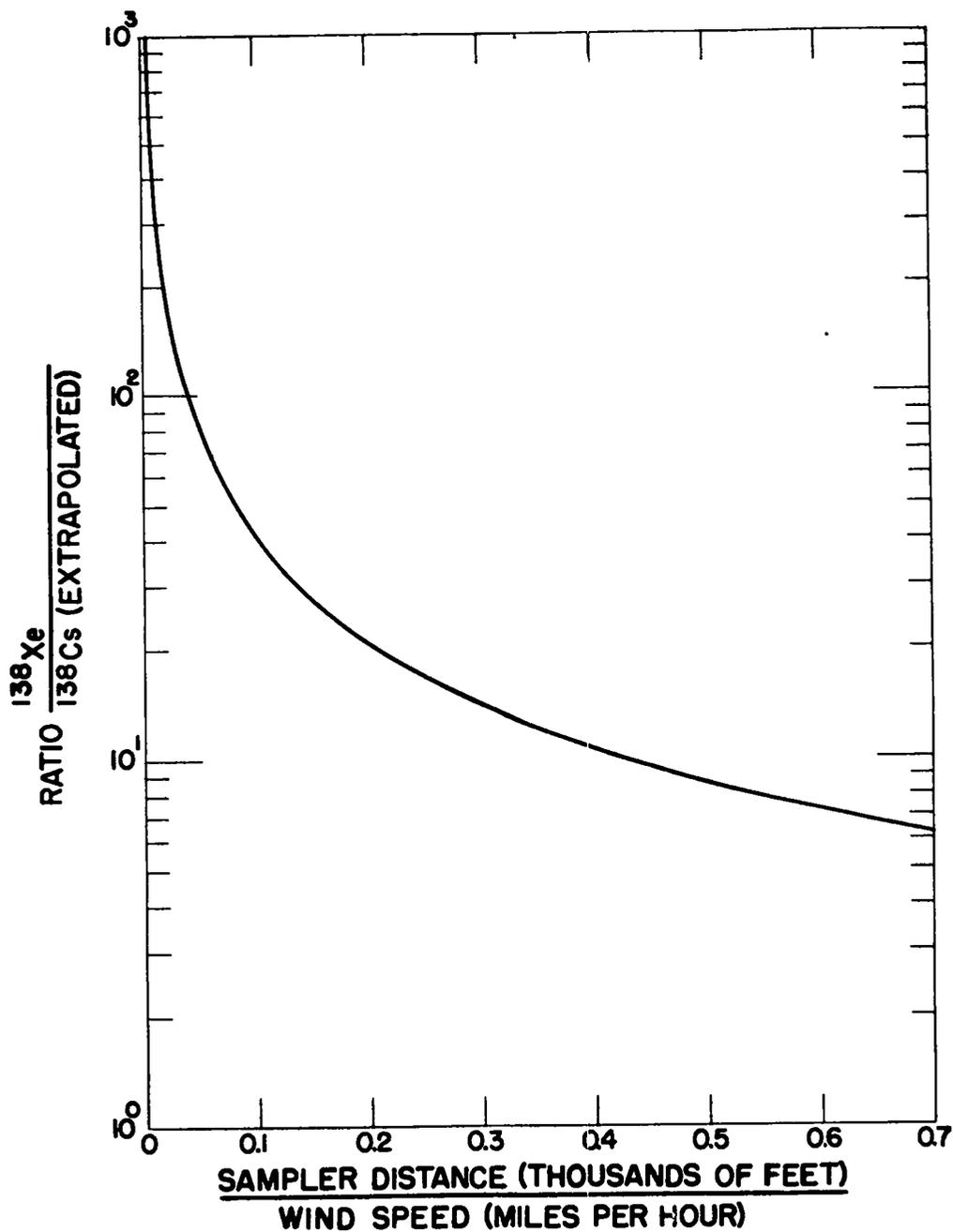


Fig. 5. Ratio of  $^{138}\text{Xe}$  to extrapolated  $^{138}\text{Cs}$ .

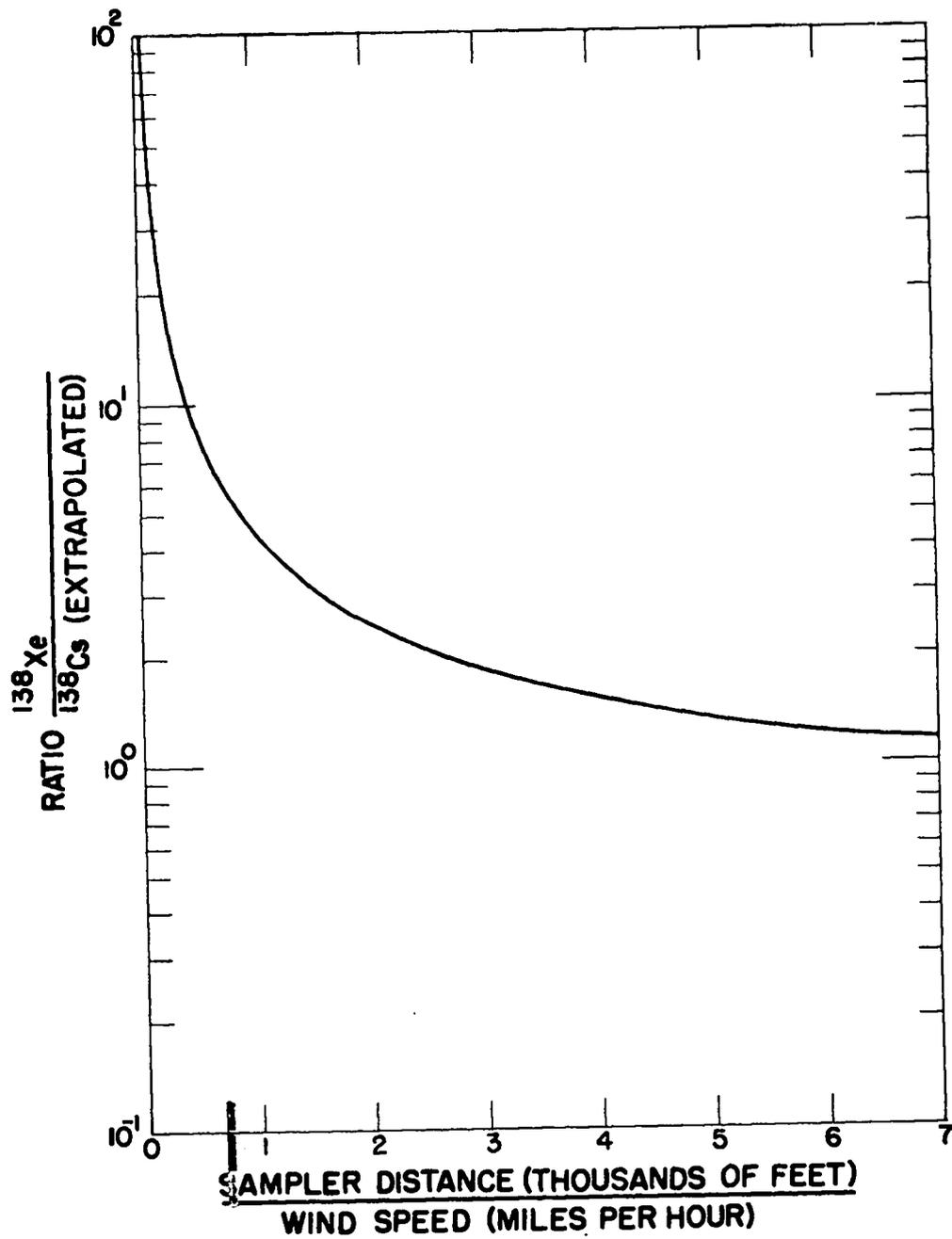


Fig. 6. Ratio of  $^{138}\text{Xe}$  to extrapolated  $^{138}\text{Cs}$ .

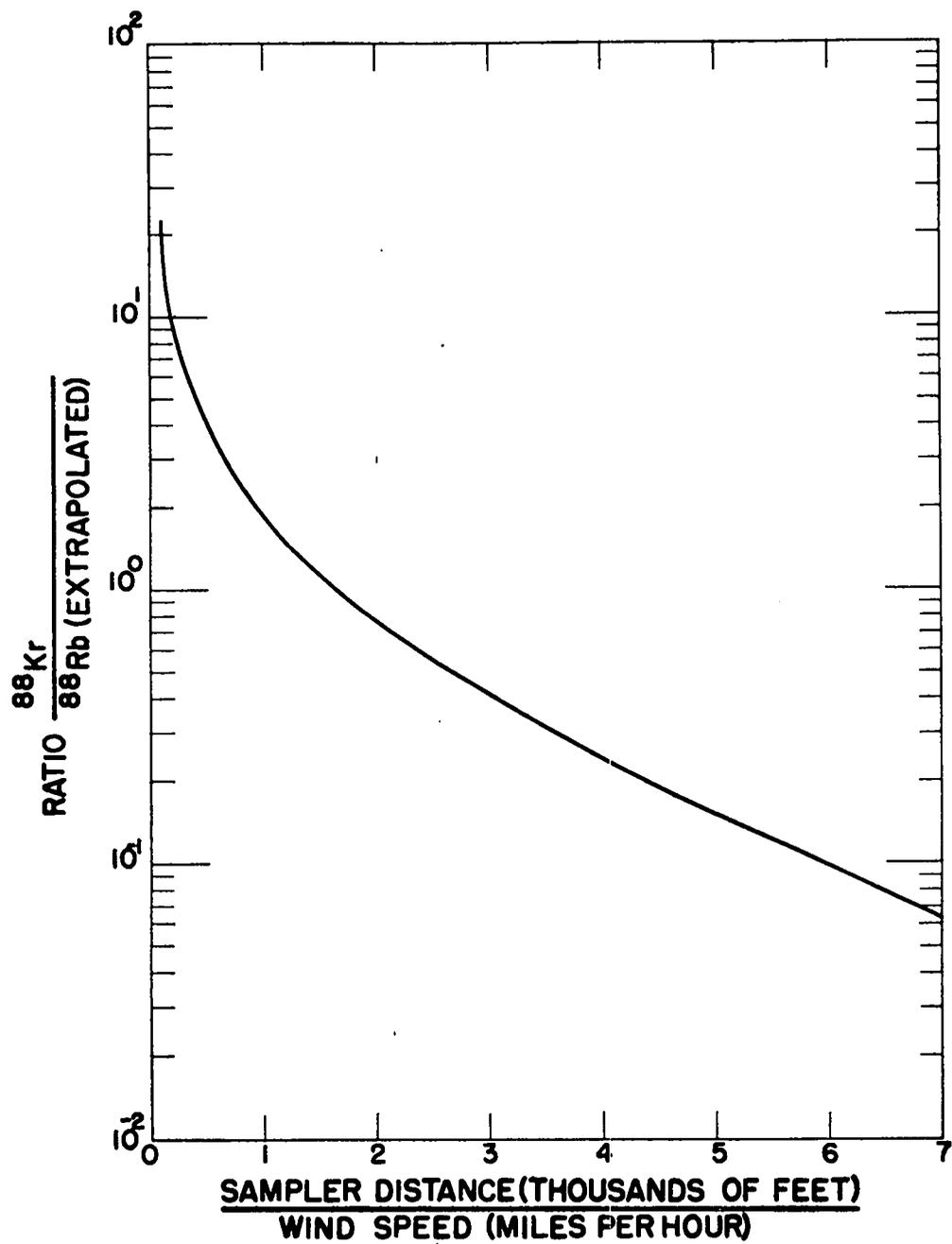


Fig. 7. Ratio of  $^{88}\text{Kr}$  to extrapolated  $^{88}\text{Rb}$ .

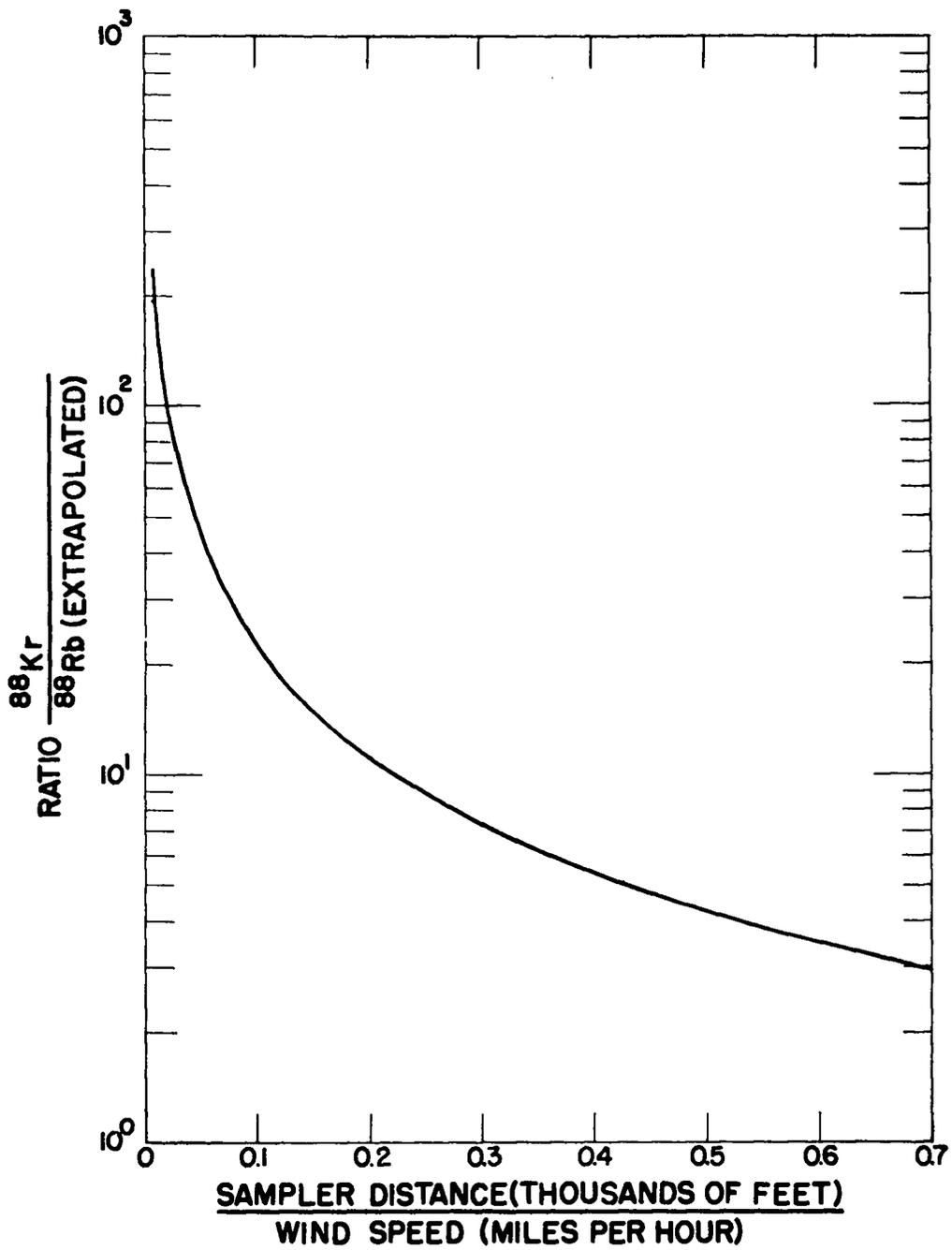


Fig. 8. Ratio of  $^{88}\text{Kr}$  to extrapolated  $^{88}\text{Rb}$ .