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THEORETICAL POSSIBILITIES AND CONSEQUENCES OF MAJOR ACCIDENTS IN U-233 AND Pu-239 FUEL FABRICATION AND RADIOISOTOPE PROCESSING PLANTS

By
C. E. Guthrie
J. P. Nichols

April 1964

Oak Ridge National Laboratory
Oak Ridge, Tennessee

UNITED STATES ATOMIC ENERGY COMMISSION • DIVISION OF TECHNICAL INFORMATION

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CHEMICAL TECHNOLOGY DIVISION

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OAK RIDGE NATIONAL LABORATORY
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U.S. ATOMIC ENERGY COMMISSION

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FOREWORD

by

Harold L. Price, Director of Regulation, USAEC

The Background and Purposes of this Study

The Atomic Energy Act of 1954 removed the restrictions imposed by the Atomic Energy Act of 1946 upon the private possession and use of large quantities of fissile material and upon the private construction of nuclear reactors and other atomic energy facilities.

At the same time, in recognition of the potential hazards to the health and safety of the public from such activities, the Congress included in the Atomic Energy Act of 1954 provisions that gave the Atomic Energy Commission the responsibility and authority to regulate these activities to assure that they would be conducted in such a manner as not to endanger public health and safety.

Since the adoption of the Atomic Energy Act of 1954, private uses of atomic energy materials and facilities have expanded and developed to the point where there are now in the United States more than 11,700 outstanding licenses authorizing the holders to use source, byproduct, and special nuclear materials,* 121 licenses authorizing the construction or operation of nuclear reactors and one license authorizing construction of a plant to reprocess spent nuclear fuel.

Before any individual or firm may acquire possession of atomic energy materials or facilities, or engage in their use, a license must be obtained from the Commission. Each applicant for a license must demonstrate that he is qualified by training and experience, and that his procedure and equipment are such, that the proposed activities will not endanger public health and safety. Each licensee must comply with regulations of the Comission and all special requirements incorporated in the license by the Commission; Commission representatives inspect licensed activities to assure compliance with the Commission's requirements.

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*Of these, more than 2,700 licenses have been issued by states which have entered into agreement with the AEC under Section 274 of the Atomic Energy Act of 1954, as amended.

The safety experience of the atomic energy industry has been outstanding. This experience has demonstrated that atomic energy activities can and are being carried out without jeopardy either to the employees or to the general public; and that by appropriate care in the planning and execution of atomic energy activities the potential hazards can be kept from materializing into dangerous conditions. This gratifying experience is attributable to the care and skill exercised by the individuals and organizations in the atomic energy industry as well as to the effectiveness of the Commission's regulatory and other safety programs.

There is a unique element in the Commission's regulatory program which is an important part of prelicensing safety reviews. Each applicant who wishes to operate a nuclear facility is required to analyze his facility and the proposed operations for various possible mechanisms or circumstances by which severe accidents might theoretically occur. He is then required to demonstrate that by the design of his facility and the procedures for its operation appropriate safeguards will exist to prevent the occurrence of such accidents. In addition, the applicant is required to assume that the worst of these accidents (the so-called "maximum credible accident") occurs despite the preventive safeguards, and then to demonstrate that he has incorporated sufficient consequence limiting safeguards in his facility so that the public outside the plant will not be endangered.

In 1956, the Joint Committee on Atomic Energy requested the Commission to undertake a study of the possible effects of serious reactor accidents. The study, carried out for the Commission by the Brookhaven National Laboratory and completed in March of 1957, was entitled, "Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants," WASH-740, U. S. Atomic Energy Commission. That study included an analysis of the possibilities and consequences of highly improbable, but theoretically conceivable, catastrophic accidents, and was a part of the background against which Congress considered indemnity legislation.

The Act was designed to remove the deterrent to industrial participation in atomic energy presented by the possibility of enormous liability claims in the very unlikely event of a catastrophic nuclear accident and to assure that, if such an accident were to occur, funds would be available to satisfy public liability claims.

The Price-Anderson Act requires each reactor licensee to maintain "financial protection" (typically, nuclear energy liability insurance) in amounts (up to $60 million) prescribed by the Commission to cover public liability claims. In addition, whenever a licensee is required to furnish financial protection, the Act provides $500 million of government indemnity over and above the amount of financial protection.

The Price-Anderson Act applies to all licensed nuclear reactor and other licensed "production" and "utilization" facilities. The Commission is, however, given discretion as to whether or not the Price-Anderson Act should be applied to other kinds of atomic energy activities.

In 1959, the Commission requested the Convair Division, General Dynamics Corporation, to undertake a study of theoretical possibilities and consequences of major nuclear accidents in activities involving the possession and use of substantial quantities of unirradiated uranium enriched in the isotope U^{235}. Based upon information contained in Convair's report (NYO-2980) and other information available to it, the Commission announced in 1961 that the extension of the Price-Anderson indemnity coverage to processors and fabricators who use significant quantities of unirradiated, enriched uranium was not justified inasmuch as the potential consequences of accidents did not appear to be of such a magnitude as to exceed the available private insurance capacity ($60 million).

In 1962, the Commission requested Oak Ridge National Laboratory to prepare a study of theoretical possibilities and consequences of major accidents in plants which process and fabricate uranium-233 and plutonium-239 and plants which process and fabricate very substantial quantities of radioisotopes. In conducting the study, the Oak Ridge National Laboratory was requested to examine operations currently authorized and those likely to be undertaken in the next decade by industry in the fabrication of large isotopic sources and the fabrication of reactor fuel from Pu^{239} and U^{233}.

In conducting this study, the Oak Ridge National Laboratory was requested to include not only those accidents which might be considered to be credible but also to consider in its report, as was done in the study of theoretical reactor accidents prepared by the Brookhaven National Laboratory, the possibilities and consequences of accidents which are theoretically possible but so highly improbable as to be considered incredible.

For this study, we share the opinion about the upper-limit, theoretically possible accidents which was expressed about the theoretically possible reactor accidents considered in the Brookhaven report,

> "We are not aware of such a study having been undertaken for any other industry. We venture to say that if a similar study were to be made for certain other industries with the same free rein to the imagination, we might be startled to learn what the consequences of conceivable major catastrophic accidents in those other industries could be in contrast with the actual experience in those industries."*

It should be emphasized that the major theoretical accidents considered in this report are of a magnitude that the Commission firmly believes will not occur; if their occurrence were considered to be more than of theoretical concern, the activities would not be licensed by the Commission.

In a very real sense, the Price-Anderson Act, and this study, should be viewed not as indicating any belief in the credibility of the major theoretical accidents analyzed in this report, but rather as one more indication of the preoccupation of the Congress and the Commission with protection of the public health and safety in the conduct of atomic activities, and as an ultimate measure in an elaborate program to protect the public health and safety--in this case, to provide compensation for the public against accident consequences which the best available scientific experts advise are incredible but not theoretically impossible.

In this study the authors have of necessity made basic assumptions with respect to such matters as possible accidents, the dispersive characteristics of radioactive materials, meteorological and health physics parameters in order to estimate effects of accidents in dollars. In all cases, the authors have made assumptions and presented descriptions which, in their opinion, are as realistic as possible. Engineering designs and operational procedures have been described for "typical" situations and for illustrative purposes. However, in any particular situation, different choices might well be more appropriate.

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*Excerpt from the letter from the Atomic Energy Commission to the Joint Committee on Atomic Energy, dated March 22, 1957, forwarding the Brookhaven report to the Joint Committee on Atomic Energy.

The information contained in this report will be considered by the Commission, together with other information which may become available, in deciding whether or not the indemnity provisions of the Price-Anderson Act should be extended to various activities considered in this report.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance of a large number of persons.

The need for the study was initially suggested by C. K. Beck, AEC Deputy Director of Regulation. All phases have been made in close cooperation with the AEC Division of Licensing and Regulation, coordinated through J. J. DiNunno. Financial support was provided by the AEC Division of Reactor Development through S. A. Szawlewicz, Chief, Nuclear Safety Research and Development Branch. The study was made under the direction of F. R. Bruce, ORNL Director of Radiation Safety and Control. The meteorological calculation techniques were contributed by W. M. Culkowski and W. F. Hilsmeier of the Oak Ridge U.S. Weather Bureau Research Station.[a]

Many people reviewed drafts of the study and/or contributed helpful comments and suggestions. Among these are: R. E. Baker, C. K. Beck and R. Lowenstein, AEC Division of Licensing and Regulation; A. P. Kenneke, AEC Division of Radiation Protection Standards; Charles Farbstein, AEC Office of the General Council; Alex Perge, AEC Division of Operational Safety; H. V. Clukey, L. H. McEwen and J. M. Smith, General Electric (San Jose); J. J. Fuquay, C. C. Gamertsfelder, R. L. Junkins, and E. C. Watson, General Electric (HAPO); K. V. Best, J. R. Mann, C. W. Piltinsgrud, E. A. Putzier, and C. L. Schuske, Rocky Flats; J. R. Eichelberger, M. R. Hertz and D. L. Scott, Mound Laboratory; and J. C. Bresee, W. E. Browning, W. B. Cottrell, F. L. Culler, B. R. Fish, E. Lamb, R. W. Schaich and W. S. Snyder, ORNL. Recognition here is not meant to imply that the individual has seen or approved this report.

This report as well as many drafts have been patiently typed by Mrs. Fran Quillen, secretary of the Long Range Planning Group of the Chemical Technology Division, ORNL.

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[a]W. M. Culkowski, "Deposition and Washout Computations Based on the Generalized Gaussian Plume Model," ORO-599 (Sept. 30, 1963).

THEORETICAL POSSIBILITIES AND CONSEQUENCES OF MAJOR ACCIDENTS IN U^{233} AND Pu^{239} FUEL FABRICATION AND RADIOISOTOPE PROCESSING PLANTS

C. E. Guthrie
J. P. Nichols

ABSTRACT

Radioisotope source fabrication and Pu^{239} and U^{233} fuel fabrication in privately owned radiochemical plants are expected to increase greatly during the next decade. Inventories of specific isotopes in plants of the future may be factors of 2 to 1000 larger than those common in today's plants. The potential economic loss from off-site damage due to theoretical accidents in such future plants was evaluated.

With the containment and engineered safeguards included in radiochemical plants it is concluded that accidents having more than a theoretical possibility of occurring would not result in a monetary loss due to damage to the surroundings in excess of the limits of private insurance now available ($60,000,000).

It is possible to postulate accidents in plants processing the more hazardous isotopes (e.g., Sr^{90}, Pu^{238}, Pu^{239}, Cm^{244}) which would theoretically result in off-site damage in excess of $60,000,000. Such accidents, however, have such an extremely low probability of occurrence as to be considered incredible. On the other hand, in plants processing some isotopes (e.g., Kr^{85}, I^{131}, Pm^{147}) even the most severe accident imaginable (100% release during unfavorable weather) would not be expected to result in a loss of more than a few million dollars.

INTRODUCTION

As the need for radioisotopes in research, irradiation sources, and power sources expands, private industry is playing an ever-increasing role in their manufacture and processing. In addition, private industry may, within the next few years, make plutonium and U^{233} fuel elements for recycle to power reactors. The purpose of this study was to examine the operations currently authorized and those likely to be undertaken by industry in the next decade in the fabrication of large isotopic sources and reactor fuel from Pu^{239} and U^{233} and to determine: (1) what kinds of accidents might conceivably be experienced by facilities conducting such operations, (2) whether a monetary estimate could be made of the damages, if any, beyond the confines of the facilities from conceivable accidents, and (3) how any monetary estimate so derived would compare with the limits of private liability insurance available.

Major accidents that might occur in radiochemical plants are, in general, of the same nature as might occur in any chemical plant (fires, vessel ruptures, chemical explosions, etc.). The unique feature of radiochemical plant accidents is the possibility that as a result of the accident radioactive material may be dispersed into surrounding areas. Such dispersal may not only result in unavoidable exposure of the public to radioactivity, but may also require measures to limit further exposure to radiation from materials deposited in the area and/or on food supplies. A criticality excursion can be shown to be, in itself, a relatively minor accident with regard to off-site consequences.[1]

Facilities processing materials such as Pu^{239}, U^{233}, or large inventories of radioactive fission products are designed with numerous safeguards against the inadvertent release to the surrounding environs of products hazardous to the health and safety of people. The granting by the AEC of authorization to operate such facilities is done only after consideration of the safeguards involved and a determination that the likelihood of a severe accident having off-site effects is sufficiently

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[1] References at end of Summary.

small that there is no undue risk to health and safety of the public. The safeguards provided generally include: (1) limits on the quantities of materials, (2) ventilation systems planned to minimize the spread of contamination within the building, (3) exit air streams which are filtered and monitored, and (4) two independent barriers between the radioactive materials and the outside. By design and operating plans, both the probability of an accident and the probability of release of radioactivity during an accident are maintained at very low levels. The combination of a very low probability of an accident occurring and a very low probability of a containment failure in case of an accident results in an extremely low risk to the public.

It is much easier to estimate plant capacities in 10 to 20 years than to foresee the advances in technology that may develop in the same period. Undoubtedly there will be many improvements in processes, safeguards, and containment in radiochemical plants. These are not reflected in the accident postulated in this report. The damage consequences are, therefore, undoubtedly on the high side since estimated future inventories were used, while only present safety technology was considered.

As with any study of this kind, the conclusions are only as reliable as the basic assumptions. While every effort was made to choose the best available meteorological, health physics, and economic bases for the calculation, the answers must of necessity be largely qualitative, and no claim is made for their absolute accuracy. For the conditions assumed, the general conclusions reached are believed to be valid even if the figures are off by an order of magnitude. In analyzing any specific situation, care should be taken to review the basic assumptions and modify them where appropriate.

This report is arranged so that each section is, insofar as possible, a complete entity, including references. The summary, besides presenting the salient points, is a prose index of the material in each section.

SUMMARY

The Four Categories of Hypothetical Plant Accidents

A review of radiochemical plant accident experience to date (Section 1) shows that there has been no major off-site contamination from radiochemical plant accidents, and consequently all were much less severe than the accidents postulated in this report. Although there is not sufficient radiochemical plant operating experience to establish a probability for a major accident, in general, the frequency and severity of accidents in such plants to date have been lower than those in the related chemical industry.

Large-scale processing and fabrication of large isotope sources and Pu^{239} and U^{233} fuel elements are areas in which private industry is not yet significantly involved but is likely to enter in the next ten to twenty years. The 1980 yearly production is predicted to be about 10^4 kg of Pu^{239} fuel, 10^8 curies of Sr^{90} sources, and 100 kg of Pu^{238} isotope sources (Section 2). Routine inventories in the "typical" private plant today range from 1/2 to 1/1000 of the inventories assumed in this study for future plants. Inventories in Government plants today, however, in some cases are comparable to those assumed in this study for private plants of the future.

For purposes of this report hypothetical radiochemical plant accidents have been divided into four categories in order of decreasing probability of occurrence and increasing severity of off-site consequences.

Category 1 includes those accidents where a portion of the plant becomes contaminated or an employee is exposed to radiation but no radioactive material escapes from the facility. This type of accident could be caused by some not particularly unusual mishap such as a pipe leak, dropped sample, etc. Damage in this category of accident is confined to the facility and would result in plant cleanup and downtime. There would be no off-site damage.

Category 2 includes those accidents where in addition to damage to the facility a small amount of radioactive material escapes from the plant containment systems. This type of accident could result from some occurrence such as a fire, explosion, criticality excursion, etc. Although the

facility may be lost and possibly some employees injured, there would be only very minor, if any, public damage involved in this type accident. Experience has shown that this type of accident has a very low probability of occurrence. To date all major radiochemical plant accidents have been in Category 1 or Category 2 where no appreciable contamination was spread off site.

An example of a postulated accident in this category for Pu^{239} would be a fire which ruptures a glove box train containing 10 kg of plutonium carbide. This accident is assumed to release 400 g of plutonium to the laboratory, where it is picked up in the building ventilation system. A small amount is assumed to escape the laboratory building through the absolute filter system. The calculated damage for this postulated accident as well as accidents of comparable severity for other selected isotopes is shown in Table 0.1.

Table 0.1 Summary of Theoretical Damage from Category 2 Hypothetical Accidents

	Maximum Release from Building		Off Site Damage from Maximum Release	
Isotope	Percent of Glove Box Inventory Involved	Quantity	Maximum*	Minimum*
Pu^{239}	0.04%	4 g	$\$1.5 \times 10^5$	*
Sr^{90}	0.01%	25 c	1×10^6	$\$1 \times 10^4$
Pu^{238}	0.2%	0.08 g	1×10^6	*
U^{233}	0.04%	4 g	None	None

*Maximum damage (unfavorable meteorological conditions) has been calculated for all isotopes; minimum damage (favorable meteorological conditions) has been calculated only for Sr^{90} to demonstrate the effect of meteorological conditions.

Category 3 encompasses those theoretical accident possibilities that could conceivably result in off-site spread of appreciable quantities of radioactive materials. Accidents in this category have an extremely low probability of happening. This is evidenced by the fact that postulation of such accidents requires the assumption of a sequence of events, each

of which is, in itself, highly unlikely. Further, no accidents of this severity have occurred in almost 20 years of AEC radiochemical plant operation. Unless prelicensing safety review of a facility showed that the possibility of a Category 3 type accident was extremely remote, it would not be licensed for operation.

An example of a postulated Category 3 accident involving Pu^{239} would be one initiated by a criticality excursion in a PuC-graphite fuel storage area containing 100 kg of plutonium. The heat from the criticality excursion is assumed to start a graphite fire followed by a CO explosion which ruptures the storage vault. The outer building is assumed to remain undamaged but some plutonium and volatile fission products (FP's) from the criticality excursion are assumed to escape the building via the exhaust air filter system. The calculated damage from this postulated accident as well as accidents of comparable severity for other selected isotopes is shown in Table 0.2.

Table 0.2. Summary of Theoretical Damage from Category 3 Hypothetical Accidents

	Maximum Release from Building		Off Site Damage from Maximum Release	
	Percent of Cell Inventory	Quantity	Maximum	Minimum*
Pu^{239}	0.04% nonvolatile 100% volatile	40 g plus volatile FP's	$\$5 \times 10^6$	*
Sr^{90}	0.09%	220 c	1.5×10^7	$\$1.5 \times 10^4$
Pu^{238}	0.022%	2.2 g	5×10^7	*
U^{233}	0.04% nonvolatile 100% volatile	20 g plus volatile FP's	2×10^6	*

*Maximum damage (unfavorable meteorological conditions) has been calculated for all isotopes; minimum damage (favorable meteorological conditions) has been calculated only for Sr^{90} to demonstrate the effect of meteorological conditions.

Category 4 encompasses those theoretical accident situations wherein all containment and other safeguards systems are overcome, and widespread dispersal of radioactive material to the atmosphere is postulated. With

the multiple independent safeguards included in radiochemical plants, this type of accident is believed to have such an extremely low probability that it is deemed incredible. The sequence of prerequisite events that would be required to lead to this category of accident are far beyond any reasonable expectation. On the other hand, accidents having consequences in this category are considered in this report because theoretically one cannot conclude that they are completely impossible. Since, as noted, such accidents are not expected to occur (i.e., are "incredible") they fix the upper limit for this study of potential damage.

An example of a postulated accident in Category 4 involving Pu^{239} is assumed to be a criticality excursion in a PuC-graphite fuel storage area containing 100 kg of plutonium. The heat from the criticality excursion is assumed to start a graphite fire. Successive CO explosions are assumed to rupture the storage vault and then the building, releasing the smoke from the fire to the atmosphere. The calculated damage from this postulated accident as well as accidents of comparable severity for other selected isotopes is shown in Table 0.3.

Table 0.3 Summary of Theoretical Damage from Category 4 Hypothetical Accidents

Isotope	Maximum Release from Building		Off Site Damage from Maximum Release	
	Percent of Inventory	Quantity	Maximum*	Minimum*
Pu^{239}	4%	4000 g + FP	$\$4 \times 10^8$	*
Sr^{90}	20%	50,000 c	5×10^9	$\$1.5 \times 10^6$
Pu^{238}	20%	2000 g	5×10^{10}	*
U^{233}	4% nonvolatile 100% volatile	2000 g plus fission products	3×10^6	*
Kr^{85}	100%	10^5 c	None	None

*Maximum damage (unfavorable meteorological conditions) has been calculated for all isotopes, minimum damage (favorable meteorological conditions) has been calculated only for Sr^{90} to demonstrate the effect of meteorological conditions.

Although one might imagine accidents (e.g. release of entire radioactive inventory of a radiochemical facility) greater than those assumed in Category 4, we cannot conceive of any combination of circumstances by which this could happen. Therefore, we made no attempt to consider the damage consequences of any theoretical accident beyond Category 4. Category 4 is considered in this report not because it is ever expected to occur but because it provides an upper limit estimate of the damage under the worst conceivable circumstances.

Basic Assumptions

In order to evaluate the potential economic loss from radiochemical plant accidents, ranges of area contamination and personnel exposure were assumed and a damage value assigned to each range. A detailed discussion of the basis for these assumptions can be found in Section 3. The monetary loss due to personnel exposure was, in most cases, insignificant compared to the loss from land contamination.

A population density increasing from zero at the site to 500 persons per square mile at 20 miles and farther from the site was found to be reasonably typical for fuel fabrication and radioisotope processing plants (Section 4). This is essentially the same distribution used in the comparable reactor liability study.[2] A uniform population density of 100 persons per square mile was also used in calculating the loss economics to allow facile conversion of the results to various sites.

The area and downwind distance as a function of isopleths of exposure and ground contamination were calculated with the Gaussian plume-dispersion model, using experimentally verified values of dispersion coefficient, velocities conducive to deposition of particles (deposition velocities), and washout rates for practical, consistent sets of weather conditions (Section 5). Washout maximizes contamination areas, and inversion maximizes personnel exposure. In most cases the maximum loss is for the washout condition. The damage for a given release may vary by a factor of 10^3 from the most unfavorable (washout) to the most favorable (sunny day) weather conditions (see Table 0.2 and 0.3).

The potential economic loss as a function of the curies released for a number of isotopes was calculated and is presented in Section 6. The

damage varies for the different isotopes depending on the relative biological hazard and other properties of the isotope.

An analysis of theoretical accidents at the upper range of Categories 2, 3 and 4 for several isotopes (summarized in Table 0.1, 0.2 and 0.3) is presented in more detail in Section 7. It is re-emphasized that all these postulated accidents are probably overestimated since they are more severe than any that have occurred to date and assume present rather than future safety technology. The damage calculated for the Category 3 accident is believed to represent the probable maximum damage. Category 4 type of accidents are believed to be incredible and are studied only in order to extend the damage estimates to cover every conceivable possibility no matter how remote the probability.

Conclusions*

It is concluded that with the containment and engineered safeguards included in a radiochemical plant, accidents having more than a theoretical possibility of occurring would not result in a monetary loss due to damage to the surroundings exceeding the limits of private insurance currently available ($60,000,000). It is theoretically possible to postulate accidents (Category 4) in plants that would be processing large quantities of the more hazardous isotopes (e.g., Pu^{238}, Pu^{239}, Sr^{90}) wherein off-site damage in excess of $60,000,000 could be incurred. However, such accidents have such an extremely low probability of occurring as to be considered incredible. On the other hand, in plants processing many other isotopes (e.g., U^{233}, Kr^{85}) even those theoretical accidents (Category 4) which would breach containment during unfavorable weather would not cause loss of more than a few million dollars.

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*These conclusions are valid only so long as the basic assumptions used in calculating the potential economic loss are valid. The two most sensitive assumptions are: (1) that the population and land values are low in the immediate neighborhood of the facility, and (2) that the absolute filters maintain their integrity in Category 1, 2, and 3 accidents.

REFERENCES (Introduction and Summary)

1. H. T. Williams et al., Safety Analysis of Enriched Uranium Processing, NYO-2980 (March 18, 1960).

2. Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants, WASH-740, USGPO, p 78 (Mar. 1957).

1. RADIOCHEMICAL PLANT ACCIDENT POTENTIAL AND EXPERIENCE

In almost every human endeavor there are serious potential hazards to health and safety. Riding in an airplane or automobile can be disastrous. Gas and electricity in the home are accompanied by serious hazards. Dangers are inherently present in transportation and manufacture of explosives and poisonous gases. Yet all these endeavors are regularly carried on with only very infrequent realization of any significant portion of the hazards.

Major accidents that might occur in radiochemical plants (plants that process or fabricate radioactive material) are in general of the same nature as might occur in any chemical plant. During the years the Government's nuclear facilities have been in operation, fire and explosion have been the principal causes of Commission property damage and loss. Only about 11% of the damage resulted from hazards classed as specific to nuclear activities.[1] The types of damage connected with radioactivity that might theoretically occur in radiochemical plant accidents are (1) radiation exposure to workers, (2) ingestion of radioactive materials by workers, (3) contamination of on-site areas and buildings, (4) external exposure or internal ingestion by people in off-site areas, and (5) contamination of off-site areas. Damage from accidents during the 20 years of AEC radiochemical plant operation has been limited to the first three types. There has been no off-site exposure to personnel or contamination.

At this time the public appears to have an unknowing fear of hazards connected with nuclear materials and appears much less willing to accept them compared with the hazards associated with the chemical industry in general. This appears to be primarily because: (1) The industry, being born with the atomic bomb, earned an immediate reputation for mass destruction. (2) The subsequent discussions and conflicting information on fallout have not improved the public image of radioactive materials compared to other materials which may also cause damage (e.g. pesticides, chlorine, etc.). (3) The large amount of discussion, research and study being done on Nuclear Safety, in order to ensure plant safety and assure the public, has tended to distort the picture with regard to the hazards involved (i.e., because there is more nuclear safety work being done, the public appears to infer that there is much greater risk). In actuality, because of the research in Nuclear Safety, the industry is one of the few

where the hazards have been thoroughly studied beforehand and steps taken to prevent and minimize the consequences of accidents. The safety record for nuclear operations is the best in the chemical industry.[1] Similar attention at the beginning to hazards of ammonium nitrate manufacture and transport, for example, might have prevented many serious ammonium nitrate explosions* which have occurred. About 2.5×10^6 tons per year of ammonium nitrate fertilizer are manufactured in the United States, with little or no evidence of public concern despite the potential hazard and past safety record.

Protection Against Accidents

Basically there are two lines of defense against the hazards existing in radiochemical plants: (1) Appropriate means are taken to prevent accidents, and (2) protection is provided against the consequences of an accident if one should occur.

The extent to which efforts are carried to prevent accidents and minimize the consequences of accidents, are indicated in the following general practices:

1. Radioisotopes are processed and handled in batches of limited size; extraordinary precautions are taken to ensure their confinement to intended locations.

2. Most processes are carried out behind heavy shielding within sealed cells or glove boxes.

3. Ventilation systems are so planned that the spread of contamination is minimized; recirculated air is frequently filtered; all exit streams are filtered and monitored, and released only if concentrations are below preset levels.

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*Among the most severe:[2,3,4,5]
1918 Morgan Station, New Jersey: $20,000,000 damage.
1942 Tessenderloo, Belgium: 250 killed, 1000 injured.
1947 Texas City, Texas: ~$300,000,000 damages, 512 killed, >3500 injured.
1947 Brest, France: 20 killed, 500 injured.
Among the more recent:
1960 Traskwood
1961 Norton

4. Plant designs and operations are so arranged that at least three and usually more independent mishaps, malfunctions, or misoperations must occur before an accident results.

5. The containment system is always designed to retain the radioactive materials resulting from the maximum credible accident.

6. There must always be at least two independent barriers between the normal locations of radioactive materials and the outside.

Thus, by design* and operating plans, the probability of an accident is reduced to an extremely low level, and should an accident occur, the likelihood of there being serious consequences is also very low, even for workers in the plant. For the general public this likelihood is far lower. The total hazard, being composed of the probability of the accident multiplied by the probability of hazardous consequences, is obtained by multiplying one very low probability by another. The resulting total risk to the public is extremely low.

Despite the most extraordinary precautions in design and care in operation, it is inevitable that accidents will happen. Failures of components, malfunction of systems, and misoperations of controls cannot be completely avoided. The basic approach to design and operation of fuel and radioisotope processing plants is: first, to minimize the likelihood of failure, malfunctions and misoperations; and second, to ensure that the basic safety objectives are still achieved, recognizing that despite all precautions untoward events will occur.

Theoretical Categories of Accidents

In order to analyze radiochemical plant accidents for purposes of this report, four categories of accidents are postulated in order of decreasing probability of occurrence and increasing severity of off-site consequences:

Category 1 includes those accidents where a portion of the plant becomes contaminated or an employee is exposed to radiation but no radioactive material escapes from the facility. This type of accident could be

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*The design details of radiochemical plants depend on the particular type isotope handled and will be discussed more thoroughly in Section 2.

caused by some not particularly unusual mishap such as a pipe leak, dropped sample, etc. Damage in this category of accident is confined to the facility and would result in plant cleanup and downtime. There would be no off-site damage.

Category 2 includes those accidents where, in addition to damage to the facility, a small amount of radioactive material escapes from the plant containment systems. This type of accident could result from some occurrence such as a fire, explosion, criticality excursion, etc. Although the facility may be lost and possibly some employees injured, there would be only very minor, if any, public damage. Experience has shown that this type of accident has a very low probability of occurrence. To date all major radiochemical plant accidents have been in Category 1 or Category 2 where no appreciable contamination was spread off site.

Category 3 encompasses those theoretical accident possibilities that could conceivably result in release off site of appreciable quantities of radioactive materials. Accidents in this category have an extremely low probability of happening. This is evidenced by the fact that postulation of such accidents requires the assumption of a sequence of events, each of which is, in itself, highly unlikely. Further, no accidents of this severity have occurred in almost 20 years of AEC radiochemical plant operation. Unless prelicensing safety review of a facility showed that the probability of a Category 3 type of accident was extremely remote, it would not be licensed for operation.

Category 4 encompasses those theoretical accident situations wherein all containment and other safeguards systems are overcome and widespread dispersal of radioactive material to the atmosphere is postulated. With the multiple independent safeguards included in radiochemical plants, this type of accident is believed to have such an extremely low probability that it is deemed incredible. The sequence of prerequisite events that would be required to lead to this category of accident are far beyond any reasonable expectation. On the other hand, accidents having consequences in this category are considered in this report because theoretically one cannot conclude that they are completely impossible. Since, as noted, such accidents are not expected to occur (i.e., are "incredible") they fix the upper limit for this study of potential damage.

Although one might imagine accidents (e.g. release of entire radioactive inventory of a radiochemical facility) greater than those assumed in Category 4, we cannot conceive of any combination of circumstances by which this could happen. Therefore, we made no attempt to consider the damage consequences of any theoretical accident beyond Category 4. Category 4 is considered in this report not because it is ever expected to occur but because it provides an upper limit estimate of the damage under the worst conceivable circumstances.

Dispersal and Release Mechanisms

Extensive off-site damage can result only if radioactive materials are dispersed and released from the facility to the atmosphere. Fires and explosions are the primary type of accident that could furnish sufficient energy for the dispersal and release of radioactive materials off site.

Dispersal Mechanisms

In any plant there are materials that will support combustion. Organic solvents, plastic boots from manipulators, rubber gloves, and blotting paper from cleanup operations are all common materials in radiochemical plants. The smoke and hot gases from fires in contaminated areas can entrain and disperse radioactive particulate material. Protection from serious fires in a radiochemical plant, as in any other plant, must rely on fire prevention and the quick detection and extinguishing of small fires before they reach serious proportions. Special care must be taken in designing fire extinguishing systems to ensure that they do not increase the spread of contamination or cause a criticality accident in putting out the fire. Once carefully designed, these systems have a very high degree of reliability in confining and extinguishing fires.

Explosions are much more serious as a dispersal and release mechanism, and more difficult to prevent than fire. Explosions could be caused by either chemical reactions or expanding steam produced by a criticality accident. Chemical explosions could result from nitrated hydrocarbons or air explosions of solvent vapors or mists, hydrogen produced in the dissolution of metals, finely divided metal powders, or carbon monoxide produced as a result of a fire. The main protections against a chemical explosion are careful screening of reagents to prevent the production of explosive

compounds or mixtures and adequate ventilation to prevent buildup of flammable materials to the explosive range in enclosed spaces.

Due to the large negative temperature coefficient in solutions and the large void coefficient associated with radiolytic gas generation combined with the difficulty of adding solution rapidly, accidental criticality involving chemical solutions of fissionable material is a relatively minor dispersal mechanism compared with chemical explosions. Total fission yields of about 10^{20} fissions spread over several hours are possible in solution criticality accidents if reactivity is added slowly.[6] This type of accident would not provide an explosive-type dispersive mechanism. The maximum initial burst fission yield has been estimated[6] to be about 10^{18} to 10^{19} fissions, depending on the solution volume, with the maximum yield per unit volume being no more than 10^{16} fissions per liter for solutions at room temperature. This corresponds to an instantaneous heat generation of about 127 Btu per pound of solution, and radiolytic gas production of 1.1 ft^3 per cubic foot of solution. This would be insufficient energy to raise the solution to boiling if it were below 85°F initially. If the solution were at a higher temperature initially, a lesser number of fissions would occur because of the incremental voids due to steam formation. The inertial pressure caused by rapid generation of radiolytic gas could be sufficient to rupture most process vessels but would hardly provide sufficient energy to rupture containment.

In addition to solutions, fissionable materials are handled in fuel fabrication facilities as dry chemical compounds such as oxides, carbides, and fluorides during chemical processing and fabrication. Accidental criticality in a dry system of these materials would be relatively minor because of the low density of the material and the built-in neutron source from α-n reactions in the chemical compound. Accidental flooding of such systems could lead to fission yields of 2×10^{18} fissions, maximum.[6] Uranium-233 or Pu^{239} in pure metallic form (where up to 10^{21} fissions are possible) would not be expected to be present in a fuel processing and fabrication plant because these metals do not make good power reactor fuels.

The short-term energy release from a criticality accident involving 100 liters of solution is about 30,000 Btu's maximum, most of which would be absorbed in the solution. This compares with the potential energy

release of about 800,000 Btu's in a solvent-vapor explosion in a 20 ft x 20 ft x 20 ft cell. One can see by the energy releases that compared to a solvent explosion a criticality accident, by itself, is a rather minor dispersive mechanism. The main off-site hazard from a criticality accident is gaseous fission products released through the ventilation system.

While the above conclusions as to the magnitude of a criticality excursion are based on studies[6] of the U^{235} system, they also are valid as an upper limit for the U^{233} and Pu^{239} systems. According to Hansen,[7] the energy release in a criticality burst is approximately proportional to the square root of the ratio of the absolute reactivity addition rate to the initial neutron source strength. Considering the properties of the three fissionable isotopes, the reactivity addition rate for a given maximum mass assembly rate would be within the same order of magnitude, while the built-in neutron source strengths for U^{233} and Pu^{239} would be more than ten times higher than for U^{235}, causing a lower energy release from U^{233} or Pu^{239} than from U^{235}. Within the scope of this report it is considered sufficient to make the assumption that the damage due to the release of fission products from a U^{233} or Pu^{239} criticality accident is at a level no greater than that for the U^{235} case.

Once dispersed, as a fine smoke or mist in the cell or glove box, it is theoretically possible for radioactive material to be released to the atmosphere through the filtered ventilation systems or by successive leaks from the laboratory, room, or cell (primary containment) to the building (secondary containment) and through leaks in the building walls to the atmosphere.

It has been shown in ORNL radiochemical plant hazard studies[8,9,10,11] that, in the event of accidents, the activity released from successive leaks through the primary and secondary containment walls is insignificant, compared with the possible release through the ventilation filters. In a contained accident, the blast effects of an explosion are confined to the region of primary containment (glove box, laboratory, storage room, or cell). Although a radioactive aerosol may leak through cracks or penetrations in the primary containment wall and become mixed with the air in the secondary containment zone (building) during the period when the primary containment zone is pressurized, the leaked air is ordinarily not sufficient

to raise the secondary containment pressure above atmospheric. In the event that flow from the secondary containment to the atmosphere is possible, perhaps because of a lee vacuum induced on the building by a high wind, the leak rates and ventilation rates in typical plants are such as to cause the predominant release of activity through the primary and secondary containment ventilation filters. This would be the primary cause of release in Category 2 and Category 3 accidents, the distinction between the two categories being determined by the amount of material dispersed and the efficiency of the filters for removal of the material.

The so-called "AEC absolute" filters that are widely used in radiochemical plants are the weakest link in the containment of accidents. The susceptibility of the filters to both physical and chemical degradation necessitates that their integrity and efficiency be ensured by routine in-situ testing or by preplacement testing plus careful installation and operation. The filters must be protected from excessive corrosion and excessive loadings of dust or water, and they must be located so that they can withstand the blast wave from credible explosions without rupture. In typical facilities, the tortuous path and expansions and contractions of the ventilation duct are sufficient to reduce the blast wave from credible explosions to a tolerable level at the filters.

Experience with AEC absolute filters operating at the rated flow revealed that they have a better than 99.95% efficiency for removing particles of size greater than 0.3 μ, that the efficiency decreases to a minimum of approximately 87% for particles of 0.05 to 0.1 μ, and that the efficiency is greater than 87% for smaller particles.[12] Cheever showed that these filters are approximately 99.5% efficient in removing smoke from a plutonium fire, varying in size from 0.004 to 0.03 μ, and that the addition of one to six backup filters in series did not significantly improve the efficiency.[13]

In this report it is assumed that filters have removal efficiencies of 99% for particles smaller than 0.05 μ, 87% for particles 0.05 to 0.1 μ, 95% for particles 0.1 to 0.3 μ, 99.95% for particles 0.3 to 5 μ, and 100% for particles larger than 5 μ. Smokes from fires of metal, solid carbonaceous materials, or organic liquids (which would be predominantly 0.01 to 0.1 μ in diameter when initially produced but which agglomerate rapidly) are assumed to be 99% removed in filters.

When radioactive liquid aerosols are formed by an explosion in a tank of radioactive solution in a cell, the concentration of solution in the air entering the ventilation system is not expected to exceed that in rain or drizzle: 100 to 1000 mg of solution per cubic meter of air. Use of the approximate particle-size distribution of such an aerosol from the ORNL hazard studies[8,9] and the assumed filter efficiencies results in a filter effluent concentration of 0.14 mg/m^3. It is assumed for such an accident that one cell volume (about 100 m^3) of air containing 0.14 mg of radioactive solution per cubic meter is released to the atmosphere.

Radiochemical Plant Accident Experience

"The past is prologue." It is, therefore, pertinent to review the past radiation accidents in order to know what might be expected of the future. During the 20 years in which radiochemical plants have been in operation, mostly at Government sites, the frequency and severity of accidents have been significantly lower than that in the related chemical industry. Only a few of the accidents that have occurred in radiochemical plants involved radioactive material (about 11% of the total loss), and none resulted in damage off site.[1] The cost of the worst accidents involving radioactive materials resulted primarily from decontamination or replacement of buildings and equipment. No accident where damage resulted primarily from nuclear materials is believed to have exceeded $500,000.

Past radiation accidents that are pertinent to possible future accidents in fuel or radioisotope handling facilities are summarized in Table 1.1. Many of these accidents are reviewed in Nuclear Safety,[14,15] in the proceedings of the Karlsruhe Criticality Symposium,[7] and in a recent book, Nuclear Liability.[16]

The conclusions one can draw from these accidents are: (1) In general they were caused by a series of unrelated mishaps or errors. (2) In comparison with a fire or explosion, a criticality accident in a radiochemical plant is relatively minor; it can, however, be the forerunner of some more serious accident. (3) No accident to date has been more severe than a Category 2 accident. (4) No radiation accident to date is believed to have resulted in damage to the facility in excess of $500,000. (5) None of the accidents resulted in damage off site.

Table 1.1 Causes and Effects of Some Radiation Accidents

Accident	Category	Causes and Effects	Cost
Hanford, Pu^{239} criticality accident	1	Control rod removed from Pu^{239} solution too fast; 3×10^{16} fissions, single burst; dispersed Pu to building.	
subsequent fire	2	Subsequent fire believed due to spontaneous combustion in decontamination waste required abandonment of building because of recontamination.	*
RFP, Pu^{239} fire	2	Fire in Pu^{239} metal dispersed smoke to building and burned filters no large personnel exposures or downwind contamination.	~$400,000
Y-12 Plant, Oak Ridge, criticality accident	1	U^{235} solution in nonsafe geometry; 1.3×10^{18} fissions total; 8 employees suffered sublethal exposures.	*
LASL, criticality accident (1958)	1	Agitator created critical geometry in Pu solution tank; 1.5×10^{17} fissions no physical damage; one lethal, 2 sublethal employee exposures.	*
ICPP, criticality accident (1959)	1	U^{235} solution siphoned from safe to unsafe geometry in shielded cell; no physical damage; no excessive personnel exposures.	$62,000 ($U^{235}$ loss)
ORNL waste-system release	2	55 curies Ru^{106} and Ce^{144} released to process waste system; activity collected in ORNL-controlled creek.	~$10,000
Evaporator explosion, ORNL, Building 3019	2	Explosion in in-cell evaporator containing 150 g Pu^{239}; six-tenths of a gram blown through open cell door; building and adjacent facilities contaminated.	$350,000
Mound, glove-box ruptures	1	Glove-box explosions dispersed alpha material to the room on two occasions; no significant personnel exposure.	$1900 + $31,000
Hanford, dissolver fire-explosion	2	Fire and explosion of uranium metal in a chemical dissolver damaged dissolver and contaminated process canyon.	$250,000
ICPP, criticality accident (1961)	1	U^{235} solution transferred to unsafe geometry in shielded cell; no physical damage; no excessive personnel exposures.	None

* Cost figures not available but not believed to exceed $500,000 in facility damage or cleanup cost.

The following is a more detailed summary of the accidents:

Plutonium-239 Criticality Accident at Hanford Atomic Products Operation [7]

On November 16, 1951, a criticality accident occurred in a Hanford Atomic Products Operation building when the control rod from a Pu^{239} solution criticality experiment was removed too rapidly. A total of 8×10^{16} fissions occurred in a single burst before the assembly was made subcritical by void formation and insertion of the safety rod. Plutonium nitrate solution contaminated the experimental area and a portion of the building. Several months later, before the decontamination was completed, a fire occurred in the material used for decontamination and caused a redistribution of contamination. The building was abandoned. The fire served to emphasize that unrelated subsequent accidents may occur and compound the consequences of the original one.

Plutonium-239 Fire at the Rocky Flats Plant [16,17]

A fire on September 11, 1957, involving about 26 kg of Pu^{239}, resulted in gross contamination of a portion of the process building. The fire apparently started by the spontaneous ignition of a Pu^{239} skull (casting residue) in a Plexiglas glove box. The Plexiglas walls and neoprene gloves on the box were ignited and caused the fire to spread to glove boxes throughout the room. Although the fire burned the ventilation filters, and there were at least two minor explosions in the room, there was no excessive ground contamination around the stack or on the site. Most of the process building was returned to operation within one day, and the remainder was decontaminated successfully.

Criticality Accident at the Y-12 Plant [7]

On June 16, 1958, a criticality accident occurred in an Oak Ridge Plant (Y-12) when U^{235} solution was accidentally collected in a vessel of non-safe geometry. Three unusual occurrences were required to cause the accident: (1) A nonsafe container was used as a result of misinterpretation of safety rules. (2) Fissile solution collected in an unanticipated location because of a leaky valve and accidental combination of other valve settings. (3) An experienced operator observed the slow flow of distinctively colored uranyl nitrate solution without recognizing the hazard and shutting off the flow. A total of 1.3×10^{18} fissions occurred over a 20-min period before the reaction was finally shut down by a flow of water that decreased the

concentration of the solution below the critical concentration. No physical damage resulted, and very little activity was dispersed, but eight employees suffered sublethal radiation exposures.

Criticality Accident in 1958 at Los Alamos Scientific Laboratory[7,18]

On December 30, 1958, a criticality accident occurred in an unshielded plutonium processing area at the Los Alamos Scientific Laboratory when plutonium solids were inadvertently flushed to a tank containing aqueous and organic solutions during a cleanup operation. The solids dissolved and extracted into the organic layer. The starting of the agitator in the tank caused the organic layer to assume a critical geometry. A total of 1.5×10^{17} fissions occurred before the system was made subcritical. There was no damage to the facility or significant contamination, but one employee was killed and two others suffered sublethal exposures because of the prompt neutron and gamma radiation.

Criticality Accident in 1959 at the Idaho Chemical Processing Plant[7]

On October 16, 1959, a criticality accident occurred in a shielded cell at the Idaho Chemical Processing Plant when a U^{235} solution accidentally siphoned from safe vessels to a large tank. A total of 4×10^{19} fissions occurred in the 800 liters of solution over a period of 15 to 20 minutes until the reaction was shut down by boiling and ejection of solution. There was no physical damage, and no one received a significant radiation dose through the thick concrete shield. Seven persons outside the building received whole body exposure up to a maximum of 50 rems from the cloud and contamination that resulted from the release of a portion of the fission products through the off-gas system to the plant stack. No off-site personnel exposure was experienced because of the large exclusion radius of the site.

Release of Radioactive Chemicals into the Process Waste System at Oak Ridge National Laboratory[19]

At ORNL, on October 29, 1959, an evaporator steam coil leaked and released 2000 curies of mixed fission products to the process waste system, which normally contains only a low level of contamination. Impoundment of water in an ORNL-controlled creek was necessary to prevent the uncontrolled release of about 55 curies of Ru^{106} and Ce^{144} into the Tennessee River. This quantity of radioactivity would have been discharged to the river if

a rise in activity in the waste-water system had not been detected in routine surveys.

Explosion in an Evaporator in ORNL, Building 3019 (ref. 19,20)

On November 20, 1959, a chemical explosion occurred in an evaporator containing plutonium within a shielded cell in Oak Ridge National Laboratory Building 3019. The explosion resulted from a complex combination of circumstances during decontamination of the evaporator: (1) An unanticipated material was present, phenol in a proprietary decontamination agent. (2) The design of the evaporator was such that it could not be completely drained. (3) Due to a combination of operational error and procedure changes, (a) the evaporator containing residual decontamination agent was not flushed with water before the addition of dilute nitric acid, and (b) the nitric acid was allowed to boil and concentrate in the evaporator. This series of circumstances resulted in the nitration of the phenol and an explosion. The evaporator contained as estimated 150 g of plutonium as solution and precipitate at the time of the explosion; an adjacent vessel, a steam stripper, contained about 1000 g of plutonium as scale and precipitate. The explosion dispersed about 150 g (probably the entire contents of the evaporator) of the plutonium to the cell, and an estimated 0.6 g was blown through a cell door directly to the outside air. No one was injured or received an overexposure to radiation; however, a portion of the ORNL site was contaminated above ORNL standards. In addition, the operating area of the building was contaminated by air flow through open pipe chases and other penetrations that communicated through the cell wall. About 1.5 g of Pu^{239} was removed in the cell ventilation filter system, and no significant quantity of Pu^{239} was released through this system to the plant stack.

Glove-box Ruptures at Mound Laboratory[14,15]

Alpha active material has been released to the building on two occasions when glove boxes were ruptured. In one release (Aug. 6, 1959) a spontaneous explosion in a glove box caused 39 curies of polonium to be dispersed into the building. In the other (July 6, 1960), gas pressure built up in a box, blew out the gloves, and dispersed radioactive material into the room. Radiation exposures were low and the buildings were successfully decontaminated.

Fire and Explosion in a Dissolver at Hanford Atomic Products Operation[21]

On April 17, 1960, fire and explosions involving nitric acid and uranium metal occurred in a dissolver in a shielded canyon at Hanford Atomic Products Operation. There was no exposure of operating personnel to radiation and no significant release of activity to the building or to the plant environs. Damage was limited to the loss of the dissolver and contamination of the process canyon. This is an example of a credible accident in which the consequences were confined to the plant by adequate design of containment barriers and the ventilation systems.

Criticality Accident in 1961 at Idaho Chemical Processing Plant[7,22]

On January 25, 1961, a criticality accident occurred in a shielded cell at Idaho Chemical Processing Plant when a U^{235} solution was accidentally forced into a vessel of unsafe geometry because of a plugged line. A total of 6×10^{17} fissions occurred before the solution gravitated back into a subcritical geometry. There was no physical damage, and no whole-body exposures to workers were greater than 100 mrem from the radiation that penetrated the thick concrete shield or from the fission product cloud.

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2. RADIOCHEMICAL PLANT DESIGN CHARACTERISTICS AND PLANT INVENTORIES ASSUMED FOR THIS STUDY

The quantity of radioactive material that could theoretically be released to the environment in radiochemical plant accidents is a function not only of the possible dispersive release mechanisms discussed in Section 1 but also of the design of the plant and the inventory of radioactive material. It is much easier to predict plant capacities in 10 to 20 years than to predict the advances in technology that may develop in the same period. Undoubtedly there will be many improvements in processes, safeguards and containment in radiochemical plants. These are not reflected in the plant design characteristics assumed in this report.

Estimated Production of Isotope Sources and U^{233} and Pu^{239} Fuel

Estimates of the possible yearly production of Pu^{239} and U^{233} fuel and isotope sources by private manufacturers are given in Table 2.1. At present, the production rate of Pu^{239} and U^{233} fuel by private sources is about 50 and 25 kg/year, respectively, consisting almost entirely of the fabrication of experimental fuel pins and fuels for use in criticality experiments. At the anticipated installed reactor capacity of 2×10^4 Mw_{th} in 1970, the plutonium discharged from power reactor that year will be 1200 kg; plutonium production in 1980 should be about 10 times greater. Plutonium utilization should not lag much behind production, at least by 1980. Including recycle streams, processing and fuel fabrication would amount to about 40 kg of plutonium per day by 1980 and might amount to as much as 4 kg/day by 1970. Uranium-233 processing is most likely to be only a tenth as much during this period.

The most extensive programs for alpha-emitting radioisotopes other than reactor fuel involve their use as heat sources. These sources are particularly useful in space programs since the radiation level for a comparable amount of heat can ordinarily be obtained with significantly less shield weight than for a beta-gamma emitter. Estimates of private manufacturer's capability and possible production for the isotopes of greatest interest in this class (Cm^{242}, Pu^{238}, Cm^{244}, and Po^{210}) are tabulated in Table 2.1.

Table 2.1. Estimates of Possible Yearly Production of Pu^{239} and U^{233} Fuel and of Radioisotopes by Private Manufacturers

Fuel Type or Isotope	Projected Production Rate		
	1963	1970	1980
Pu^{239}	50 kg	1200 kg	12,000 kg
U^{233}	25 kg	100 kg	
Cm^{242}[a]	0	80 g	
Pu^{238}[a]	3 kg	50 kg	100 kg
Cm^{244}[a]	0	50 kg	
Po^{210}[a]	20 g	100 g	
Sr^{90}[a]	3×10^6 curies	1.5×10^7 curies	10^8 curies
Cs^{137}[a]	10^6 curies	1.3×10^7 curies	9×10^7 curies
Tm^{170}		10^6 curies	
Tl^{204}		10^6 curies	
Sb^{124}		10^5 curies	
Pm^{147}[a]	2×10^4 curies	4×10^7 curies	2.7×10^8 curies
Ir^{192}		10^5 curies	
I^{131}		10^4 curies	
Kr^{85}		10^5 curies	
Co^{60}		3×10^6 curies	
Ce^{144}[a]	10^6 curies	9×10^7 curies	6×10^8 curies

[a]Estimates of potential capacity of private manufacturers (see ref 1, end of this section).

The projected potential capacity of private manufacturers for the production of beta-gamma-emitting radioisotopes are also shown in Table 2.1. The demand for these isotopes for gamma irradiation sources and for underwater, underground, and space-program heat sources is rapidly increasing.

The quantity of each isotope stored or processed at one time (plant inventory) will depend not only on the demand for that isotope but also on such factors as heat release, radiation intensity, and criticality. The inventory of material in a plant at any time will be limited to the amount that can be conveniently and safely handled. Table 2.2 summarizes the current maximum inventory (either approved or requested)[2] for present private facilities and for the inventory that has been assumed in this study for future plant. Plant inventories assumed for the future range from 2 to 1000 times the current private plant inventories, depending primarily on how extensively private industry is currently engaged in processing the individual isotope. In general, the inventories assumed for future plants are of the same order of magnitude as typical present inventories in Government plants.

Design and Inventories Assumed for Radiochemical Plants of the Future

The general basis for the design of radiochemical plants is that the shielding and containment features must be such that during normal operation the allowable limits of personnel dose and air and water contamination as specified in Title 10, Code of Federal Regulations, Part 20, are not exceeded. There are no universally applied bases for the design for the case of an accident, but the containment features must restrict the release of activity in the event of an accident to a level that will not cause off-site damage due to personnel exposure or ground contamination. At least two and usually three independent barriers must be between the isotope being processed and the environment. Fuel fabrication and radioisotope processing plants may be divided on the basis of the required plant design details into three general categories: (1) the fabrication of Pu^{239} or U^{233} nuclear fuel, (2) the processing of short-half-life alpha-emitting isotopes, and (3) the processing of beta-gamma Bremsstrahlung emitters. The third category may be further subdivided into facilities for processing (a) primarily beta emitters, (b) hard gamma emitters, and (c) volatile fission products.

Table 2.2. Maximum Current Inventory of Maximum Requested Inventory of Radioisotopes in Private Fuel or Radioisotope Processing Plants as Compared with the Inventories Assumed for this Study

Isotope	Inventory in Plant	
	Current Maximum or Maximum Requested[a]	Assumed for this Study
Co^{60}	1,000,000 curies	5,000,000 curies
Kr^{85}	1500 curies	50,000 curies
Sr^{90}	400,000 curies	20,000,000 curies
Sb^{122}	100,000 curies	-
Sb^{124}	20,000 curies	50,000 curies
I^{131}	200 curies	500 curies
Cs^{137}	10,000 curies	3,000,000 curies
Ce^{144}	10,000 curies	1,000,000 curies
Pm^{147}	10,000 curies	50,000 curies
Tm^{170}	200,000 curies	500,000 curies
Tm^{171}	50,000 curies	-
Ir^{192}	15,000 curies	200,000 curies
Au^{198}	100,000 curies	-
Au^{199}	100,000 curies	-
Po^{210}	5,000 curies	90,000 curies
U^{233}	25,000 g	50,000 g
Pu^{238}	12 g	10,000 g
Pu^{239}	50,000 g	100,000 g
Cm^{242}	3.6 g	20 g
Cm^{244}	-	10,000 g

[a]See ref 2 at the end of this section.

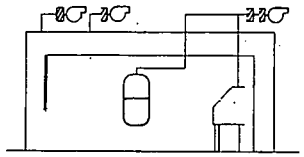
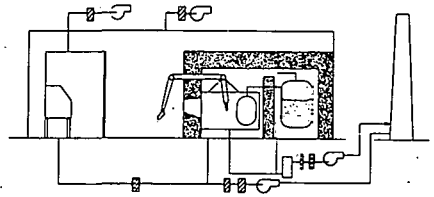
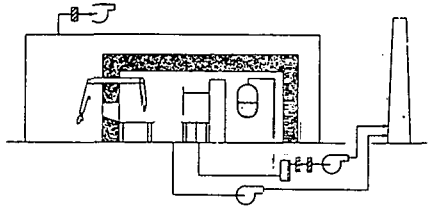
Pu^{239} or U^{233} Fuel Fabrication

The design characteristics and inventory assumed for a typical Pu^{239} or U^{233} fuel fabrication plant of the future are summarized in Table 2.3. Plutonium-239 or U^{233} is handled in glove boxes (first barrier) located in a laboratory (second barrier), which, in turn, is ordinarily located in a building (third barrier). The glove boxes are tightly sealed and are maintained at a vacuum of 0.2 to 0.5 in. (water gauge) with respect to the laboratory and building. Ventilation air for the boxes is drawn in through absolute filters, passes through the box, and exits through a roughing filter and double AEC "absolute" filters to a fan that discharges at the roof of the building or to a stack. The laboratory and building are normally ventilated by drawing air into the building through roughing filters and cracks and then exhausting it through roughing and absolute filters by means of a suction fan at the roof. Normally, the laboratory and building are of "fireproof" construction and are protected with automatic water-spray systems. The laboratory may contain combustible materials such as Plexiglas, vinyl-tile floors, blotter paper, flexible tubing, rubber gaskets and gloves, clothing, natural gas, and containers of combustible solvents and gases. The glove boxes, with thin rubber gloves and/or manipulators, normally contain only small quantities of combustible materials, but experience shows that these small quantities of combustible materials can be enough to rupture the glove box and disgorge a portion of its contents in the event of an internal fire or explosion. Some shielding is necessary surrounding the glove boxes. In the case of U^{233}, gamma rays are emitted by decay products of the U^{232} contaminant. In the case of plutonium recycled from power reactors, neutrons are produced by spontaneous fission of the Pu^{240} and Pu^{242} isotopes.

The fuel-fabrication plant also ordinarily contains closed tankage for the storage of large quantities of Pu^{239} or U^{233} solution, and storage space for containers of completed fuel elements or fuel materials which are also double contained.

While no more than half of the minimum critical mass is ordinarily handled in a glove box in commercial facilities at present, many multiples of this mass may be handled if reasonable precautions are taken to exclude large quantities of hydrogeneous material (moderator) from the box. In

Table 2.3. Assumed Design Characteristics and Estimated Material Inventory for Fuel Fabrication and Radioisotope Processing Facilities of the Future

PURPOSE OF PLANT	SCHEMATIC ARRANGEMENT OF PLANT	ISOTOPE	ASSUMED PLANT INVENTORY		
			IN SINGLE GLOVE BOXES	IN SINGLE VESSEL OR CUBICLE	IN FACILITY
NUCLEAR FUEL FABRICATION		Pu^{239}	2500 g	100 kg	100 kg
		U^{233}	2500 g	50 kg	50 kg
PROCESSING OF ALPHA-EMITTING ISOTOPES		Cm^{242}	0.04 g	20 kg	20 kg
		Pu^{238}	40 g	10 kg	10 kg
		Cm^{244}	0.10 g	2 kg	2 kg
		Po^{210}	0.15 g	10 kg	10 kg
PROCESSING OF BETA-GAMMA EMITTING ISOTOPES		Sr^{90}	-	2.5×10^5 curies	2×10^6 curies
		Cs^{137}	-	2×10^5 curies	3×10^6 curies
		Tm^{170}	-	1×10^5 curies	5×10^5 curies
		Sb^{124}	-	1×10^4 curies	5×10^4 curies
		Pm^{147}	-	5×10^4 curies	5×10^4 curies
		Ir^{192}	-	1×10^5 curies	2×10^5 curies
		I^{131}	-	5×10^2 curies	5×10^2 curies
		Kr^{85}	-	1×10^5 curies	1×10^5 curies
		Co^{60}	-	1×10^5 curies	5×10^6 curies
		Ce^{144}	-	1×10^6 curies	1×10^6 curies

this report it is assumed that 2.5 kg of U^{233} is a reasonable upper limit for the quantity that might conveniently be handled in a single glove box. It is anticipated that the glove-box train might contain several boxes each with this mass of fissile material.

Processing of Short-Half-Life Alpha Emitters

Radioisotopes such as Cm^{242}, Pu^{238}, Cm^{244}, and Po^{210} are handled in closed vessels or cubicles (first barrier) inside a remotely operated, shielded cell (second barrier). The shielding is necessary because the isotopes also emit some gamma and neutron radiation due to spontaneous fission and alpha-neutron reactions. Tightly closed vessels and cubicles are necessary to prevent spread of contamination because of the great mobility of the alpha-active materials. The cells are normally enclosed within a building (third barrier), which contributes an additional line of containment.

Progressively lower pressures are maintained in the building, the cell, the cell cubicle, and the cell vessels, respectively. Cell vessels and cubicles are maintained at the required low pressure, usually about 1.3 in. (water gauge) below atmospheric, by an off-gas system that exhausts through double absolute filters to a stack. The cells are ventilated and maintained at 0.02 to 0.3 (water gauge) below atmospheric pressure by a system that draws air into the building through roughing filters and leaks and then discharges it at the roof of the building through a single set of absolute filters and a fan.

Smaller quantities of material for experimental studies and analytical determinations may be handled in glove boxes (first barrier) in a laboratory (second barrier) in the building (third barrier). The quantity handled in the boxes is generally limited by the penetrating radiation through the thin glove-box shields. The quantities shown in Table 1.2 as glove-box inventories were chosen on the basis that the penetrating radiation dose rate 1 ft from a glove box would not exceed about 2.5 mrem/hr (these dose rates are predominantly determined by the neutrons from spontaneous fission and alpha-neutron reactions with oxygen). The glove boxes are generally held at a pressure about 0.3 in. (water gauge) below that of the building and are normally exhausted through double or triple absolute filters to the stack.

Processing Plant Design for Beta-Gamma Emitters

The processing of beta-gamma emitters is accomplished in shielded cells. Ordinarily the cells are designed for multipurpose use, and the cell wall is made of thick concrete, sufficient to attenuate the radiation from the maximum intended inventory of the isotope emitting the gamma rays of highest energy. Bremsstrahlung from the so-called "pure beta" emitters such as Sr^{90} also requires the use of heavy shields. Operations are done remotely by manipulators.

The containment and ventilation systems are in general the same as those for alpha isotope processing described previously. Except for large quantities of materials, however, tightly closed cubicles and tanks within the cell are not ordinarily required for processing beta-gamma emitters because of the relatively low mobility and greater detectability of the material. The cells sometimes have another ventilation system for an in-cell vacuum cleaner or hood which is used in dusty operations; this system discharges through double absolute filters to the stack.

Processes Used in Radiochemical Plants

Although the detailed processes used in the fabrication of the various isotope sources differ, the work normally consists in the following general steps: dissolution of reactor irradiated targets or recovery of crude isotopes from reactor fuel processing waste; purification by ion exchange, solvent extraction, or precipitation; source fabrication by precipitation, calcination, blending, hydraulic processing, sintering, loading, sealing, and inspection.

In nuclear fuel fabrication plants, U^{233} or Pu^{239} would probably be transferred into the plant as an aqueous solution, stored in critically safe tankage, and transferred in batches of limited size to the glove-box train for processing into fuel compounds. Process steps for the production of oxide fuels normally consist of precipitation (usually along with a fertile material), calcining at a high temperature in an argon-hydrogen atmosphere, crushing, mixing, sizing, tube loading, welding, and inspection.

Pyrolytic-carbon-coated carbide fuels are made by converting Pu^{239} or U^{233} oxides (or mixtures of them with fertile oxides) to carbides by contact with carbon at high temperature in a hydrogen-argon atmosphere, and

forming pyrolytic-carbon-coated particles by fluidizing the carbide particles at high temperatures in methane or acetylene and hydrogen. Fuel elements are then made by mixing the coated particles with carbon, extruding the mixture into a "green" element, and graphitizing.

The finished element is removed from the glove box and stored until shipped. Fuel fabrication plants would also normally include dissolution and solvent extraction or ion exchange equipment for scrap recovery and/or purification of off-specification material.

REFERENCES (SECTION 2)

1. W. K. Eister, Division of Isotopes Development, AEC, Washington, personal communication, January 17, 1963.
2. J. J. DiNunno, Division of Licensing and Regulation, AEC, Washington, personal communication.

3. ECONOMIC-LOSS ASSUMPTIONS AS A FUNCTION OF PERSONNEL EXPOSURE AND AREA CONTAMINATION

Major uncertainties in the study are the questions of (1) the personnel exposure and area contamination levels for individual isotopes that would result in damage to individuals, crops and property and, (2) the potential economic loss incurred. Since there are no standards for personnel emergency exposure or contamination for individual isotopes, and no reliable economic values relating to radiation exposure or contamination at any level, it was necessary to assume economic values for various ranges of exposure and contamination. The bases used are best approximations only and cannot be taken as firmly established. A series of articles summarizing the status of our knowledge on the effects of radiation on man appeared recently in Nucleonics.[1] This provides some understanding of the effects of whole body radiation and the rationale behind radiation protection standards but little basis for establishing damage from internal exposures of individual isotopes.

Ranges of Personnel Exposure and Potential Economic Loss

The most serious exposures from radiochemical plant accidents would probably result from the inhalation of radioactive materials. It is difficult if not impossible to predict at this time exactly what intake of various radioisotopes would cause significant damage. In fact, the effects, if any, are likely to be varied, depending on the individual, the isotope, and the form in which it is ingested. For insoluble materials, the lung would sustain the largest short-term dose. We have chosen the lung as the organ of reference. It should be understood, however, that for some isotopes other organs may be more important due to long-term exposure. In addition there is little if any precedent for establishing potential economic loss resulting from internal exposures to the general population. This loss will include not only compensation to those who show damage but also costs incurred for dose determination, long-term medical observation, and other expenses involved with those who show no damage.

The allowable industrial exposure is 5 rems per 13 weeks for the lung and 10 rems per 13 weeks for the thyroid.[2] For purposes of estimating an economic loss for this study, the authors have arbitrarily assumed three

ranges of personnel exposure and loss: Range A, exposures of greater than 100 times the allowable industrial, with an assumed loss of $50,000 per person; Range B, exposures of 10 to 100 times the allowable industrial, with an assumed loss of $10,000 per person; and, Range C, exposures of 1 to 10 times the allowable industrial, with an assumed loss of $2000 per person.

Except for Kr^{85} and I^{131}, the ingested material is assumed to be insoluble, and, except for I^{131}, the lung is assumed to be the critical organ. This assumption is probably more conservative for short-lived isotopes than for long-lived ones. Table 3.1 shows the intake in microcuries and curie-seconds per cubic meter (curie-sec m^{-3}) for a 13-week dose of 5 rems to the lung for several isotopes of interest. The microcurie values are based on dose calculations by G. W. Dolphin et al.[3] The units of curie-seconds per cubic meter were used for convenience in calculating the exposure isopleths resulting from a release. A respiration rate of 220 cc/sec has been assumed for converting from microcuries to curie-seconds per cubic meter. As an example of whole-body-vs-lung exposure, a radioactive cloud of Cs^{137} of sufficient concentration and duration to provide an intake (2500 μc) resulting in an internal exposure of 500 rem in 13 weeks to the lung would give a short-term whole-body gamma dose of about 3 rems from external radiation.

Area Contamination and Potential Economic Loss

Choosing reasonable criteria for ranges of area contamination and corresponding potential economic loss is difficult because of the large number of factors that might affect the loss. In farm areas, for example, the economic loss at the time of harvest would depend directly upon the crop being raised, type of soil, rainfall, at what time in the growing season the contamination occurred, etc. Other less-tangible factors could also affect the monetary loss. For example: (1) Extent of area contaminated--if only a small area were contaminated, the land might be allowed to lie fallow as part of the normal farming cycle without economic loss. (2) Public opinion--if a farm or farm areas are known to have been contaminated, the public might not purchase crops from this area even though the crops were well below the accepted level of contamination (note the cranberry

Table 3.1. Internal Dose from Single Intake of Selected Isotopes

Isotope	13-Week Lung Dose for Unit Intake[a] (rems/μc)	Single Intake of Insoluble Material for Exposure of 5 rems[b] to Lung over 13 wks (μc)	Single Intake of Insoluble Material for Exposure of 5 rems[b] to Lung over 13 wks (curie-sec m^{-3})
Co^{60}	0.33	15	6.9×10^{-2}
Kr^{85}[b]	-	-	7.2×10^{1}[b]
Sr^{90}	0.55	9.1	4.1×10^{-2}
Sb^{124}	0.3	17	7.6×10^{-2}
I^{131}[b]	1.5[b]	6.6[b]	3×10^{-2}[b]
Cs^{137}	0.2	25	1.1×10^{-1}
Ce^{144}	0.55	9.1	4.1×10^{-2}
Pm^{147}	0.033	150	6.9×10^{-1}
Tm^{170}	0.13	36	1.6×10^{-1}
Ir^{192}	0.2	25	1.1×10^{-1}
Po^{210}	22	0.23	1.0×10^{-3}
U^{233}[c]	61	0.082	3.7×10^{-4}
U^{235}	20	0.25	1.1×10^{-3}
Pu^{238}[d]	33	0.15	6.7×10^{-4}[d]
Pu^{239}[d]	30	0.17	7.6×10^{-4}[d]
Cm^{242}	26	0.19	8.7×10^{-4}
Cm^{244}	28	0.18	8.1×10^{-4}

[a] G. W. Dolphin et al., Accumulated Dose Received in 13 Weeks and 50 Years by Body Tissues from One Microcurie Single Intake by Inhalation or Injection Through a Wound, AHSB(RP)R 20 (1962).

[b] I^{131} based on 10 rems over 13 weeks to the thyroid and Kr^{85} based on immersion dose to lung of 5 rems assuming 10^{-5} c/m^3 = 2.5 mr/hr.

[c] Based on U^{233} containing 500 ppm U^{232}. Specific activity = 2×10^{-2} curie/g of $U^{233} + U^{232}$.

[d] The limit would be considerably lower for plutonium if soluble material is ingested and the bone is considered as the critical organ. The material dispersed in the accidents considered most likely, however, is insoluble PuO_2.

crisis of a few years ago). Similar variation to the above would occur in economic losses from urban contamination.

In order to estimate the potential economic loss from contamination of extensive areas, three ranges of contamination damage and economic loss are postulated. These, as well as the personnel exposure loss ranges assumed, are summarized in Table 3.2. It should be understood that the authors assumed the ranges of personnel exposure and area contamination with corresponding loss figures for purposes of obtaining an economic estimate for this study and that they may not correspond in any way to the values which would apply in case of an actual accident.

Table 3.2. Summary of Monetary Loss Assumptions for Personnel Exposure and Area Contamination

Range	Description	Assumed Loss
A	Assumed upper range of exposure	$50,000/person
B	Assumed intermediate range of exposure	$10,000/person
C	Assumed lower range of exposure	$ 2,000/person
I	Severe contamination Long-term evacuation Total loss of value No crops ~5 years	$10,000/person
II	Moderate contamination Short-term evacuation Moderate decontamination No crops 1-5 years	$ 1,500/person
III	Minor contamination No evacuation Minor decontamination Some crops destroyed	$0.005/m^2

In the highest contamination range (Range I) it is assumed that long-term evacuation and relocation and very extensive demolition-type decontamination operations would be required in urban areas. Also, evacuation and restrictions on agriculture of five years or greater would be necessary in rural areas. The economic loss in this range is assumed to be the total

value of the property involved; that is to say, the cost of decontamination would equal or exceed the total value of the property in the area contaminated. The loss would vary tremendously on an area basis in urban-vs-rural areas. The loss per capita, however, would be about the same for urban and rural areas.[4] In 1956, the total reproducible tangible assets and land in the United States was valued at $\$1.42 \times 10^{12}$ for a population of 1.68×10^{8} people.[5] Thus the average total assets are about $8500 per person. If another $1500 is assumed for relocation expenses and loss of income during relocation, the total loss per person on the average, would be about $10,000. Therefore, $10,000 per person will be assumed for the loss in Range I. The variation in per capita personal income throughout the country may be a good indication of the variation in the per capital value of property. Using per capita income[5] as a criterion, the loss in different sections of the United States might vary from 47% (Mississippi) to 139% (Connecticut) of the $10,000-per-person average assumed for Range I.

In the intermediate range of contamination (Range II) it is assumed that short-term evacuation and moderate decontamination would be required. For urban areas this might include washing nonporous surfaces with detergent, replacing or recovering porous surfaces such as sidewalks, pavements, roofs, etc. For rural areas this would include deep plowing and letting the land lie fallow for at least one season. In successive seasons there might be further restriction on the type of crop that could be raised. The potential economic loss assumed for this type of operation is $1500 per person.

In the lowest contamination range (Range III), it is assumed that the costs incurred would result from contamination surveys and possible minor decontamination operations (hosing roofs and streets in urban areas and destroying standing crops and milk in rural areas). Since evacuation would not be necessary and there would not be extensive destruction of property except for the crops, the costs would be dependent on the area contaminated and be independent of population density. Although the crop loss on an individual acre might be high, for example, $60 for wheat and $900 for tobacco, when an extensive area is contaminated the average loss per acre would be less than this. For example, based on the total area in the state, the average value of the Kansas 1958 wheat crop was $9.50 per acre, and the

average value of the North Carolina 1958 tobacco crop was $14 per acre. Plowing costs for decontamination has been estimated[6] at $30 per acre. Firehosing urban paved areas for decontamination has been estimated[6] to cost about $2.50 per thousand square feet. A cost of 5 mills per square meter has been assumed for Range III. This corresponds to a survey and decontamination cost of 46 cents per thousand square feet for urban areas and $20 per acre for the survey cost and crop loss for rural areas.

The contamination ranges used as a basis for calculating loss in this study are shown in Table 3.3. The basis used for choosing the various contamination ranges are discussed below. The reader is invited to revise the contamination and economic values assumed in this study to cover his situation or to make suitable adjustments as more data on the fate and effect of individual isotopes in the ecology become available.

Basis of Contamination Ranges for I^{131}, Sr^{90} and Cs^{137}

Following the Windscale accident (October 10, 1957) studies[7] were made on the I^{131}, Sr^{90} and Cs^{137} content of milk as a function of the contamination of the surrounding area. Studies had also been made on the soil-grass-milk Sr^{90} cycle[8] and of the uptake of Sr^{90} and Cs^{137} in crops[9] and milk.[8,10,11] At the time of that accident, recommendations were made[12] by the English Medical Research Council for the maximum permissible dietary contamination by I^{131}, Cs^{137}, and Sr^{90} after an accidental release in that country. These data and recommendations provide a basis for choosing contamination ranges for the isotopes studied.

At Windscale, the data[7] showed a constant ratio of I^{131} grass activity to milk activity. The relationship is: 1 $\mu c/m^2$(grass) = 0.09 μc/liter (milk). The recommendation[12] for the maximum I^{131} content of milk was 0.065 μc/liter. This would be produced on grass having a contamination level of about 7×10^{-7} curie/m^2. Although the ground gamma activity level after the accident decreased with the 8-day I^{131} half-life, the milk activity decreased with a 5 or 6 day half-life, probably due to some I^{131} becoming fixed. With this short half-life, long-term restrictions would not be a problem.

There are considerable data on the fate of Sr^{90} in the food cycle[9-11,13] and its distribution in the environment as a result of fallout.[10,14,15]

Table 3.3. Contamination Ranges Assumed for Purposes of This Report to Require Restrictions

Isotope	168-hr MPC[a] Minimum (c/cc)	$t_{1/2}$ (yr)	No Restrictions (curie/m^2)	Range III Minor Restrictions: No evacuation Minor decontamination Some crops destroyed (curie/m^2)	Range II Moderate Restrictions: Short-term evacuation Moderate decontamination No crops for ≥ 1 yr (curie/m^2)	Range I Severe Restrictions Long-term evacuation Total loss of value No crops for ≥ 5 yr (curie/m^2)
Co^{60}	3×10^{-9}	5.2	$<3 \times 10^{-5}$	3×10^{-5} to 3.4×10^{-4}	3.4×10^{-4} to 6×10^{-3}	$>6 \times 10^{-3}$
Sr^{90}[a]	10^{-10}	27.7	$<1 \times 10^{-7}$	1×10^{-7} to 1×10^{-6}	1×10^{-6} to 1.1×10^{-5}	$>1.1 \times 10^{-5}$
Sb^{124}	7×10^{-9}	0.167	$<7 \times 10^{-5}$	7×10^{-5} to 4.6×10^{-2}	$>4.6 \times 10^{-2}$	-
I^{131}[a]	3×10^{-9}	0.022	$<6.5 \times 10^{-7}$	$>6.5 \times 10^{-7}$	-	-
Cs^{137}[a]	5×10^{-9}	26.6	$<5 \times 10^{-6}$	5×10^{-6} to 5×10^{-5}	5×10^{-5} to 5×10^{-4}	$>5 \times 10^{-4}$
Ce^{144}	2×10^{-9}	0.78	$<2 \times 10^{-5}$	2×10^{-5} to 1.4×10^{-3}	1.4×10^{-3} to 1.2	>1.2
Pm^{147}	2×10^{-8}	2.6	$<2 \times 10^{-4}$	2×10^{-4} to 2.7×10^{-3}	2.7×10^{-3} to 9.6×10^{-2}	$>9.6 \times 10^{-2}$
Tm^{170}	1×10^{-8}	0.354	$<1 \times 10^{-4}$	1×10^{-4} to 7×10^{-2}	$>7 \times 10^{-2}$	-
Ir^{192}	9×10^{-9}	0.203	$<9 \times 10^{-5}$	9×10^{-5} to 2.7×10^{-2}	$>2.7 \times 10^{-2}$	-
Po^{210}	7×10^{-11}	0.378	$<7 \times 10^{-7}$	7×10^{-7} to 4.5×10^{-3}	4.5×10^{-5} to 3.6	>3.6
U^{233}[b]	1.4×10^{-11}	7.7×10^{4}	$<1.4 \times 10^{-7}$	1.4×10^{-7} to 4×10^{-6}	1.4×10^{-6} to 1.4×10^{-5}	$>1.4 \times 10^{-5}$
U^{235}	4×10^{-11}	7×10^{8}	$<4 \times 10^{-7}$	4×10^{-7} to 4×10^{-6}	4×10^{-6} to 4×10^{-5}	$>4 \times 10^{-5}$
Pu^{238}	7×10^{-13}	86.4	$<7 \times 10^{-9}$	7×10^{-9} to 7×10^{-8}	7×10^{-8} to 7×10^{-7}	$>7 \times 10^{-7}$
Pu^{239}	6×10^{-13}	2.4×10^{4}	$<6 \times 10^{-9}$	6×10^{-9} to 6×10^{-8}	6×10^{-8} to 6×10^{-7}	$>6 \times 10^{-7}$
Cm^{242}	4×10^{-11}	0.444	$<4 \times 10^{-7}$	4×10^{-7} to 1.9×10^{-5}	1.9×10^{-5} to 4.5×10^{-1}	$>4.5 \times 10^{-1}$
Cm^{244}	3×10^{-12}	17.9	$<3 \times 10^{-8}$	3×10^{-8} to 3×10^{-7}	3×10^{-7} to 4×10^{-6}	$>4 \times 10^{-6}$

[a] Contamination levels for these isotopes based on dairy farming. Limits for all other isotopes based on resuspension and inhalation.

[b] Based on U^{233} containing 500 ppm U^{232}. Specific activity = 2×10^{-2} curie/g $U^{233} + U^{232}$.

The data on the uptake in milk,[8] which also includes the effect of sequential plowing on Sr^{90} uptake, probably provides the best basis for choosing Sr^{90} contamination ranges. At a contamination level of 5550 μμc/m^2 and two years of plowing and reseeding, the uptake ratio of Sr^{90} to calcium in grass was 20 μμc of Sr^{90} per gram of calcium. A further decrease in the Sr^{90}/Ca ratio by a factor of 2 might be obtained by the application of lime. (This would depend on the original calcium content of the soil. Unless the soil were highly calcium deficient, unlikely in good farming practice, a much higher factor could not be expected by limeing.) In the conversion from grass to milk, there is a further discrimination in calcium over strontium by a factor of about 10 (refs. 8, 16). Thus, the milk produced on contaminated land that has been limed and plowed on successive years (long-term uptake in milk, in other words) would be 1.8×10^5 mμc of Sr^{90} per gram of calcium (in milk) per curie per square meter of ground. The recommended limit[12] on Sr^{90} in milk for long-term intake is 2 mμc of Sr^{90} per gram of calcium. Thus the contamination limits for long-term restriction, Range I, would be 1.1×10^{-5} curie/m^2. The uptake of Sr^{90} from ground that has not been plowed on successive years was about 5 times higher.[8] It is assumed that it would take perhaps five years to realize the factor-of-5 benefit from repeated plowing, and the factor-of-2 benefit from limeing. In addition, during the five years, radioactive decay would yield an added factor of 1.1. Combining these factors yields a factor of 11 between long-term and one year's restrictions. One year's restriction, Range II, would thus result from contamination levels 91% lower than long-term restrictions. Short-term restrictions, Range III, are assumed to occur at a factor of 100 lower than Range I levels.

The equations developed by Russell and co-workers[17] for calculating the concentration of Sr^{90} in milk from current and previous fallout furnish an independent basis for estimating Sr^{90} contamination ranges. These equations are:

$$\text{Short term:} \quad C = D_p + 0.23\, D_t,$$

$$\text{Long term:} \quad C = 1.1\, D_p + 0.14\, D_t,$$

where

C = μμc of Sr^{90} per gram of calcium in milk,

D_p = mc/km^2/year of Sr^{90} deposited,

D_t = mc/km^2 of Sr^{90} in soil.

The first term, D_p, is the contribution of fallout on the grass, and the second term, D_t, is the contribution due to uptake from the ground. The distinction between short term and long term is the reduced uptake as a result of plowing and leaching over several years.

A comparison of the contamination ranges based on the Morgan[8] and Russell[17] data is shown in Table 3.4. In using the Russell equations for calculating the values shown in Table 3.4, an allowable milk concentration, C, of 2000 μμc Sr^{90} per gram of calcium was used; for Range I and II there is assumed to be no uptake from newly deposited material, i.e., $D_p = 0$; and for Range III the effect from a single release $Q(c/m^2)$, was assumed to be equivalent to a continuous release of that material over a month, and ground uptake was neglected (i.e., $D_p = 1.2 \times 10^{-10} Q$ and $D_t = 0$). The contamination ranges calculated from the two sets of data are surprisingly consistent. The ranges based on the Morgan data have been used in this study.

Table 3.4. Comparison of Sr^{90} Contamination Ranges Using Alternate Bases

	Morgan Data[8] Basis (curie/m^2)	Russell Equation[17] Basis (curie/m^2)
Range I	$>1.1 \times 10^{-5}$	$>1.4 \times 10^{-5}$
Range II	$>1.0 \times 10^{-6}$	$>8.7 \times 10^{-6}$
Range III	$>1.0 \times 10^{-7}$	$>1.6 \times 10^{-7}$

Range III for Sr^{90} is from 2200 dis min^{-1} 100 cm^2 to 22,000 dis min^{-1} 100 cm^2. For comparison, the ORNL standard[18] for unrestricted areas is 1000 dis min^{-1} 100 cm^2 transferable beta contamination.

The recommended[12] maximum concentration of Cs^{137} in milk is 150 mμc/liter for children. At Windscale[7] the measured ratio of Cs^{137} in grass to Cs^{137} in milk was about 4 mμc per square meter of grass per mμc per liter of milk. Based on this ratio, a milk contamination level of 150 mμc/liter would result from a grass contamination level of 600 $m\mu c/m^2$. The average ratio of the Cs^{137} content in clover to the Cs^{137} content of

ground in the plant uptake studies[9] was 1120 mμc in the clover per mc in the ground. Assuming the ratio to be constant, a ground contamination of 5×10^{-4} curie/m^2 would produce grass having a content of 600 mμc/m^2, and milk with a content of 150 mμc/liter. Since Cs^{137} has little mobility in soil, contamination at these levels or above would lead to long-term (Range I) restriction. It is assumed that one-year restrictions (Range II) would result from contamination a tenth as great and that short-term restrictions (Range III) would result from contamination only a hundredth as great. To illustrate the rapid decrease in grass contamination with time and the extremely high initial contamination for Ranges I and II, the postulated grass contamination levels for Sr^{90} and Cs^{137} are tabulated as a function of time after the incident for the three ranges in Table 3.5.

Contamination-Level Basis for Other Isotopes

For other isotopes, where plant uptake data are not available, a resuspension factor has been used in combination with the maximum permissible concentration in air (MPC_a) to establish a lower contamination level for Range III. Ranges II and I are then calculated based on a model developed from the Sr^{90} and Cs^{137} plant uptake studies.

Experiments[19] have indicated a resuspension factor of 4×10^{-5} units per cubic meter in air per 1 unit per square meter of surface activity for a dusty operation in a confined space. In uranium feed plants, factors of 2×10^{-3} to 2.5×10^{-5} have been measured.[20] Other experiments and documented plant experience[21] have been given values varying from 2×10^{-3} to 4×10^{-5}, with more extensive areas involved. Oak Ridge National Laboratory data[22] range from 1.3×10^{-5} for partially cleaned contaminated surfaces to 1.7×10^{-7} for surfaces completely scrubbed but unpainted. Based on experiments on the wind pickup of particles on various types of ground, resuspension factors of 7×10^{-2} (1.5-μ particles) to 8×10^{-5} (14-μ particles) can be predicted[23] at a wind speed of 5 m/sec. Resuspension factors have been measured for ground contaminated by plutonium distributed by a non-nuclear explosion.[24] Average resuspension factors from 2.5×10^{-6} to 1×10^{-7} from 4 to 160 days past explosion, respectively, result from ground contaminated to 10 μg/m^2. These data show increasing resuspension factors with decreasing contamination levels as well as a day-to-day

Table 3.5. Postulated Grass Contamination Ranges for Sr^{90} and Cs^{137}

Grass Contamination Ranges for Milk Restrictions	$\frac{Sr^{90}}{20\ m\mu c/g\ Ca}$	$\frac{Cs^{137}}{600\ m\mu c/m^2}$
Range III		
Deposited	1×10^{-7} to 1×10^{-6} c/m^2	5×10^{-6} c/m^2 to 5×10^{-5} c/m^2
Grass contamination		
Immediately	100 to 1000 mμc/g Ca	5×10^3 to 5×10^4 mμc/m^2
After one year	2.0 to 20 mμc/g Ca	60 to 600 mμc/m^2
Long term	0.2 to 2.0 mμc/g Ca	6 to 60 mμc/m^2
Range II		
Deposited	1×10^{-6} to 1×10^{-5} c/m^2	5×10^{-5} to 5×10^{-4} c/m^2
Grass contamination		
Immediately	1000 to 10,000 mμc/g Ca	5×10^4 to 5×10^5 mμc/m^2
After one year	20 to 200 mμc/g Ca	600 to 6000 mμc/m^2
Long term	2.0 to 20 mμc/g Ca	60 to 600 mμc/m^2
Range I		
Deposited	$>1.1 \times 10^{-5}$ c/m^2	$>5 \times 10^{-4}$ c/m^2
Grass contamination		
Immediately	>11,000 mμc/g Ca	$>5 \times 10^5$ mμc/m^2
After one year	>200 mμc/g Ca	>6000 mμc/m^2
Long term	>20 mμc/g Ca	>600 mμc/m^2

variation in resuspension factor by a factor of 50. Taking these variations into account at a plutonium contamination level of 6×10^{-9} curies/m^2, the resuspension factor would be between 10^{-4} and 2×10^{-6} at 4 days and 5×10^{-5} and 10^{-6} at 160 days.

For purposes of this study an assumed resuspension factor of 10^{-4} (curies/m^3)/(curies/m^2) or 10^{-4} (μc/cc)/(curies/m^2) has been used in conjunction with the 168-hr MPC_a (ref 25) as a basis for calculating the lower contamination levels of Range III for isotopes other than Sr^{90}, I^{131}, and Cs^{137}. It is assumed that some restrictions but no evacuation (Range III) would be required at a contamination level that would result in the air being above MPC_a, i.e., the lower limit chosen for Range III is $10^4 \times MPC_a^{168}$, curies/m^2, for all isotopes other than Sr^{90}, I^{131} and Cs^{137}.

This basis is less restrictive than the food chain basis for Sr^{90}, I^{131} and Cs^{137} (the factors are a tenth for Sr^{90}, a forty-fifth for I^{131}, and a tenth for Cs^{137}); they therefore may be liberal rather than conservative for the other isotopes. For plutonium, this results in Range III lower contamination level of 130 dis min^{-1} 100 cm^{-2} (6×10^{-9} curies/m^2) which compares with a level of 30 dis min^{-1} 100 cm^{-2} average standard for unregulated areas at ORNL[20] and 500 dis min^{-1} 100 cm^{-2} maximum allowable on any surface of a shipping container in interstate commerce.[26]

In choosing ranges for longer-term restriction, radioactive decay must be considered in addition to the normal decrease in availability due to movement or fixation in the soil (see Table 3.4). Five years' decay would decrease the contamination level a factor of $2^{(5/t_{1/2})}$ (where $t_{1/2}$ = half-life in years), and one year's decay would decrease the level by a factor of $2^{(1/t_{1/2})}$. An increase in contamination by factors of 10 and 100 were assumed based on the Cs^{137} and Sr^{90} data from Range III to Range II and Range I, respectively, for movement in soil or fixation. Combining these with the decay fraction yields:

$$\frac{\text{Range II}}{\text{Range III}} = 10 \times 2^{(1/t_{1/2})},$$

$$\frac{\text{Range I}}{\text{Range III}} = 100 \times 2^{(5/t_{1/2})}.$$

These equations were used to calculate the lower limits for Range II and Range I based on Range III (see Table 3.3). These equations are consistent with the ratios used for Sr^{90}, I^{131} and Cs^{137}.

Much higher contamination levels would probably be more consistent with actual biological injury. There is likely to be considerable public pressure, however, in the event of a major accident, to take protective action at even lower levels. Thus, although the postulated losses may appear to be highly overestimated relative to actual injury, the exigencies of any actual situation may well prove the estimates to have been too low.

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4. ASSUMPTIONS REGARDING THE SITE AND THE DISTRIBUTION OF THE SURROUNDING POPULATION

Because of economics, large nuclear power reactors must be at least relatively near a large electric load center and have a large supply of cooling water. This severely restricts their siting and almost invariably dictates that they be built near large population centers. The designer and builder of a radiochemical facility has much more flexibility in choosing a site. These plants can be built in remote locations with less economic penalty, although they are secondarily tied to power reactors as both a customer for fuel elements and a source of isotopes.

The population density distribution of nonemployees surrounding typical existing sites,[1] and the population distributions assumed for this study are shown in Fig. 4.1. Case I, defined as a "typical population distribution," closely approximates the population-distribution assumptions of report WASH-740.[2] For case I, the integrated average population density p/a (persons per square meter) with a distance x (meters) of the site may be expressed mathematically as:

$$p/a = 1.93 \times 10^{-4} \left(1 - e^{-0.895 \times 10^{-4} x}\right).$$

Case II, a uniform population distribution of 100 persons per square mile, allows facile conversion of the damage values calculated in this report to any other uniform population distribution.

The integrated average population density as a function of radius from the site was used in calculating the damage for the typical population distribution. This allows approximate determination of the number of persons within an exposure or contamination isopleth, with only a knowledge of the area within the isopleth and the maximum downwind distance of the isopleth. A more accurate method, integration of the population density over the plume width as a function of distance from the site, would involve incorporation of a complicated population-distance function into the meteorological equations. This more rigorous approach is not justified in view of the uncertainities in the exposure and contamination levels.

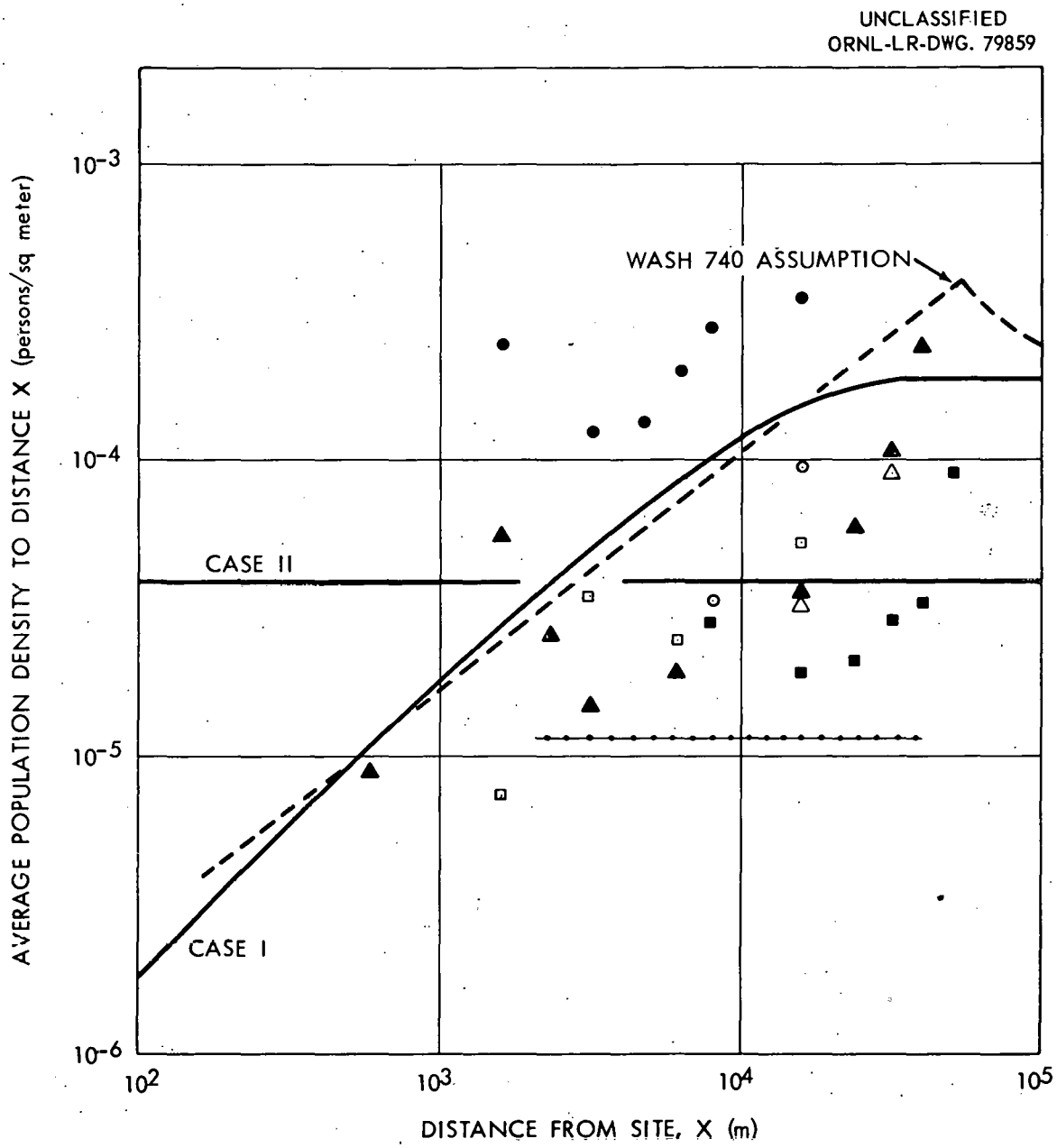

Fig. 4.1. Population Density Distribution for Typical Sites and Assumptions for Present Study. Case I - typical population distribution. Case II - uniform population density of 100 persons per square mile.

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5. MAXIMUM CREDIBLE VALUES OF CONCENTRATION, FALLOUT, AND WASHOUT FOR RELEASE OF RADIOISOTOPES

The calculations performed in obtaining the values of concentration, deposition, and washout in this paper were based on observational evidence. Little significant difference will be found from the values and methods outlined in WASH-740 (ref 1), which are based on Sutton's parameters. The approach used here, however, as developed by Gifford and Culkowski and Hilsmeier,[2-5] uses empirical weather parameters based on actual measurements rather than the unmeasurable "n" and "c" values of most diffusion equations. This is a step toward a more realistic description and evaluation of the effect of meteorological conditions. The maximum rate of deposition for a given release has been calculated. This is a function of such weather conditions as precipitation, cloudiness, and wind speed, which are elements of climatological record in almost every area.

Atmospheric Concentration of Radioisotopes After an Accidental Release

The concentration of effluent, assuming no depletion by deposition or washout, can best be described by the simple Gaussian formula:[3]

$$\chi = \frac{Q}{\pi \sigma_y \sigma_z \bar{u}} \exp\left[-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2} + \frac{h^2}{\sigma_z^2}\right)\right], \qquad (1)$$

where

χ = concentration in grams or curies per cubic meter,

Q = source strength in grams of curies per second,

$\bar{u}$ = mean wind speed in meters per second,

y = crosswind distance in meters from the plume axis, which is assumed to coincide with the mean wind direction,

h = source height in meters,

σ_y^2, σ_z^2 = dispersion coefficients in square meters.

For convenience, the three variables $\bar{u}$, χ, and Q are lumped together as $\bar{u}\chi/Q$ (herein called the concentration parameter). Hilsmeier and Gifford[4] published a set of Graphs for Estimating Dispersion Parameters from which Table 5.1 and Figs. 5.1 through 5.6 are taken. Table 5.1 is a list of meteorological stability categories. For the nonmeteorologist, the columns

Table 5.1. Meteorological Stability Categories

A: Extremely unstable conditions
B: Moderately unstable conditions
C: Slightly unstable conditions
D: Neutral conditions[a]
E: Slightly stable conditions
F: Moderately stable conditions

Surface Wind Speed (m/sec)	Daytime Insolation			Thin Overcast or ≤4/8 Cloudiness[b]	≥3/8 Cloudiness
	Strong	Moderate	Slight		
<2	A	A-B	B		
2	A-B	B	C	E	F
4	B	B-C	C	D	E
6	C	C-D	D	D	D
>6	C	D	D	D	D

[a]Applicable to heavy overcast, day or night.

[b]The degree of cloudiness is defined as that fraction of the sky above the local apparent horizon which is covered by clouds. /Manual of Surface Observations (WBAN), Circular N(7th ed.), paragraph 1210, USGPO, Washington, July 1960./

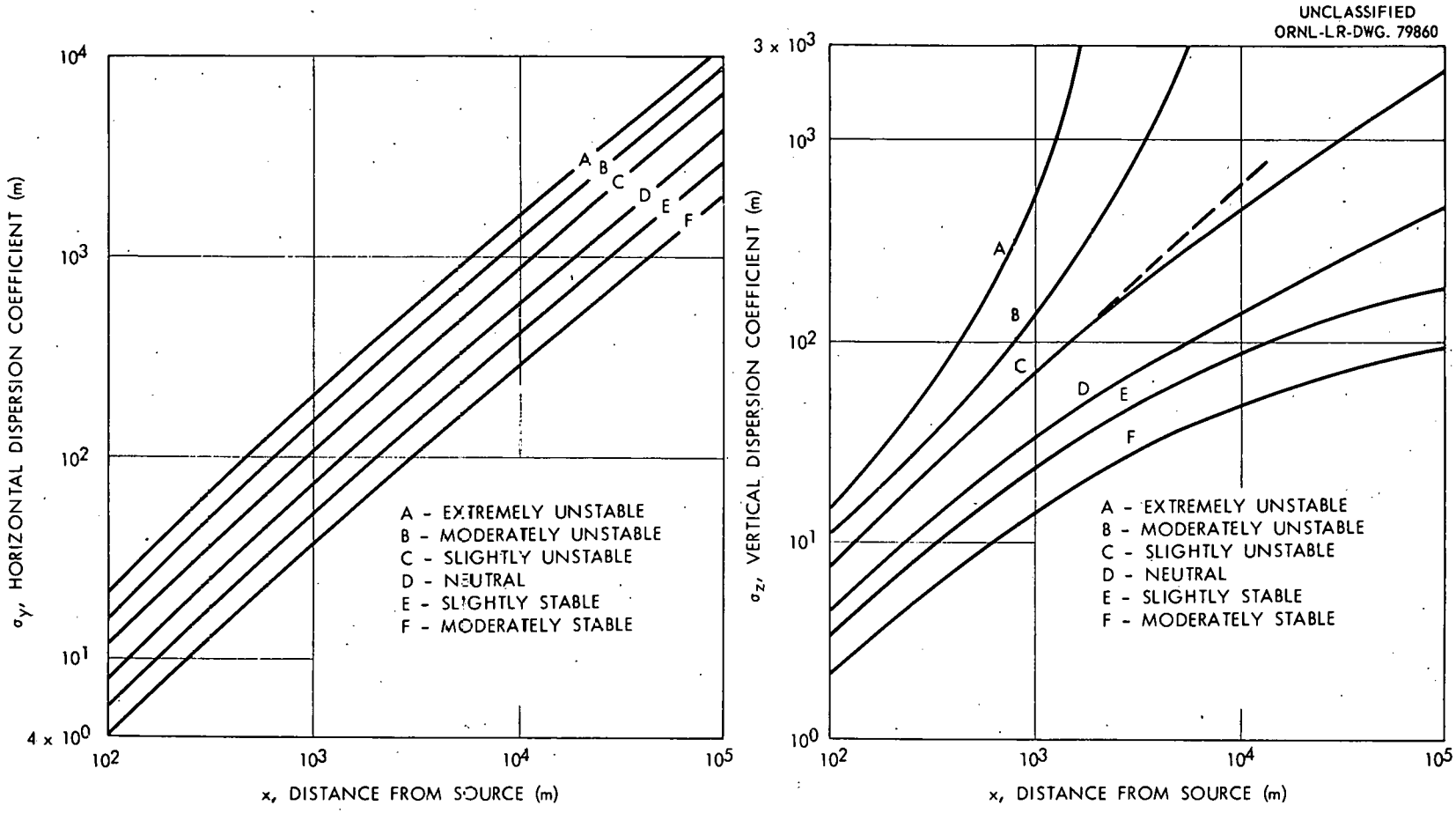

Fig. 5.1. Horizontal and Vertical Dispersion Coefficients as a Function of Downwind Distance for Various Meteorological Stability Categories.

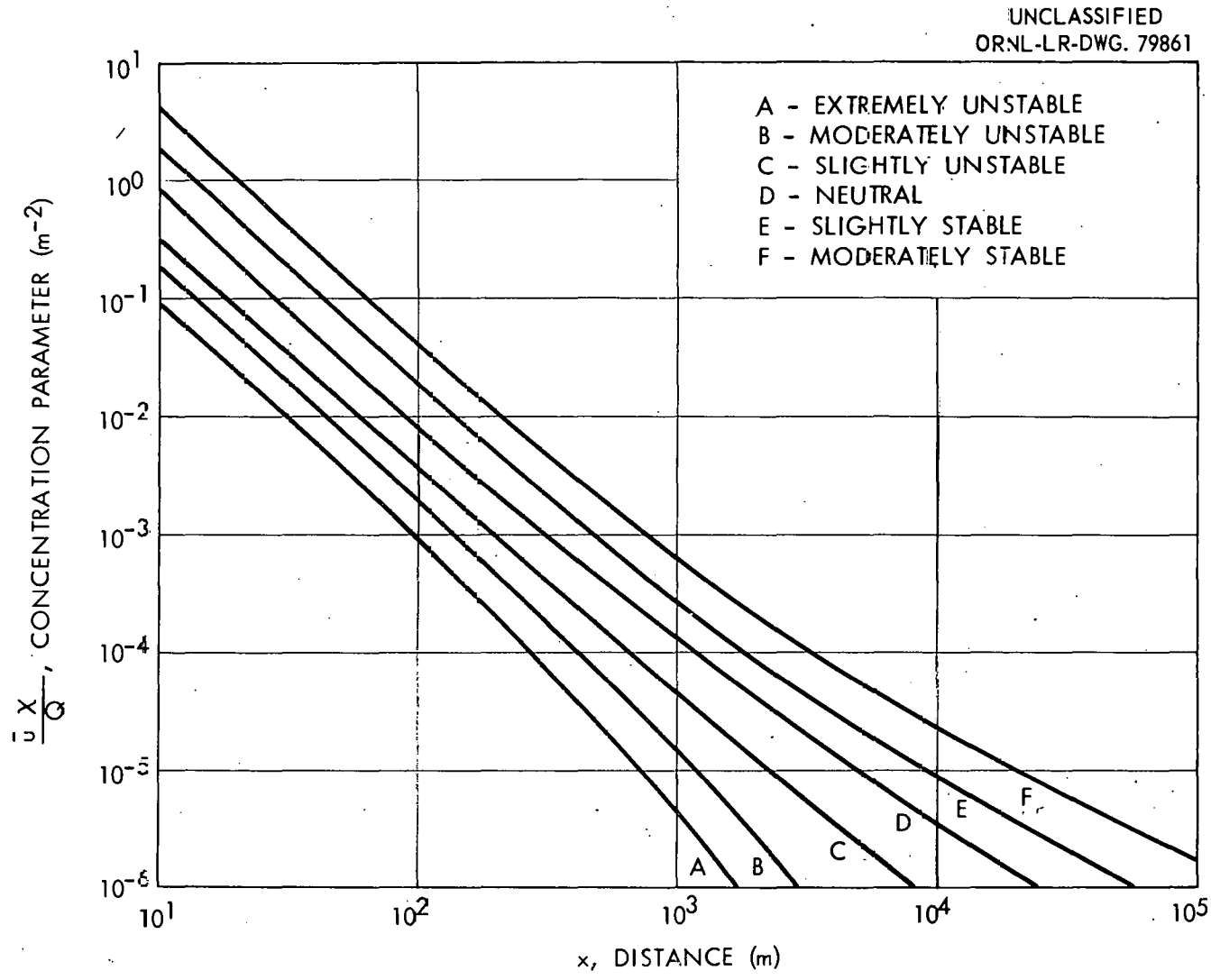

Fig. 5.2. Concentration Parameter as a Function of Downwind Distance for Various Meteorological Stability Categories. Source at Surface.

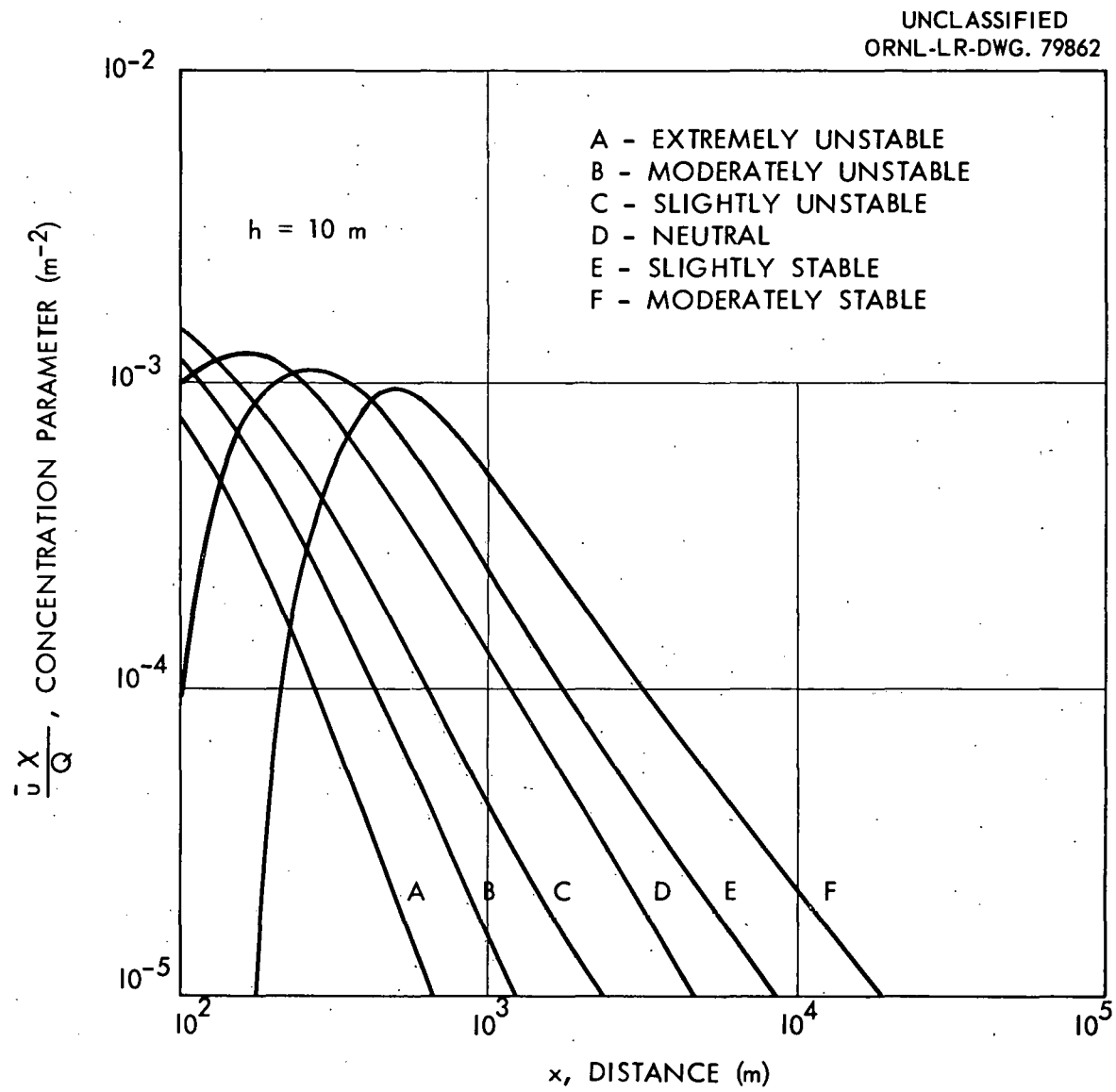

Fig. 5.3. Concentration Parameter as a Function of Downwind Distance for Various Meteorological Stability Categories. Source at 10 m.

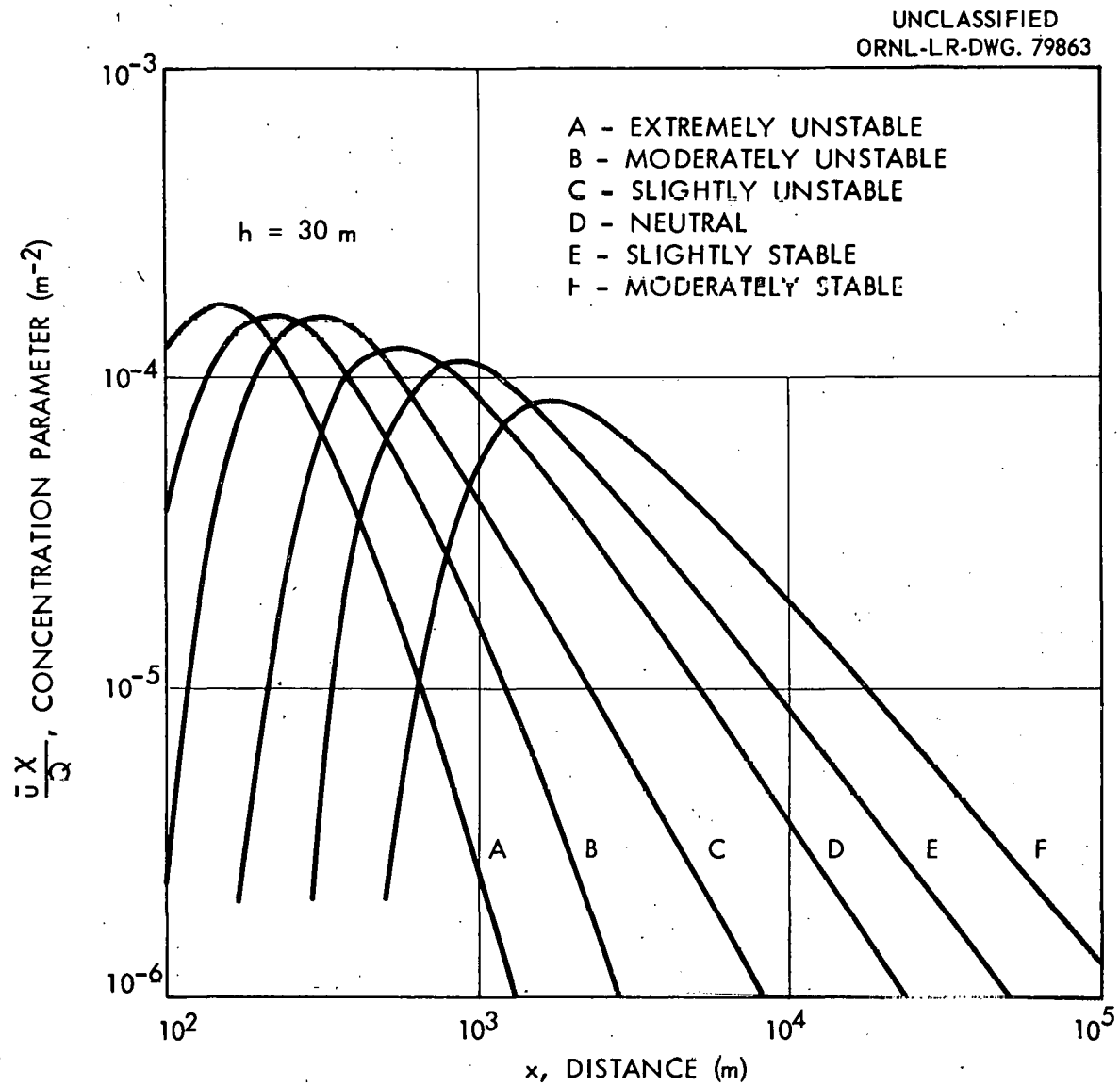

Fig. 5.4. Concentration Parameter as a Function of Downwind Distance for Various Meteorological Stability Categories. Source at 30 m.

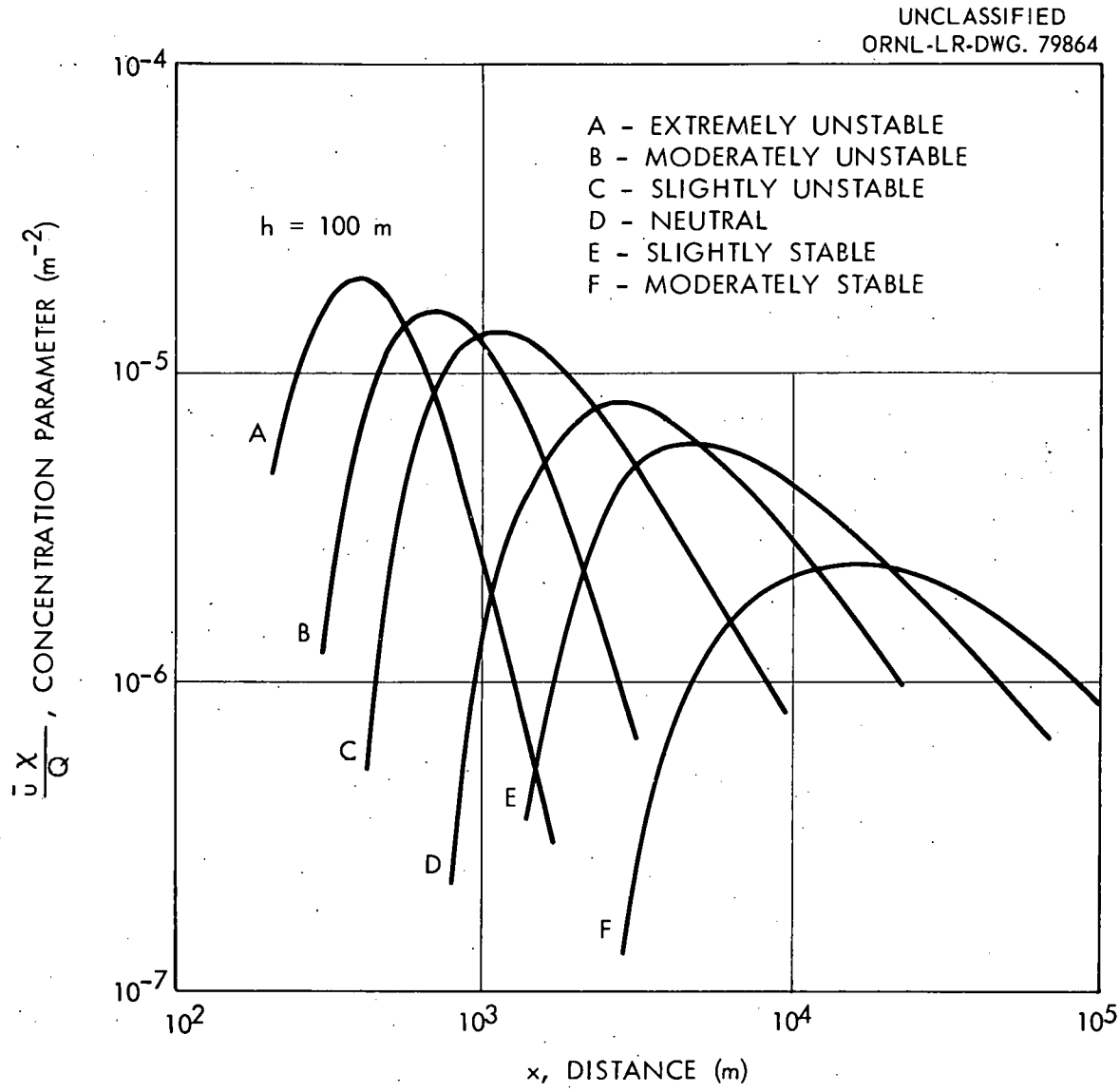

Fig. 5.5. Concentration Parameter as a Function of Downwind Distance for Various Meteorological Stability Categories. Source at 100 m.

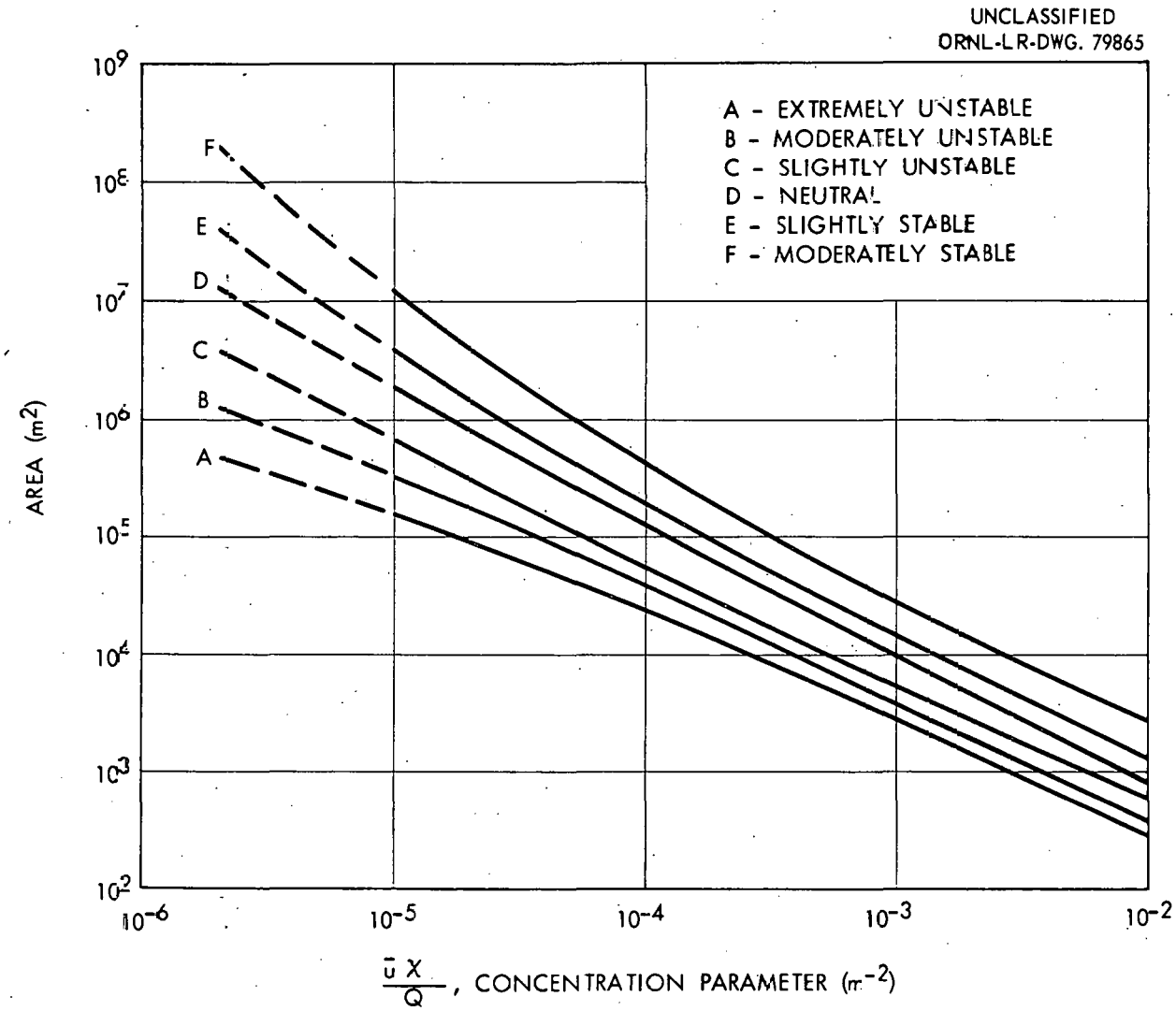

Fig. 5.6. Area Within Isopleths of the Concentration Parameter for Various Meteorological Stability Categories. Source at surface.

marked "daytime insolation" which are subdivided into "strong," "moderate," and "slight" can be read roughly as "clear (or fair)," "partly cloudy," and "cloudy," respectively. Figure 5.1 presents the recommended values of dispersion coefficients. Graphs of the concentration parameter as a function of the downwind distance (on the plume centerline) and source height, Figs. 5.2 through 5.5, were prepared by using Eq. (1) and these dispersion coefficients. Figure 5.6 is a plot of the area within isopleths of the concentration parameter for the six stability categories assuming a ground level release.

Deposition of Radioactive Materials After an Accidental Release

Deposition of radioactive material on the ground or other surfaces presents two problems: (1) long-term environmental buildup and (2) localized nonuniformity of surface contamination from accidental or intentional high-volume releases of effluent. The first problem is dealt with by Culkowski.[6] The second problem, that of high-volume anomalous releases, is of interest in considering the maximum credible amount of land contaminated beyond a given level of activity.

Basically, deposition is obtained by multiplying the concentration X by a coefficient V_g, known as the "velocity of deposition."[7] It is of interest to note that V_g has the <u>units</u> of velocity, but this need not indicate a true <u>settling</u> velocity. The units of V_g are obtained by dividing the parts depositing per unit area by the parts per unit volume of concentration adjacent to the area. Thus, a gas impacting on or filtering across a membrane may have a large V_g, whereas a large irregularly shaped particle of low density may have a small V_g. Obviously, V_g's are determined experimentally.

Gifford and Pack[8] recently published an evaluation of most of the experimental data obtained to date on deposition velocities of materials of interest in nuclear safety studies. Major conclusions were that the deposition velocity for reactive materials such as I^{131}, SO_2, and ruthenium on flat plates or bare soil is less than 0.01 m/sec and is about 0.01 to 0.03 m/sec for deposition on vegetation; the average deposition velocity of inert materials such as Cs^{137} and Sr^{90} on flat plates and vegetation is less than 0.001 and about 0.001 to 0.002 m/sec, respectively. These

results are remarkably consistent, indicating that for particles less than 10 to 15 μ in diameter (which are of primary interest in nuclear hazard studies), the relatively nonvariant effects of impaction, diffusion, and adsorption are more important than the widely varying gravitational settling velocities.

The deposition rate may be calculated from a known air concentration by:

$$Dep = \chi V_g. \tag{2}$$

The equation of continuity is utilized to account for depletion of the plume:

$$\int_0^\infty \int_0^\infty \chi V_g \, dy \, dx = Q. \tag{3}$$

Equation (3) is analytic for the first integration, but the resulting equation, from (1),

$$1 = \int_0^\infty \frac{V_g}{\bar{u}\sigma_z\sqrt{\pi}} \exp - \frac{1}{2}\left[\frac{h^2}{\sigma_z^2}\right] dx, \tag{3a}$$

can be solved analytically only if σ_z can be expressed in terms of x, which Fig. 5.1 shows to be difficult, to say the least.

To arrive at an accurate approximation, Eq. (3) was computed iteratively. The results for deposition velocities of 0.01 and 0.001 m/sec are shown in Figs. 5.7 through 5.9. Figure 5.7 shows the concentration parameter $\bar{u}\chi/Q$ and the deposition parameter $\bar{u}\chi V_g/Q$ as a function of downwind distance for a deposition velocity of 0.01 m/sec. Figures 5.8 and 5.9 show the area enclosed within isopleths of the concentration and deposition parameter for deposition velocities of 0.01 and 0.001 m/sec.

Washout of Radioactive Isotopes After an Accidental Release

Washout (termed rainout in report WASH-740), is the removal of effluent from a plume or cloud by the scavenging action of raindrops as they fall. This is by far the most important factor in estimating maximum surface contamination. Since the scavening occurs throughout the depth of the plume, height of plume rise, vertical plume dispersion, and deposition characteristics can be ignored. Three parameters - the scavenging rate Λ, the wind

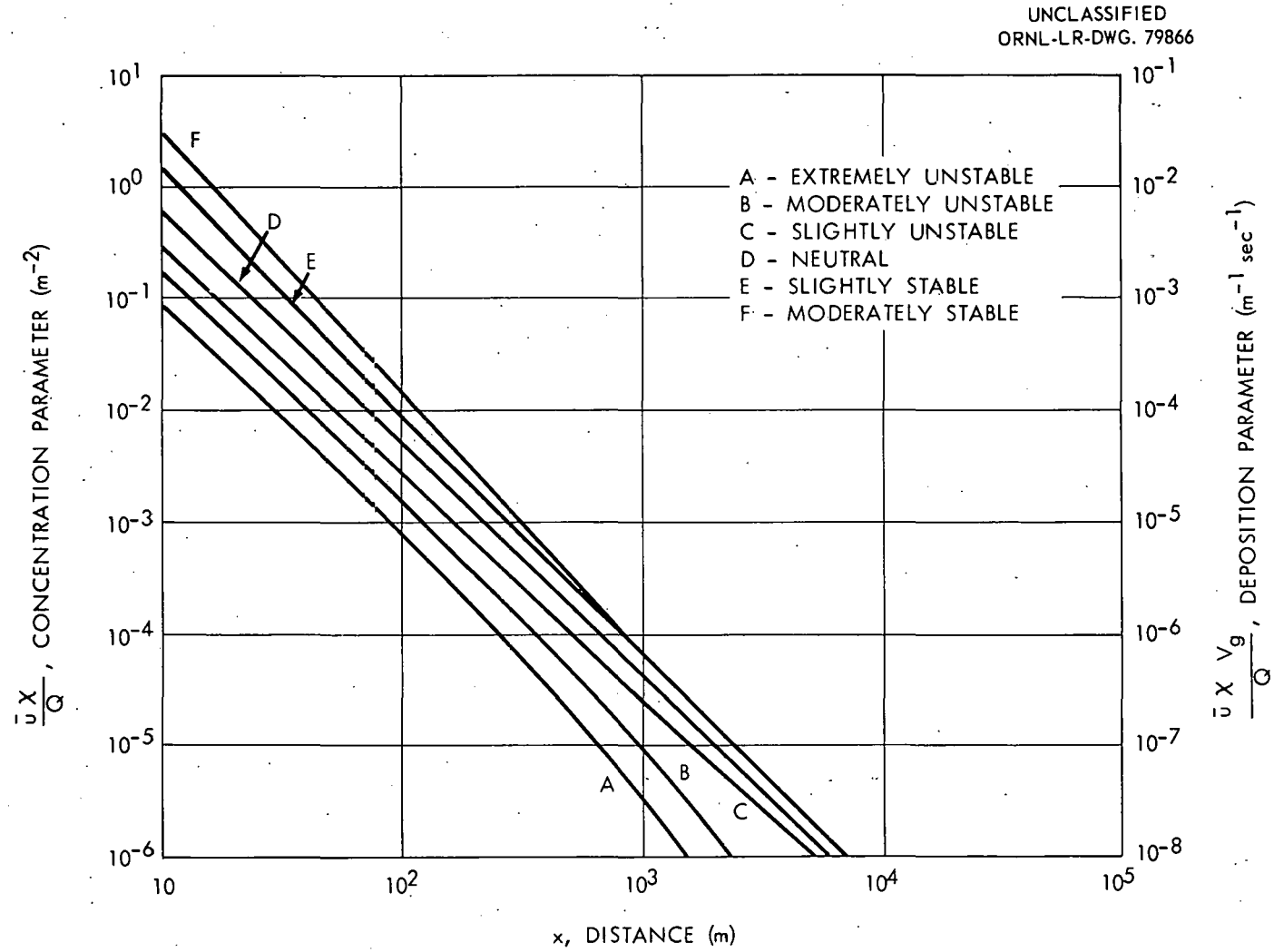

Fig. 5.7. Concentration and Deposition Parameter as a Function of Downwind Distance for Various Meteorological Stability Categories. Ground level release. Deposition velocity, 0.01 m/sec.

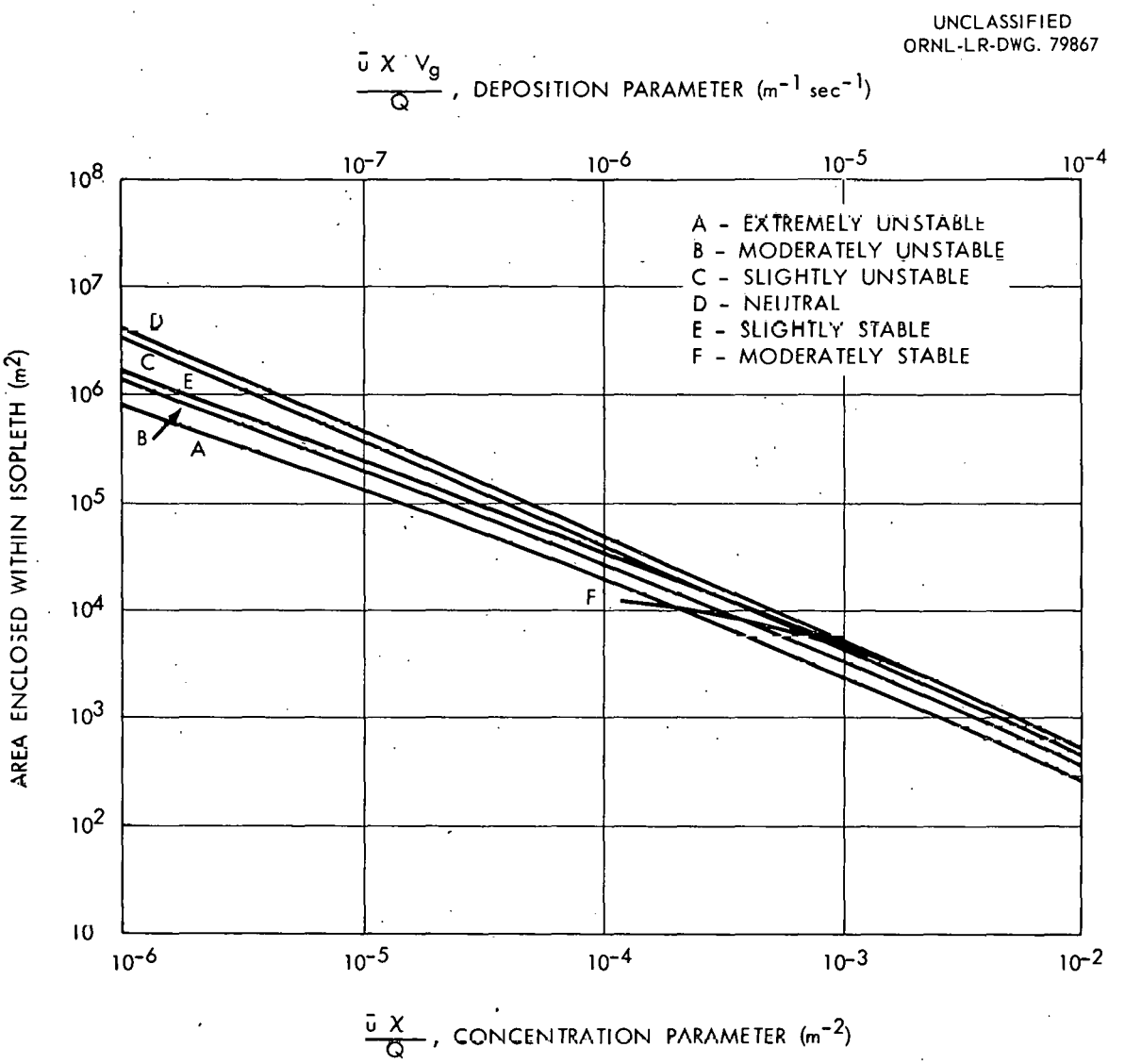

Fig. 5.8. Area Enclosed Within Isopleths of the Concentration and Deposition Parameter for Various Meteorological Stability Categories. Ground level release. Deposition velocity, 0.01 m/sec.

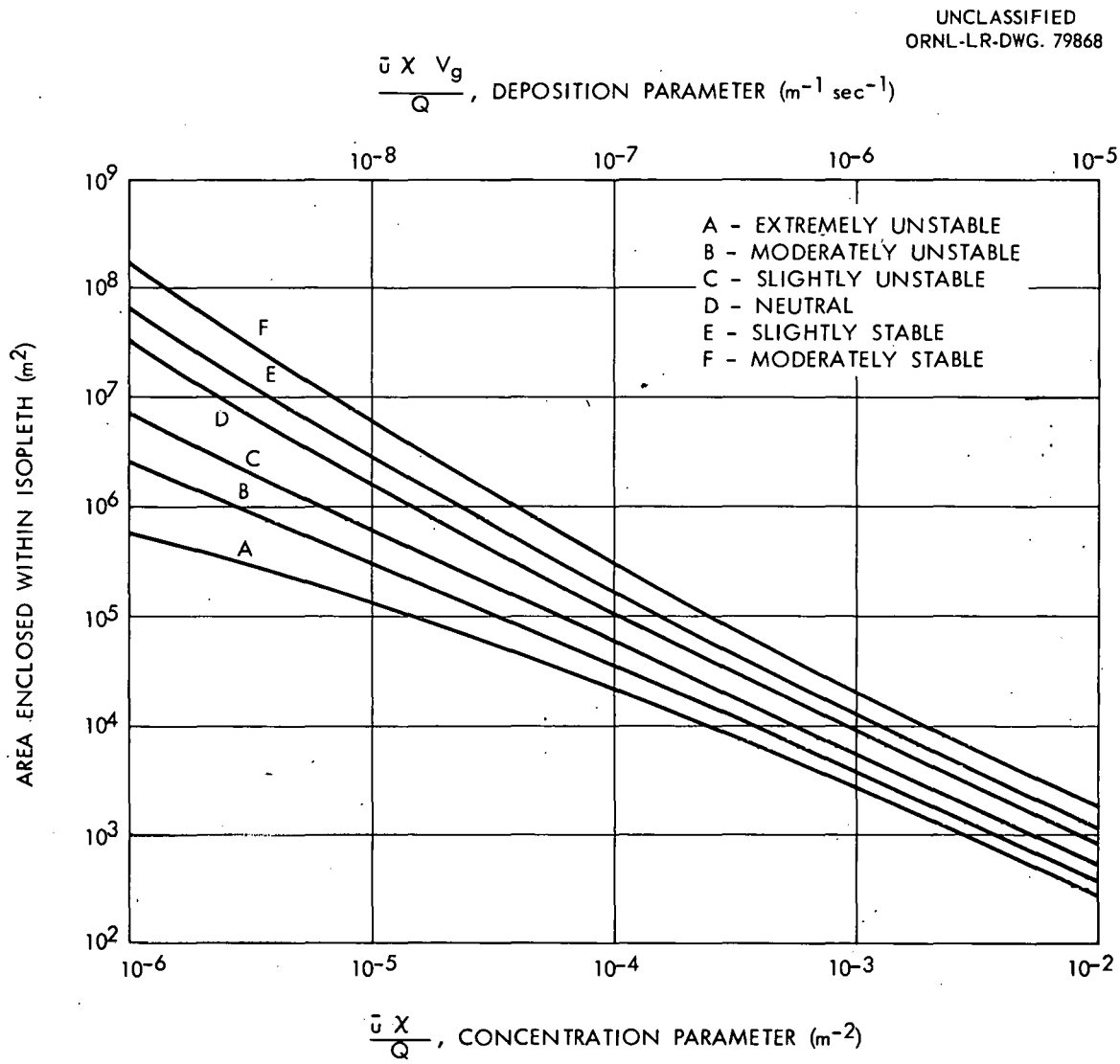

Fig. 5.9. Area Enclosed Within Isopleths of the Concentration and Deposition Parameter for Various Meteorological Stability Categories. Ground level release. Deposition velocity, 0.001 m/sec.

speed, and the horizontal dispersion coefficient σ_y control the area contamination from the washout process.

The removal of particulate matter by rain is analogous to radioactive decay; that is to say, it is completely random. Accordingly, the remaining material Q after a time t is:

$$Q = Q_o e^{-\Lambda t}, \tag{4}$$

where Λ is the washout coefficient, depending upon rainfall rate, and coalescent factors of particles. Employing Eq. (4), its derivative, and Eq. (1), we find that the rate of deposition ω may be written as:

$$\omega = \frac{\Lambda Q_o e^{-\Lambda x/\bar{u}}}{\bar{u}\sqrt{2\pi}\,\sigma_y} \exp - \left(\frac{y^2}{2\sigma_y^2}\right) \tag{5}$$

Solving Eq. (5) for the maximum ω at any given distance x, we find for ω_{max}:

$$\Lambda = \frac{\bar{u}}{x}. \tag{6}$$

Substituting Eq. (6) in (5) we have:

$$\omega_{max} = \frac{Q_o}{ex\sigma_y\sqrt{2\pi}} \exp - \left(\frac{y^2}{2\sigma_y^2}\right)$$

Calculations show that little is to be gained by using Eq. (5). After numerous calculations, assumptions of the value of Λ, etc., the values of ω vs (x,y) obtained by using Eqs. (5) and (7) are not significantly different. Use of Eq. (7), however, obviates assumptions of Λ and $\bar{u}$ and yields the maximum possible dimensions for ω, x, and y. Equation (7) will provide overestimates of the "close in" washout area for any given isopleth, as compared with Eq. (5). However, the error contributed to the total area is only about 0.01%.

Figure 5.10 shows the maximum area and maximum downwind distance vs deposition. A type "D" condition (heavy overcast day or night) was elected as typical of a rainy day (or night) and as the condition that would maximize the deposition area.

Should a calculation of a particular area or distance vs a specific weather condition be desired, Eq. (5) should be used. Table 5.2 (ref 9)

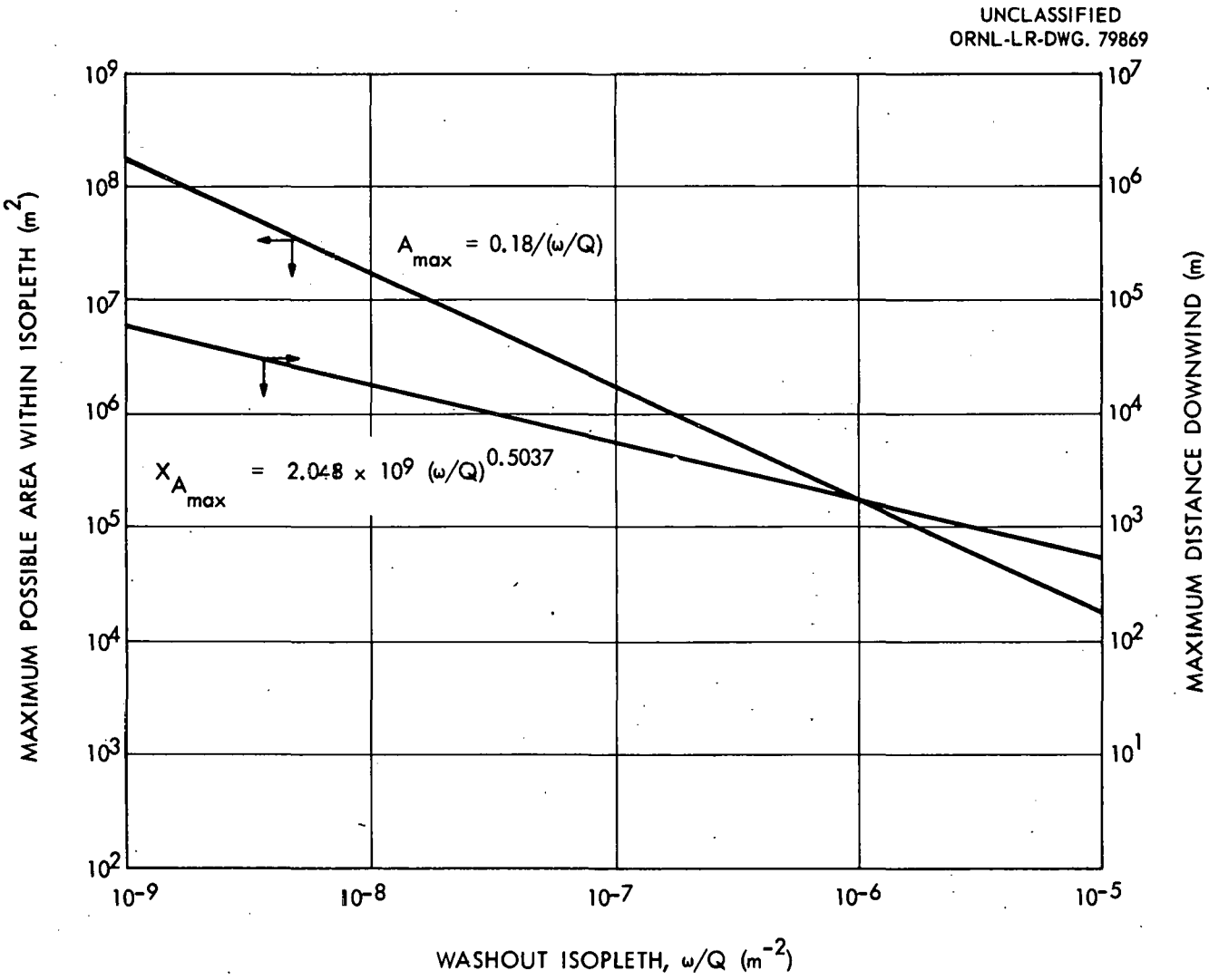

Fig. 5.10. Maximum Possible Area and Corresponding Maximum Downwind Distance for a Washout Isopleth. Type "D" (cloudy) conditions.

indicates the magnitude of the coefficient Λ when lycopodium spores were the test material.

Table 5.2. Measurements of Washout Coefficient Λ for Lycopodium Spores

Date	$\bar{u}$ (cm/sec)	Rate of rainfall (mm/hr)	Type of rain	Λ 10^{-4}/sec Observed	Λ 10^{-4}/sec Theoretical
16.8.56	320	3.91	frontal	10.2	9.7
27.9.56	543	1.12	frontal	4.2	3.6
25.10.56	845	14.1	heavy frontal	30.8	26.8
11.12.56	334	1.01	frontal	3.2	3.2
31.12.56	332	3.64	continuous rain of showery type	8.9	9.2

Conclusions with Respect to Applicability of Meteorological Conditions

The area that may be enclosed within the ground contamination isopleth is summarized in Fig. 5.11. This figure allows comparison of the maximum possible area with the area in various typical, consistent conditions. Washout in a typical frontal rainstorm causes conditions for maximum area within an isopleth relatively near the source, while a typical light rain maximizes conditions relatively far from the source. During moderately stable (inversion) conditions, dry deposition causes closest approach to the maximum-area curve at progressively greater distances from the source as the average deposition velocity is decreased. Progressively lower values of area within a given deposition isopleth generally result as the atmospheric lapse rate increases and the deposition velocity decreases.

In this study, extensive use will be made of the maximum area within a given ground contamination isopleth as it is defined by the washout condition. This condition, which maximizes the contaminated area but causes lower-than-maximum personnel exposure (Fig. 5.12), maximizes the economic loss from ground contamination for all the radioisotopes studied.

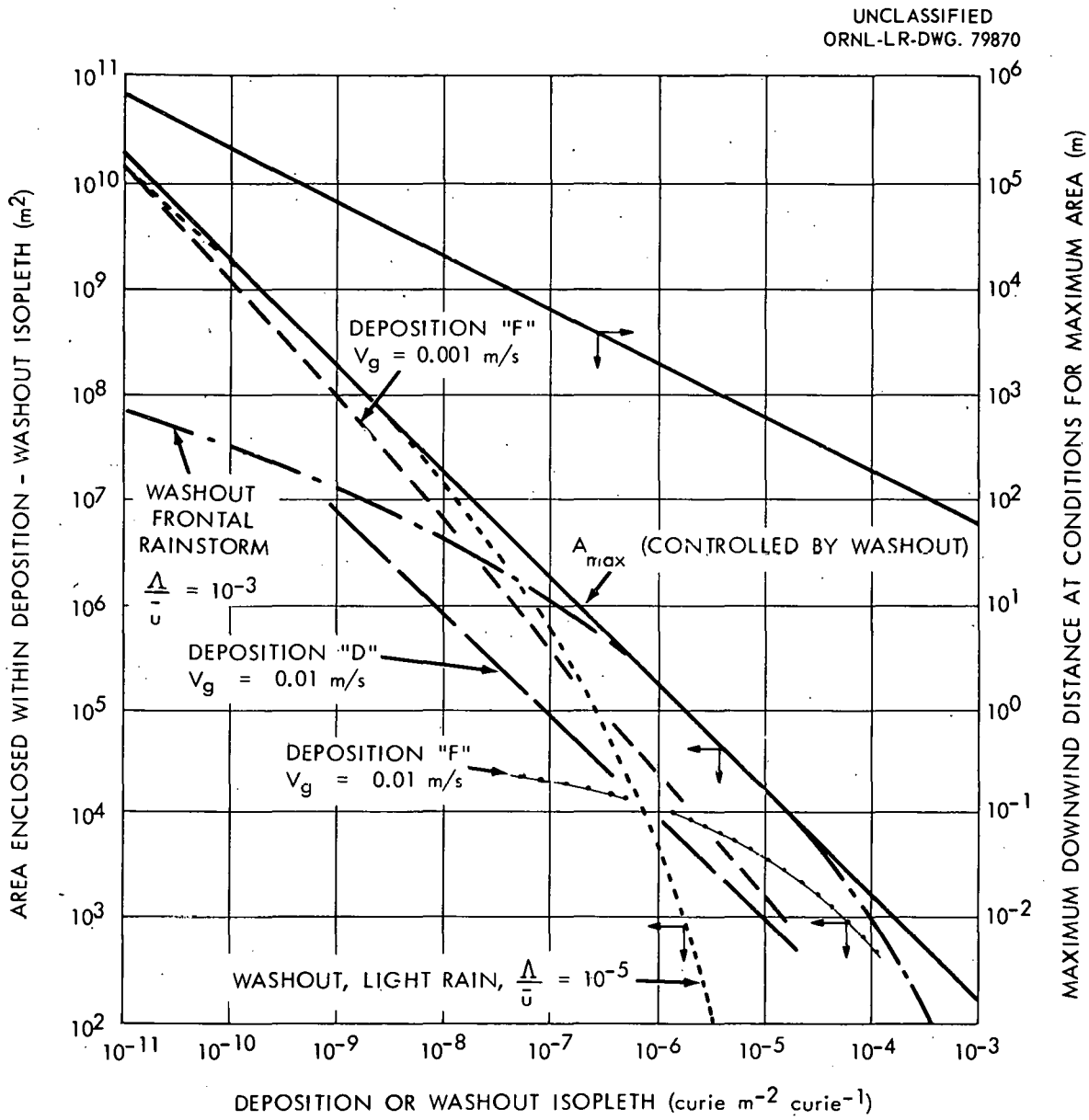

Fig. 5.11. The Enclosed Area Within a Ground Contamination Isopleth and the Maximum Downwind Distance Corresponding to the Conditions for Maximum Area.

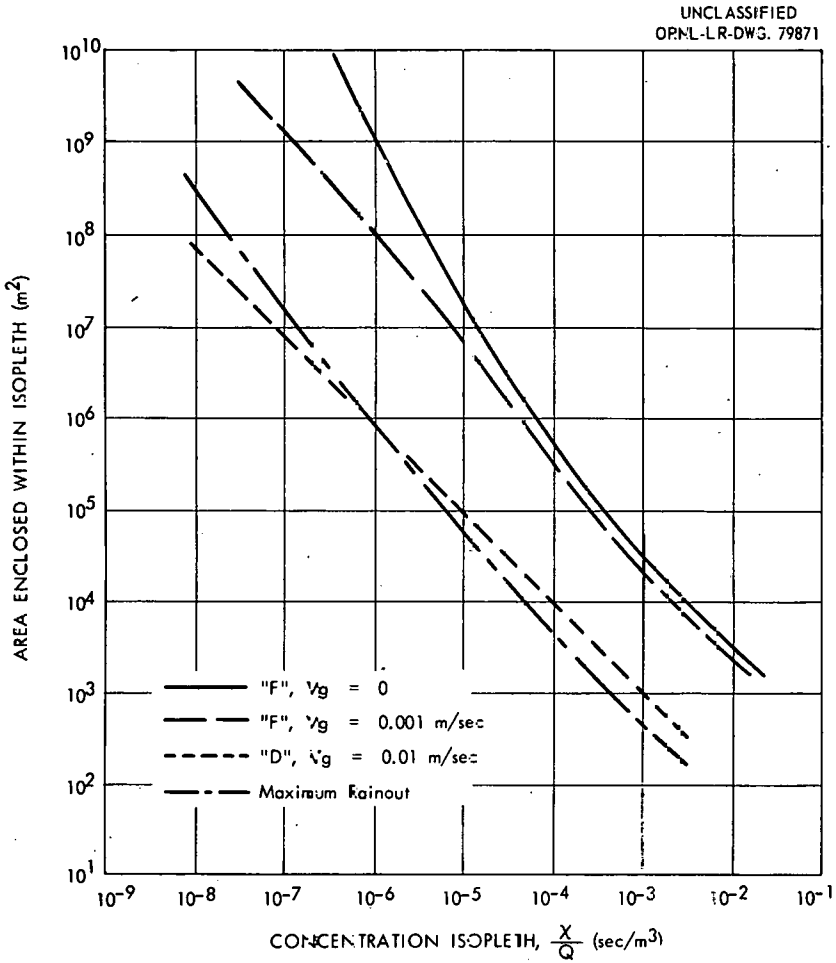

Fig. 5.12. Enclosed Area for a Concentration Isopleth.

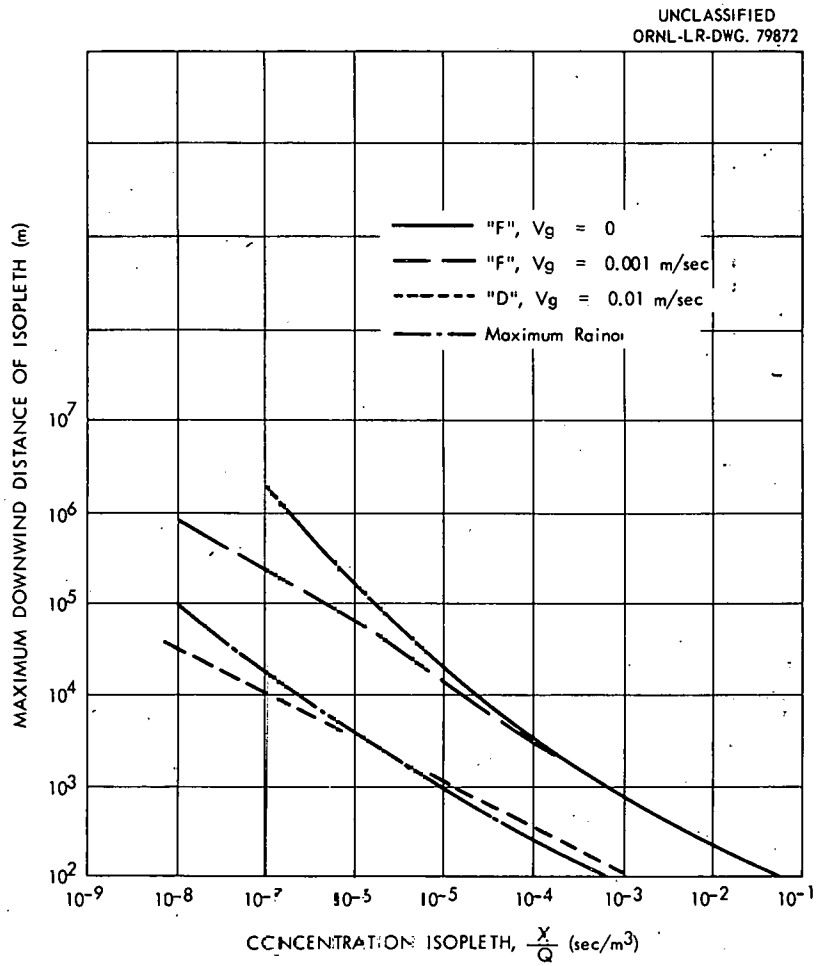

Fig. 5.13. Maximum Downwind Distance for a Concentration Isopleth.

Washout is maximized in a totally realistic manner by assuming only one parameter - a cloudy day with an unspecified rate of precipitation. The washout conditions are believed to be practicably obtainable over the range of contamination isopleths and areas calculated in this study. Further, through the use of credible deposition velocities for particles that might be released in an accident, the maximum area as defined by washout may be closely approached by dry deposition during inversion conditions. This tends to cancel out possible discrepancies in the economic-loss assumptions because of ground-wash-down effects during rain.

The equation for the maximum area, A_{max} (square meters), enclosed with a given contamination isopleth, ω/Q (curies per square meter per curie released), may be empirically approximated, within a few percent, over the range of interest, as:

$$A_{max} = \frac{0.18}{\omega/Q}.$$

This equation may be used to closely approximate the area within the several contamination ranges of interest in a given accident because the area inside of one specific isopleth controls the economic-loss calculation. Precise knowledge of the area enclosed within the other isopleths at the same conditions is not required.

The equation for the maximum downwind distance, $X_{A_{max}}$ (in meters) within a contamination isopleth, for the conditions of maximum area is:

$$X_{A_{max}} = 2.048 \times 10^{9} \left(\frac{\omega}{Q}\right)^{0.5037}.$$

The enclosed area and maximum downwind distance for a concentration isopleth are summarized in Figs. 5.12 and 5.13. The area and downwind distance are maximized for an inversion condition with a deposition velocity of zero, and progressively decreases with increasing rate of deposition.

While it is ordinarily not possible to evaluate the probability of occurrence of a violently dispersive accident, it is of value to examine the approximate frequency of occurrence of various meteorological conditions because these factors come into play in all releases to the atmosphere and have a profound effect on the extent of downwind personnel exposure and ground contamination.

The frequency of occurrence of the various meteorological categories have been determined at Oak Ridge, Tennessee,[10] and at Croydon, England[11] (Table 5.3). While these percentage frequencies are not representative of any site other than that for which the determinations were made, they do, in general, indicate the approximate spectrum of weather types. At these two locations the frequency of occurrence of lapse, neutral, and inversion conditions are roughly equal.

Table 5.3. Average Annual Frequency of Stability Categories at Croydon, England, and Oak Ridge, Tennessee

Stability Category	Description	Percent Annual Frequency at: Croydon, England	Oak Ridge, Tennessee
A	Extremely unstable conditions, very sunny summer weather	1.8	0.4
B	Moderately unstable conditions, sunny weather	8.6	8.6
C	Slightly unstable conditions, average day	16.6	35.3
D	Neutral conditions, overcast day or night	38.0	19.7
E	Slightly stable conditions, average night	12.4	22.0
F	Moderately stable conditions, clear night	14.3	9.9
G	Extremely stable conditions, cool night with heavy dew	8.1	4.1

Wind-direction frequencies for several locations in the United States are shown in Table 5.4 (ref 12). These data are typical of most of the United States in that the frequency in any 45^{o} segment of direction does not exceed 25 to 30%.

The average annual frequency of occurrence of rain and average annual rainfall at several locations in the United States is shown in Table 5.5. In most of the United States the average annual frequency of rain and total rainfall are 5-15% and 15-30 in., respectively.

Table 5.4. Average Annual Surface-Wind-Direction Frequencies at Several Locations in the United States

Wind Direction	Percent Frequency from Given Direction at:			
	Nashville	Buffalo	Albuquerque	San Francisco
N	14	6	15	5
NE	6	9	8	5
E	2	7	4	7
SE	4	8	10	7
S	18	12	11	5
SW	17	27	10	15
W	5	18	10	25
NW	10	9	12	11
Calm[a]	24	4	20	20

[a]Wind speed less than 3 mph.

Table 5.5. Average Annual Frequency of Occurrence of Rain and Average Annual Rainfall at Several Locations in the United States

Location	Average Annual Frequency of Occurrence of Rain (%)	Average Annual Rainfall (in.)
Nashville	10	45
Buffalo	15	32
Albuquerque	3	8.7
San Francisco	6	18

REFERENCES (SECTION 5)

1. C. K. Beck et al., Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants, WASH-740 (1957).
2. F. A. Gifford, "The Problem of Forecasting Dispersion in the Lower Atmosphere," USAEC DTIE (July 1961).
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4. W. F. Hilsmeier and F. A. Gifford, Graphs for Estimating Atmospheric Dispersion, ORO-545 (July 1962).
5. W. M. Culkowski, "Deposition and Washout Computations Based on the Generalized Gaussian Plume Model," ORO-599 (Sept. 30, 1963).
6. W. M. Culkowski, Estimates of Accumulated Exposures and Environmental Buildup of Radioactivity, TID-7593, pp 89-99 (1961).
7. A. C. Chamberlain, Aspects of Travel and Deposition of Aerosol and Vapour Clouds, A.E.R.E., HP/R 1261, H.M.S.O. (1953).
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10. W. M. Culkowski, Oak Ridge AEC Weather Bureau Research Station, personal communication with J. P. Nichols, ORNL, March 12, 1963.
11. "IAEA Meeting on Reactor Siting," Nuclear Safety, 3, p 16 (June 1962).
12. Airway Meteorological Atlas for the United States, U.S. Department of Commerce, USGPO (1941).

6. POTENTIAL ECONOMIC LOSS AS A FUNCTION OF THE QUANTITY OR RADIOACTIVE MATERIAL RELEASED

The potential economic loss, as a function of curies of various radioisotopes released, was calculated by using the previously established assumptions regarding economic loss as a function of contamination and exposure ranges (Section 3), population distribution (Section 4), and meteorology (Section 5). A ground-level release of the radioactive materials as vapors, small particles, or smokes was assumed. The effects of a stack release for the washout condition are essentially identical to a ground-level release. In an inversion, a stack release would result in less damage than a ground-level release, however, the height of release decreases in importance as the release becomes larger, and the effects extend to greater downwind distances. In general, washout conditions maximize the loss due to ground contamination, while inversion maximizes the loss due to personnel exposure.

The potential economic loss as a function of curies of Sr^{90} released is shown for washout and inversion conditions in Fig. 6.1 for a typical population distribution, and in Fig. 6.2 for a uniform population distribution. The loss for a strong-lapse condition representative of a sunny day is shown in Fig. 6.3 for a typical population distribution. For releases greater than 1000 curies of Sr^{90}, the damage for the sunny day condition is less by a factor 10^3 than the damage during either washout or inversion conditions.

For comparison, the economic loss predicted in report WASH-740[1] for a reactor accident releasing 150,000 curies of Sr^{90} under inversion and washout conditions is plotted in Fig. 6.1. The results of the two studies are in fair agreement.

Economic losses from the release of a large number of isotopes were calculated as a function of the quantity released and similar plots prepared. A list of the isotopes for which loss plots are presented, and an index to their location in the Appendix is included in Table 6.1. Table 6.2 is a summary of the minimum quantity of each isotope that must be released to cause 60 million dollars damage under unfavorable weather conditions.

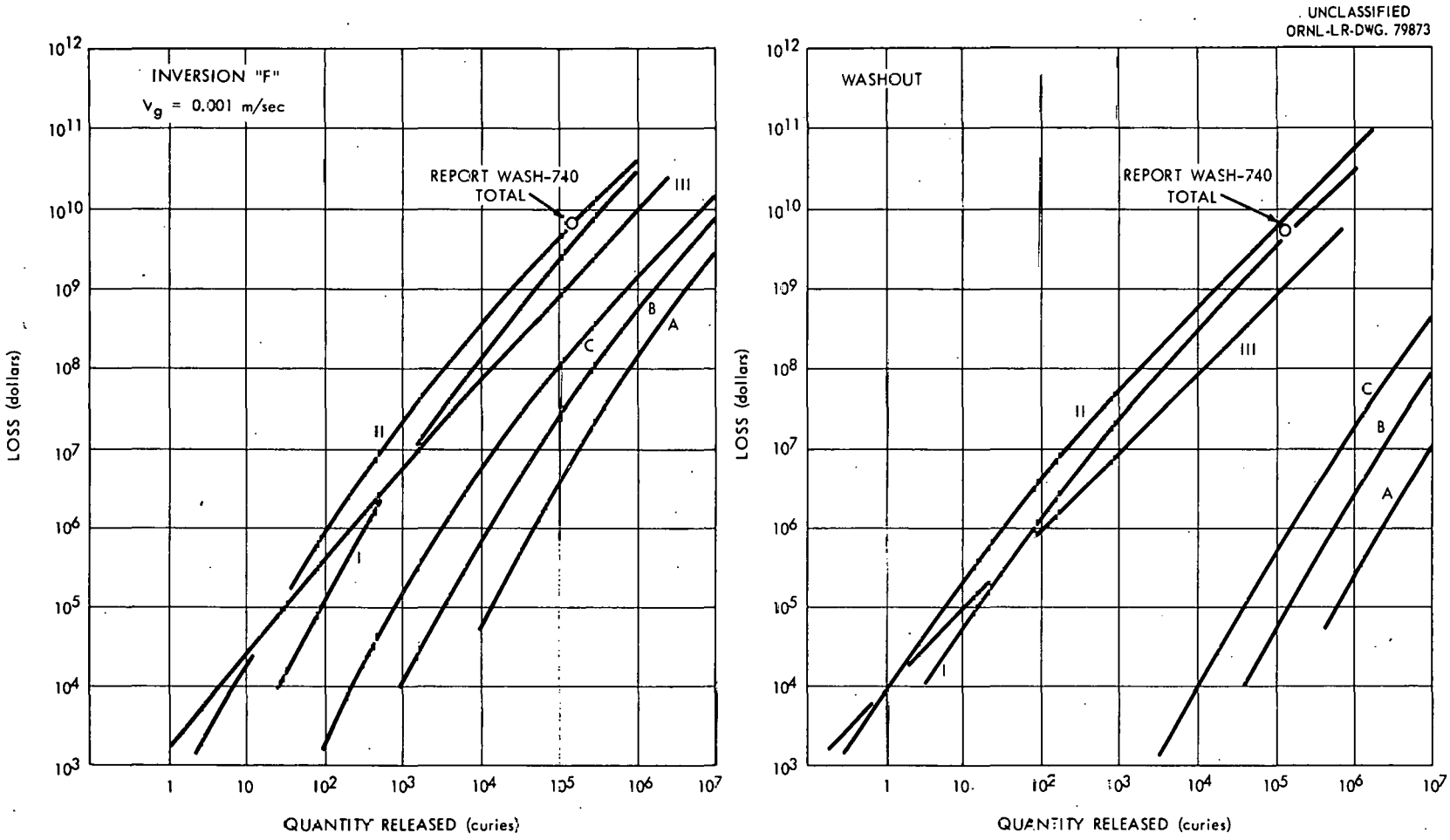

Fig. 6.1. Potential Economic Loss Resulting from Release of Sr^{90}. Typical population distribution.

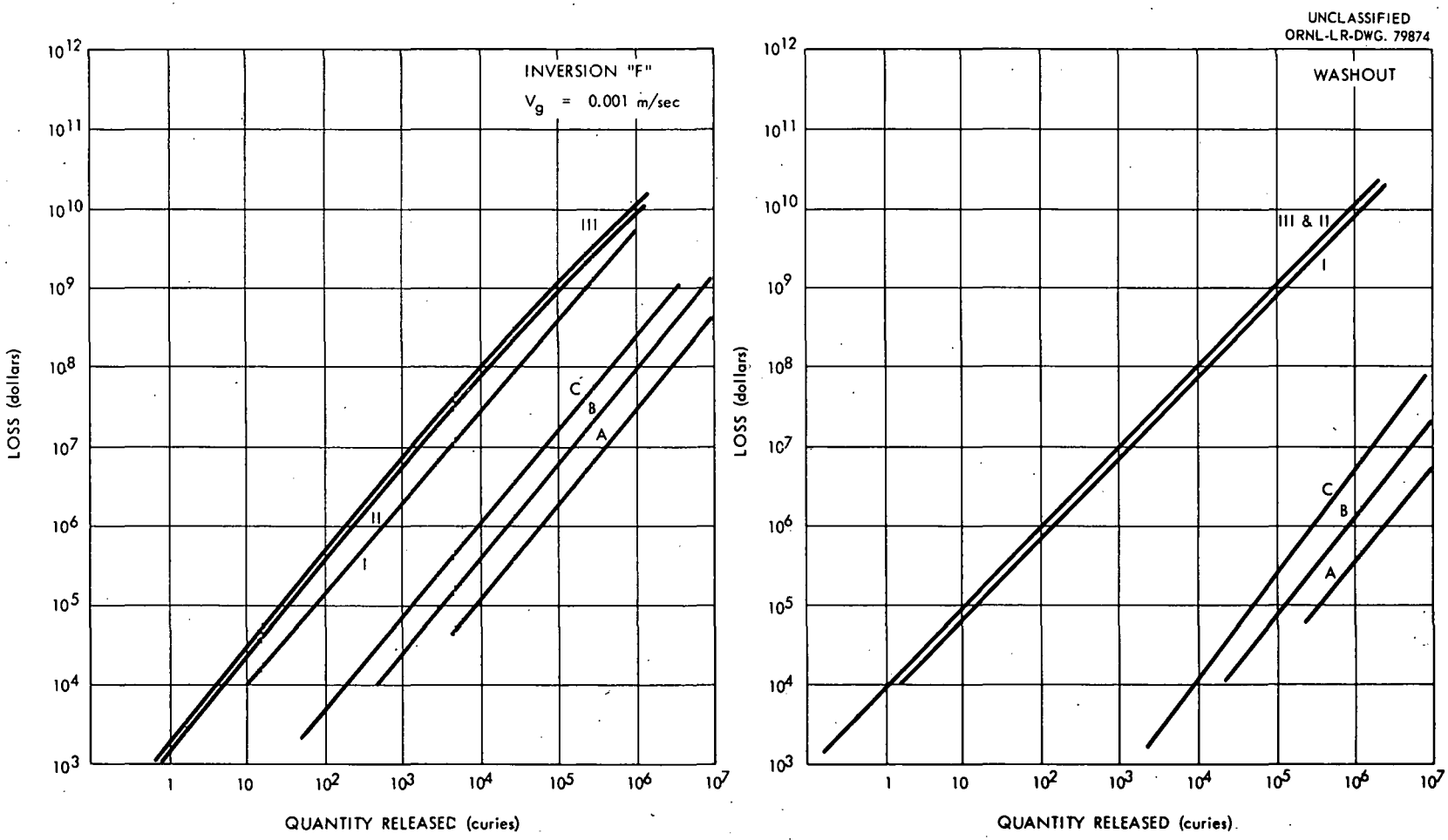

Fig. 6.2. Potential Economic Loss Resulting from Release of Sr^{90}. One hundred persons per square mile.

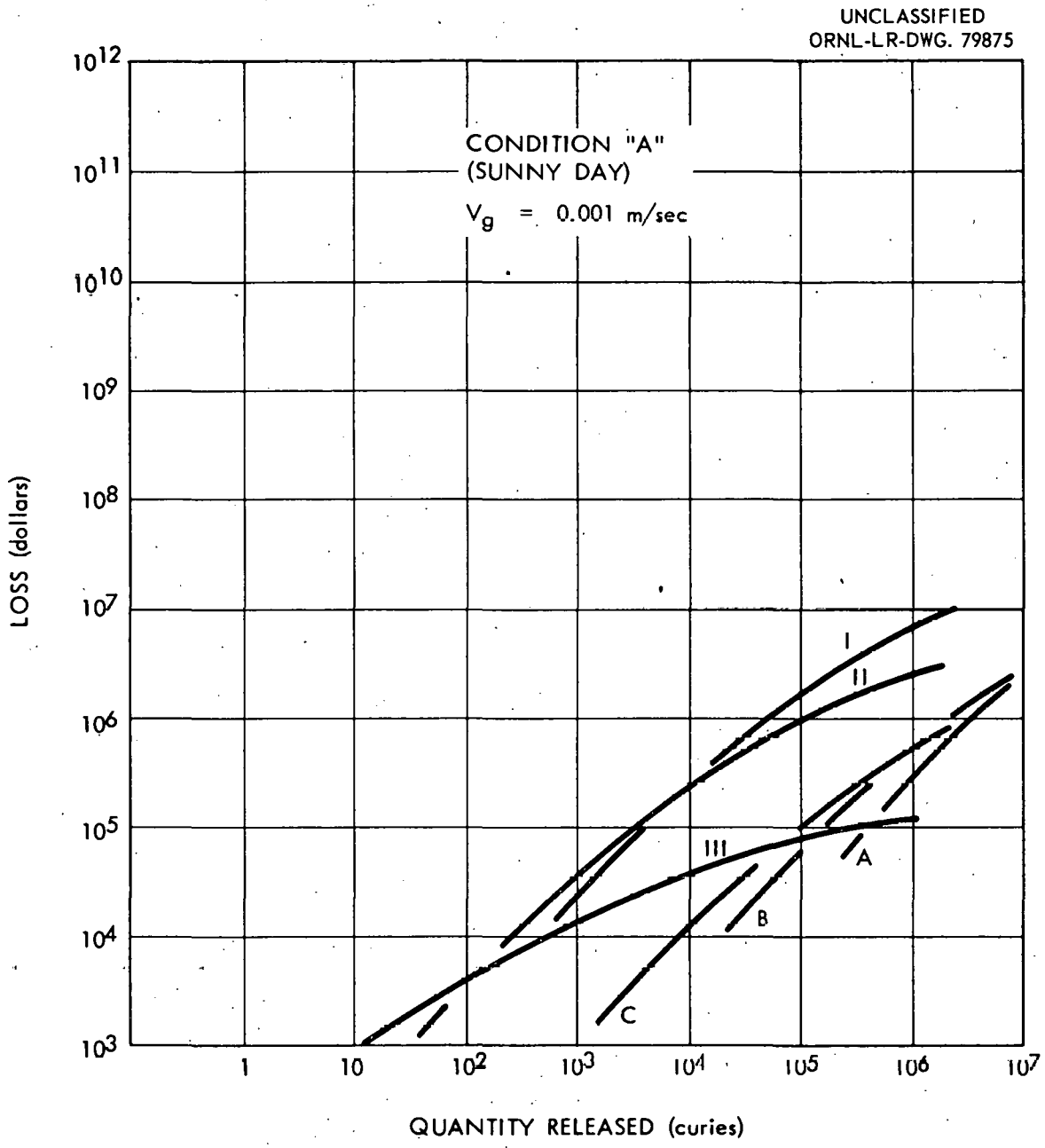

Fig. 6.3. Potential Economic Loss Resulting from Release of Sr^{90}. Typical population distribution.

Table 6.1. Index of Economic-Loss Plots in Appendix A

Isotope	Typical Population Distribution		Uniform Population Distribution, 100 people/mi^2	
	Figure	Page	Figure	Page
Co^{60}	A-1	110	A-2	111
Kr^{85}	A-3	112	A-3	112
Sb^{124}	A-4	113	A-5	114
I^{131}	A-6	115	A-6	115
Cs^{137}	A-7	116	A-8	117
Ce^{144}	A-9	118	A-10	119
Pm^{147}	A-11	120	A-12	121
Tm^{170}	A-13	122	A-14	123
Ir^{192}	A-15	124	A-16	125
Po^{210}	A-17	126	A-18	127
U^{233}	A-19	128	A-20	129
U^{235}	A-21	130	A-22	131
Pu^{238}	A-23	132	A-24	133
Pu^{239}	A-25	134	A-26	135
Cm^{242}	A-27	136	A-28	137
Cm^{244}	A-29	138	A-30	139

Table 6.2. Minimum Quantity of Material Which Must be Dispersed to Atmosphere to Cause 60 Million Dollars Damage Under Unfavorable Weather Conditions

Isotope	Most Unfavorable Weather Conditions	Minimum Amount Release for $60 Million Damage			
		Uniform Population of 100 Persons per Square Mile		Typical Population Distribution	
		(Grams)	(Curies)	(Grams)	(Curies)
Co^{60}	Inversion	500	5×10^5	80	8×10^4
Kr^{85}	Inversion	200,000	7×10^7	50,000	2×10^7
Sr^{90}	Washout	30	4×10^3	7	1×10^3
Sb^{124}	Inversion	30	5×10^5	5	1×10^5
I^{131}	Washout	0.3	4×10^4	0.3	4×10^4
Cs^{137}	Washout	500	4×10^4	100	1×10^4
Ce^{144}	Inversion	60	2×10^5	20	5×10^4
Pm^{147}	Inversion	4000	4×10^6	500	5×10^5
Tm^{170}	Inversion	100	9×10^5	30	2×10^5
Ir^{192}	Inversion	50	5×10^5	10	1×10^5
Po^{210}	Inversion	1	5×10^3	0.3	1.5×10^3
U^{233}	Inversion	9×10^4	900	2×10^4	200
U^{235}	Inversion	2×10^9	4000	3×10^8	600
Pu^{238}	Washout	9	150	4	60
Pu^{239}	Washout	2000	100	900	60
Cm^{242}	Inversion	1	4000	0.3	900
Cm^{244}	Washout	10	900	3	250

Figure 6.4 shows the potential economic loss from criticality accident as a function of the number of fissions, assuming 100% release of the fission products. These loss values were calculated using the personnel exposure and ground contamination data from report NYO-2980.[2] The "A," "B," and "C" exposure categories in that report were taken to be equivalent to our Range A, Range B and Range C, respectively. Ground contamination, as in report NYO-2980, was assumed to be controlled by Sr^{90} and I^{131}. Contamination Ranges I and II were controlled by the level of Sr^{90}. Contamination Range III was controlled by I^{131} rather than by Sr^{90}. Since I^{131} controls the contamination Range III, which is a major contributor to the economic loss, the loss economics would not be affected significantly during washout if only the halogens and rare-gas fission products were released.

The economic loss from maximum personnel exposure, calculated assuming inversion conditions, begins to rival the loss for conditions for maximum contamination (washout) as the number of fissions becomes large.

The economic, populational, and meteorological assumptions were combined to yield generalized plots (Fig. 6.5 through 6.8) of potential economic loss as a function of deposition and exposure isopleth. By use of these plots and contamination and exposure ranges, the potential economic loss can be estimated for the release of any quantity of any isotope. An example of the use of these plots for determining the economic loss from release of 1000 curies of Sr^{90} is shown in Table 6.3. Column 1 in the table lists the exposure and contamination values for the loss range. Column 2 lists the isopleth for reach range (column 1 divided by the curies released). Column 3 is the dollars loss figure obtained from Fig. 6.5 using the curve for each range with the appropriate isopleth from Column 2.

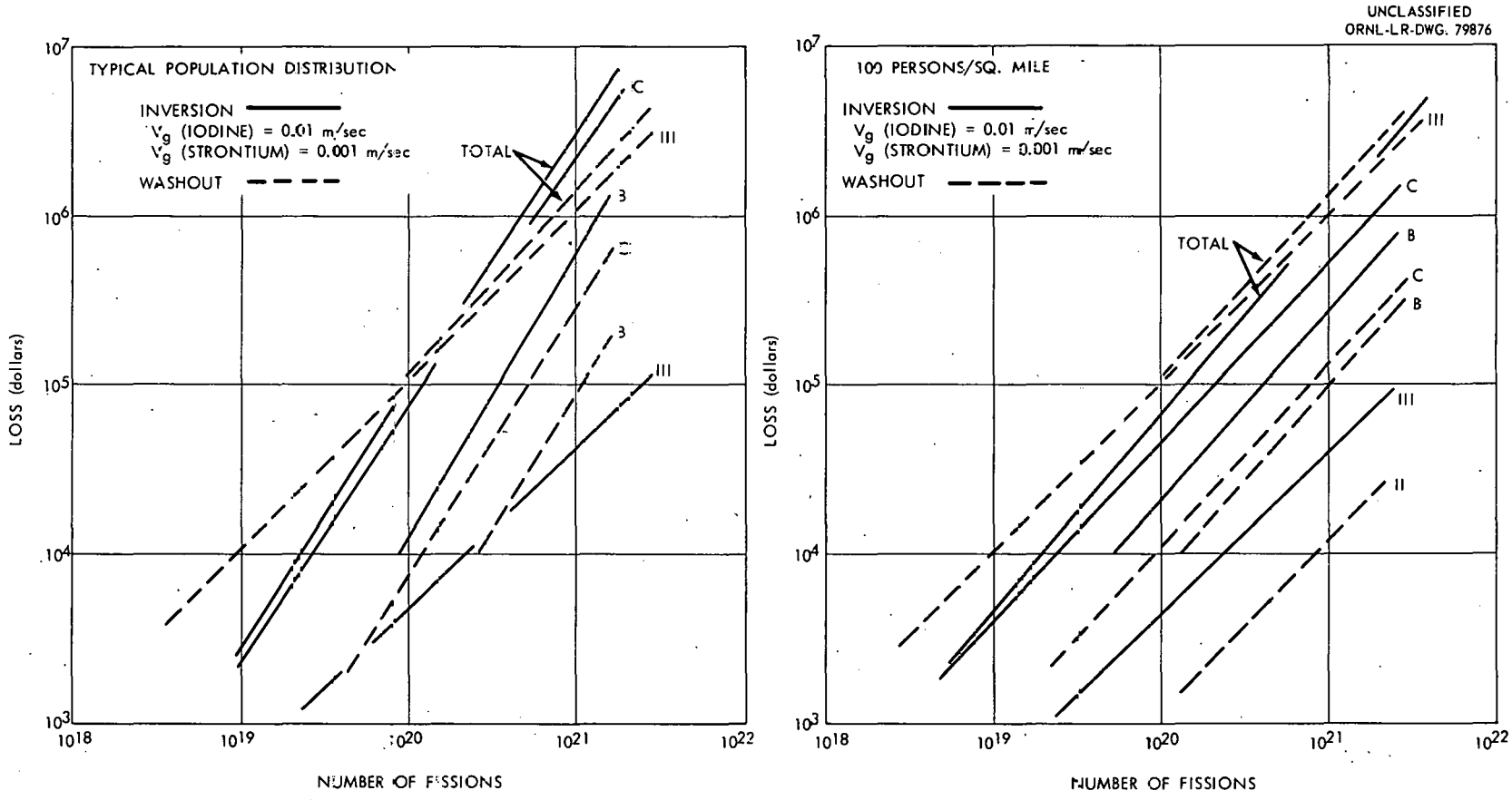

Fig. 6.4. Potential Economic Loss Resulting from Ground-Level Release of 100% of the Fission Products from a Criticality Accident as a Function of the Number of Fissions.

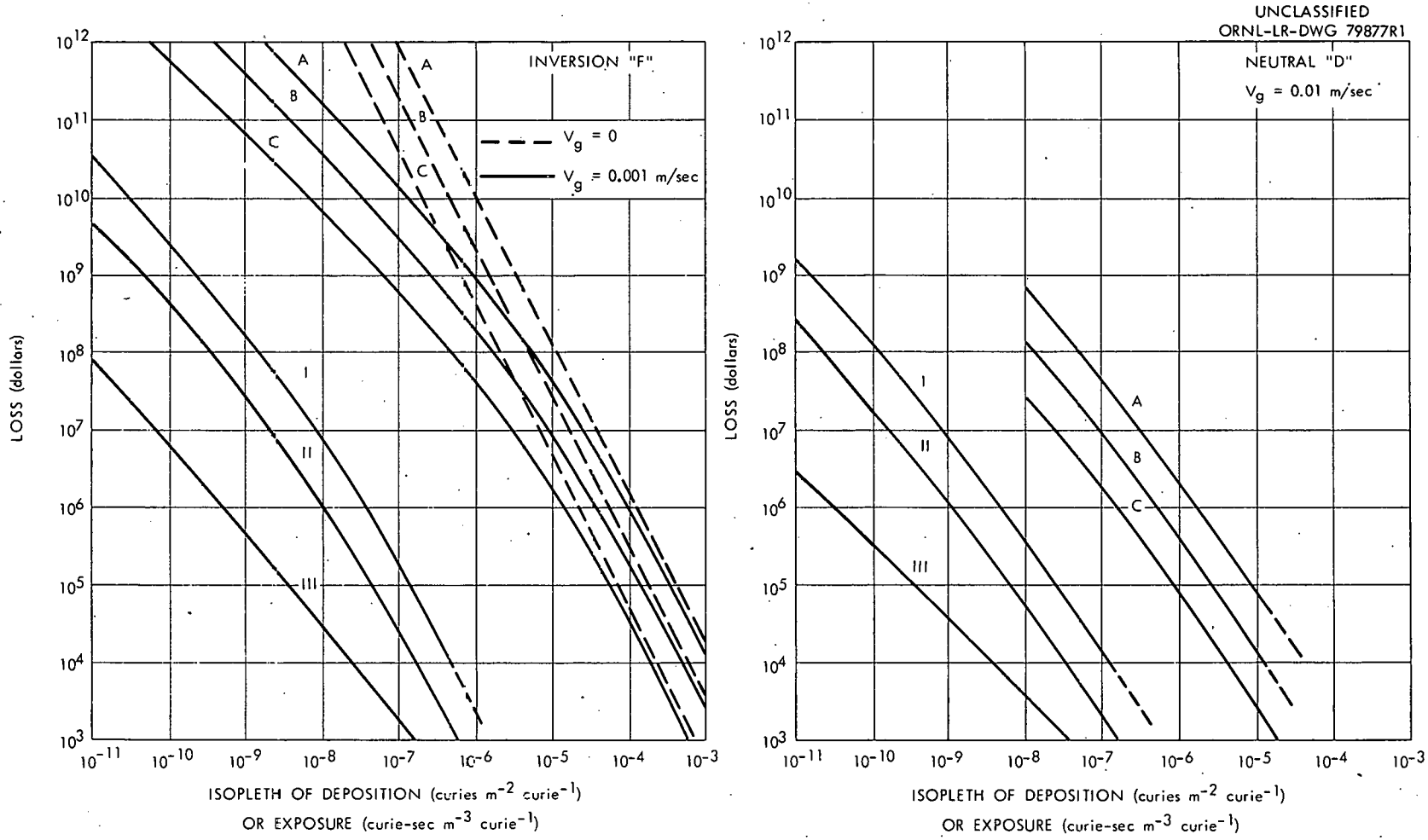

Fig. 6.5. Potential Economic Loss as a Function of Deposition of Exposure Isopleth. Typical population distribution.

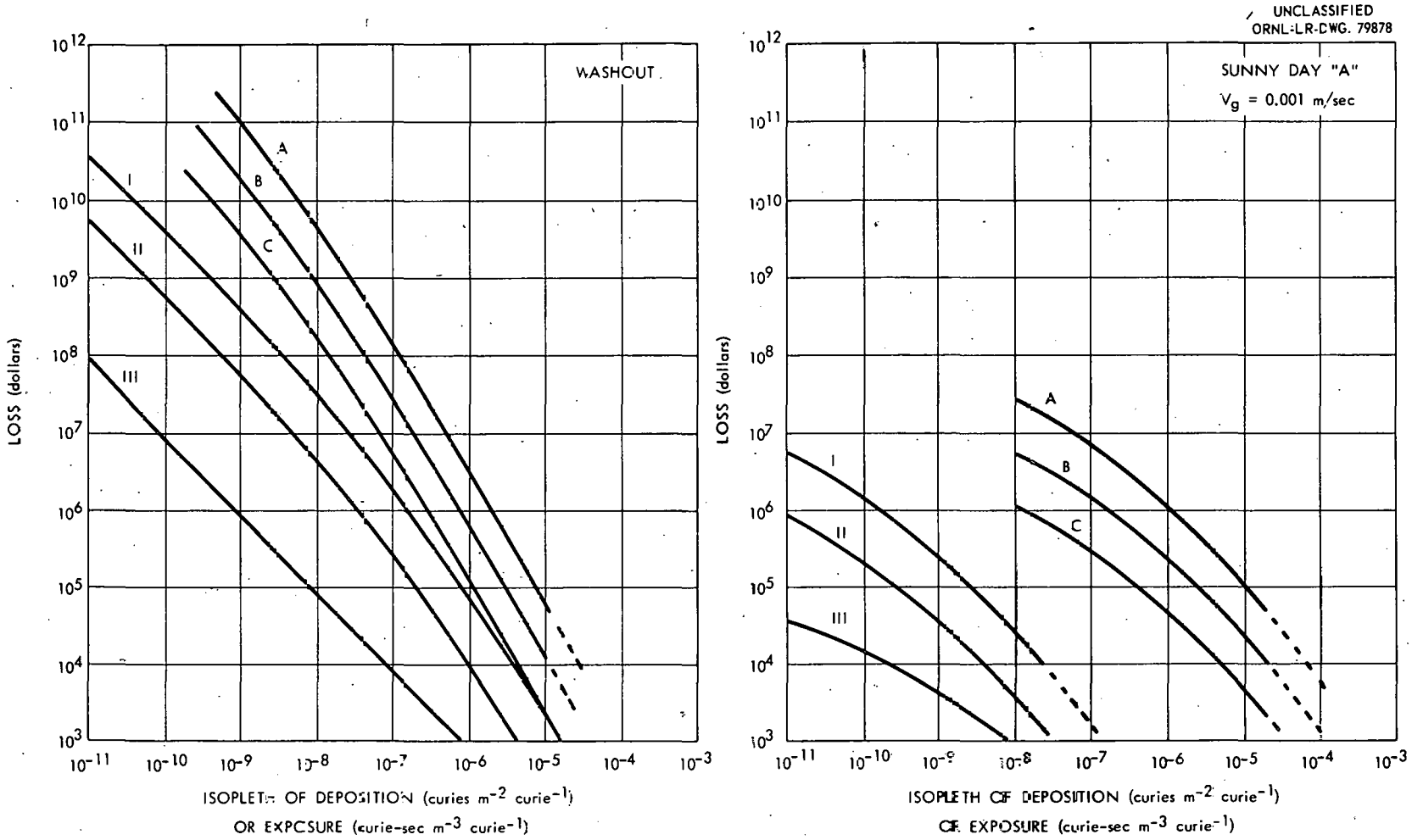

Fig. 6.6. Potential Economic Loss as a Function of Deposition or Exposure Isopleth. Typical population distribution.

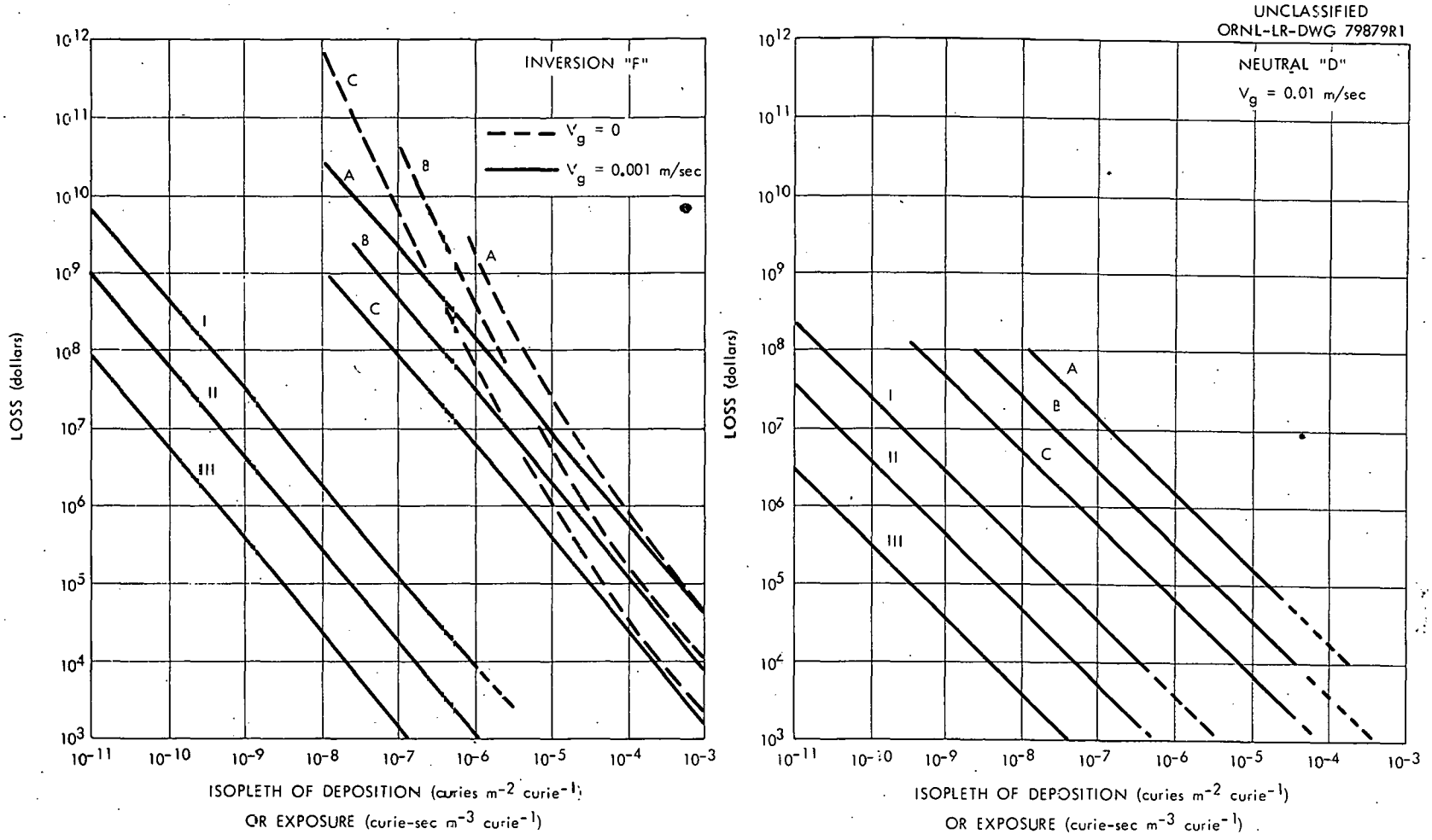

Fig. 6.7. Potential Economic Loss as a Function of Deposition or Exposure Isopleth. One hundred persons per square mile.

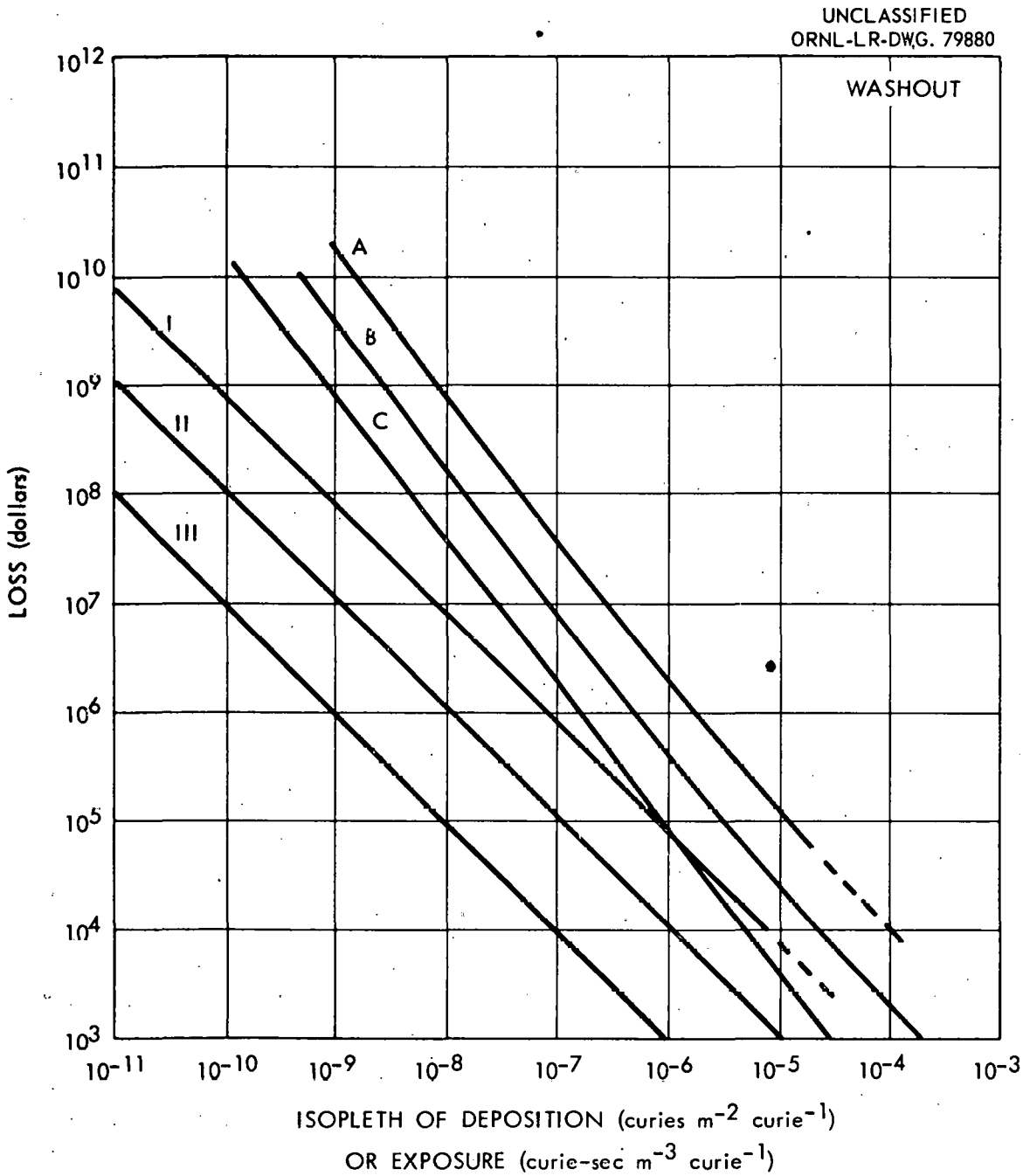

Fig. 6.8. Potential Economic Loss as a Function of Deposition or Exposure Isopleth. One hundred persons per square mile.

Table 6.3. An Example of the Use of Figs. 6.5-6.8 to Estimate the Economic Loss from Release of a Given Quantity of Radioactive Material. Assumptions: 1000 Curies Sr^{90} Released, Inversion Conditions, V_g = 0.001 m/sec, Typical Population Distribution (Fig. 6.5).

Exposure or Contamination Range	(1) Assumed Values for Sr^{90} (From Tables 3.1 and 3.3)	(2) Isopleth of Deposition or Exposure for 1000 Curie Release	(3) Economic Loss (From Fig. 6.5)
A	4.1 (curie-sec-m^{-3})	4.1×10^{-3} (sec-m^{-3})	None
B	0.41	4.1×10^{-4}	$ 1.5×10^{4}
C	0.041	4.1×10^{-5}	1.5×10^{5}
I	1.1×10^{-5} (curie-m^{-2})	1.1×10^{-8} (m^{-2})	6×10^{6}
II	1×10^{-6}	1×10^{-9}	2.5×10^{7}
III	1×10^{-7}	1×10^{-10}	6×10^{6}
Total			$ 3.7×10^{7}

REFERENCES (SECTION 6)

1. Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants, WASH-740, USGPO, p 78 (Mar. 1957).
2. H. T. Williams et al., Safety Analysis of Enriched Uranium Processing, NYO-2980 (Mar. 18, 1960).

7. OFF-SITE DAMAGE CALCULATED FOR POSTULATED ACCIDENTS IN RADIOCHEMICAL FACILITIES

Evaluation of the potential damage which could result from theoretical accidents is based on the accident categories and potential accident and release mechanisms discussed in Section 1 and the assumed design characteristics and inventories discussed in Section 2, combined with the economic loss vs release plots developed in Section 6. The maximum potential dollar losses are determined from the plots of economic loss as a function of quantity released for meteorological conditions that maximize the loss consistent with the properties of the material released in the postulated accident.

The type of accidents covered are fires and explosions for all plants and in addition criticality accidents for fuel fabrication facilities. In all cases the maximum release (and damage) has been calculated for each category so that the postulated accidents and calculated damage are maximum for the particular category and type of plant. The damage figures given are for the weather conditions which result in the maximum loss. For accidents involving Sr^{90} in each category, the loss resulting for the most favorable (least damage) weather conditions has also been calculated for comparison. The damage calculated has been based on postulated future inventories but with present technology in plant design and containment; the damage values are, therefore, probably on the high side.

Category 1 Accidents

Category 1 includes those accidents where a portion of the plant becomes contaminated or an employee exposed to radiation but no radioactive material escapes from the facility. This type of accident could be caused by some not particularly unusual mishap such as a pipe leak, dropped sample, etc. Damage in this category of accident is confined to the facility and would result in plant cleanup and downtime. There would be no off-site damage. Several Category 1 accidents which have occurred were discussed in Section 1. The following hypothetical accident is postulated as an example of the most severe accident in this category.

Explosion in Aqueous Solution Tank - A radiolytic hydrogen-air explosion in a tank of aqueous solutions could scatter the contents of the tank

as a liquid aerosol to the cell air. It would not be expected to rupture the containment or filters. Assuming that the concentration of the radioisotope in the solution would be no more than 10 w/liter or about 2000 curies/liter, about 0.03 curie of solution would be released to the atmosphere through the filters and stack (one cell volume at 0.14 mg/m^3). There would be no economic losses from off-site damage for any of the radioisotopes considered.

Category 2 Accidents

Category 2 includes those accidents where in addition to damage to the facility a small amount of radioactive material escapes from the plant containment systems. This type of accident could result from some occurrence such as a fire, explosion, criticality excursion, etc. Although the facility may be lost and possibly some employees injured, there would be only very minor, if any, public damage involved in this type accident. Experience has shown that this type of accident has a very low probability of occurrence. To date all major radiochemical plant accidents have been in Category 1 or this category where no appreciable contamination was spread off site. This category is the most severe that has occurred or is expected to occur. Several Category 2 accidents which have occurred were discussed in Section 1. The following hypothetical accidents, postulated as the maximum severity Category 2 accidents, are summarized in Table 7.1.

Postulated Mechanisms and Effects of Category 2 Accidents in Plants for Processing Beta-Gamma Emitters

Fire in Organic Solution Vessel - A solvent extraction contactor containing 1.25×10^4 curies (5% of the maximum inventory of Sr^{90} in a single vessel) is postulated to catch fire and disperse 20% of its contents (2500 curies) as smoke to the cell air. The 25-curie release through the filters (assumed 99% efficient) is 0.2% of the inventory of the tank or 0.01% of the maximum inventory in a single vessel. For such an accident involving Sr^{90}, the maximum potential economic loss is $1,000,000 for the unfavorable weather conditions (washout) and $10,000 for the favorable weather conditions (sunny day). The loss results are expressed for Sr^{90} only, since it is the worst case for a nonvolatile material in this category. Losses for

Table 7.1. Hypothetical Maximum Severity Category 2 Accidents

Type of Plant	Isotope	Events	Assumed Fraction Release	Quantity Released	Off-site Damage	
					Maximum (washout or inversion)	Minimum (Sunny day)
Beta-Gamma Isotope Processing	Sr^{90}	In-cell fire of organic solution from solvent extraction system	0.01% of material in a single vessel	25c	$\$10^6$	$\$10^4$
Fuel Fabrication Plant	Pu^{239}	Metal or carbide fire in glove box system releases 4% to laboratory	0.04% of contents of 4 glove boxes	4g Pu^{239}	$\$1.5 \times 10^5$	-
	U^{233}			4g U^{233}	None	-
Alpha Isotope Processing	Po^{210}	Glove-box fire and explosion filters remain intact	0.2% of contents of glove box	.0003g Po^{210}	$\$1.5 \times 10^3$	-
	Pu^{238}			.08g Pu^{238}	$\$1 \times 10^6$	-
	Cm^{242}			.00008g Cm^{242}	None	-
	Cm^{244}			.0002g Cm^{244}	None	-

any other radioisotopes may be obtained through the use of the calculated fractional release and the loss as a function of the release from the figures in Appendix A.

Postulated Mechanisms and Effects of Category 2 Accidents in Plants Fabricating Pu^{239} and U^{233} Fuel

Metal or Carbide Fire-Explosion in Glove Box - This is the classical "contained" accident in a glove-box laboratory. An explosion and/or fire is postulated to initiate in a glove-box train (four glove boxes each containing 2.5 kg of fissionable material), the metal or carbide burns, the smoke is dispersed to the laboratory by the explosion or by gloves burning followed by pressurization of the boxes. The building containment and filters are assumed to remain intact. Four percent of the carbide fuel in four glove boxes is assumed to be released (400 g as oxide smoke as it burns and to be carried into the ventilation system. Assuming that the filters are 99% efficient in removing this type of smoke, 4 g of the fuel is calculated to be released to the atmosphere. There is no off-site economic loss for such an accident involving U^{233} but, for Pu^{239}, the maximum potential economic loss is $150,000.

Mechanisms and Effects of Category 2 Accidents in Plants that Process Alpha Emitters

Minor Glove-box Fire-explosions - A fire-explosion is postulated to rupture a glove-box and disperse its contents as smoke (<0.1 μ particles) to the laboratory. The building containment is not breached, and the filters do not fail. This is assumed to result in 20% of the glove-box inventory (see Section 2) being discharged as smoke to the laboratory; of this, 1% is assumed to escape through the filters. There would be no off-site damage for this type of accident involving Cm^{242} or Cm^{244}, but the Category 2 potential damage involving Pu^{238} or Po^{210} is $1,000,000 and $1,500, respectively.

Category 3 Accidents

Category 3 encompasses those theoretical accident possibilities that could conceivably result in release off site of appreciable quantities of radioactive materials. Accidents in this category have an extremely low probability of happening. This is evidenced by the fact that postulation of such accidents requires the assumption of a sequence of events, each of

which is, in itself, highly unlikely. Further, no accidents of this severity have occurred in almost 20 years of AEC radiochemical plant operation. Unless prelicensing safety review of a facility showed that the probability of a Category 3 type accident was extremely remote, it would not be licensed for operation. Since no Category 3 accidents have occurred, the analysis must be based entirely on hypothetical accidents. The following hypothetical accidents, postulated as the maximum severity Category 3 accidents, are summarized in Table 7.2.

Postulated Mechanisms and Effects of Category 3 Accidents in Plants for Processing Beta-Gamma Emitters

Explosion During Powder Handling - A powder explosion or other source of agitation could disperse into the cell air the fine powder resulting from a precipitation or calcination operation. The containment and filters would be expected to remain intact. Measurements of the particle-size distribution of sintered $SrCO_3$ and $SrTiO_3$ have been made by E. Lieberman.[1] These studies indicated particle-size distribution for $SrCO_3$ of 93.41 wt % $\geq$ 0.3 μ, 5.72% 0.1 to 0.3 μ, and 0.87% less than 0.1 μ. Butler[9] found similar particle-size distributions for CeO_2. For this particle distribution and the filter efficiencies as a function of particle size assumed in Section 1, the overall filter efficiency would be 99.56%. Assuming that 20% of the material reaches the filter (99.56% efficient), 0.088% of the inventory of powder in the cell is released through the filters (220 curies). The maximum potential economic loss is 15 million dollars for an accident involving Sr^{90} during washout conditions, and $15,000 for the same release during "sunny day" conditions.

Postulated Mechanisms and Effects of Category 3 Accidents in Plants Fabricating Pu^{239} and U^{233} Fuel

Criticality Accident in a Storage Tank - The results show in report NYO-2980, coupled with accident experience and rationalizations of the characteristics of Pu^{239} or U^{233} indicate that a nuclear excursion in the storage tank could result in about 10^{20} fissions over several hours (which would be sufficient to evaporate most of the solution). It is assumed that the tank contains about 1000 liters of solution at a Pu^{239} or U^{233} concentration of 50 g/liter and utilizes fixed poison, soluble poison, or configurational methods for criticality control.

Table 7.2. Hypothetical Maximum Severity Category 3 Accidents

Type of Plant	Isotope	Events	Assumed Fraction Release	Quantity Released	Off-site Damage	
					Maximum (washout or inversion)	Minimum (Sunny day)
Beta-Gamma Isotope Processing	Sr^{90}	Explosion in furnace during calcining operation	0.088% of inventory in cell	220 c	$\$1.5 \times 10^7$	1.5×10^4
Fuel Fabrication Plant	Pu^{239}	Criticality accident in storage tank involving 10^{20} fissions total	0.2% non-volatile 100% volatile	10C g + volatile fission products	$\$5 \times 10^6$	-
	U^{233}	Criticality accident in carbide fuel storage of 10^{21} fissions	0.04% non-volatile 100% volatile	20 g + volatile fission products	$\$2 \times 10^6$	-
Alpha Isotope Processing	Po^{210}	Explosion in cell cubicle	0.022% of inventory in cubicle	0.0022 g	$\$4 \times 10^4$	-
	Pu^{238}			2.2 g	$\$5 \times 10^7$	-
	Cm^{242}			0.0044 g	$\$2 \times 10^5$	-
	Cm^{244}			0.44 g	$\$7 \times 10^6$	-

It is postulated that this accident results from misoperation, an internal hydrogen-air explosion, or an external fire which lead to supercritical conditions. Twenty percent of the 50 kg of Pu^{239} of U^{233} is assumed to be dispersed in the cell, and 1% of the dispersed fuel material is assumed to be released through the filter, as well as nearly 100% release of the newly created volatile fission products. While the release of fission products alone would result in economic losses of only $110,000, the effect of release of 0.2% of the fuel material causes maximum potential losses of $5 x 10^6 for the accident involving Pu^{239} and $150,000 for the accident involving U^{233}.

Fission products from the criticality accident contribute relatively little additional loss, compared with the release of Pu^{239} only. It is for this reason that a more rigorous analysis with respect to fission products generated in possible Pu^{239} criticality accidents is not included in the scope of this report.

Carbide Fuel Storage Area Criticality Accident - A postulated Category 3 accident which would be more serious than the one just described for U^{233} (because of higher fission yield and gaseous fission product release) and less serious than the one just described for Pu^{239} (because of lower fuel release) would be a criticality accident and fire in a carbide fuel storage area. It is postulated that a criticality accident over an extended period of 10^{21} fissions would lead to a fire in the carbide fuel. Assuming that the containment is not ruptured, 4% of the fuel is assumed to be released as smoke to the ventilation systems, and 1% of the smoke is assumed to be released to the atmosphere through the filters (0.04% overall release). It is assumed that 100% of the volatile fission products would be released to the atmosphere. For such an accident involving Pu^{239}, the maximum economic loss is approximately $3,000,000, predominantly controlled by Pu^{239}. For the accident involving U^{233} fuel, the maximum economic loss is approximately $2,000,000, all due to the fission products released.

Postulated Mechanisms and Effects of Category 3 Accidents in Plants That Process Alpha Emitters

Cubicle Explosion - It is assumed that a powder of H_2-air explosion in a cell cubicle ruptures the cubicle and scatters 20% of its contents of fine powders in the cell air but does not break the secondary containment or rupture the filters in the ventilation system. Use of the assumed

filter efficiencies as a function of particle size and a conservative particle size distribution 98.8% $\geq$ 0.3 μ, 1.1% between 0.1 to 0.3 μ, and 0.1% less than 0.1 μ results in a calculated filter efficiency of 99.89% and release of 0.022% of the cubicle inventory through the filters to the atmosphere. As discussed in Section 1, the release through leaks in secondary containment would be small compared with the release through filters. The calculated maximum potential economic losses are 54, 7.0, 0.16, and 0.036 million dollars for Pu^{238}, Cm^{244}, Cm^{242}, and Po^{210}, respectively.

Category 4 Accidents

Category 4 encompasses those theoretical accident situations wherein all containment and other safeguards systems are overcome and widespread dispersal of radioactive material to the atmosphere is postulated. With the multiple independent safeguards included in radiochemical plants, this type of accident is believed to have such an extremely low probability that it is deemed incredible. The sequence of prerequisite events that would be required to lead to this category of accident are far beyond any reasonable expectation. On the other hand, accidents having consequences in this category are considered in this report because theoretically one cannot conclude that they are completely impossible. Since, as noted, such accidents are not expected to occur (i.e., are "incredible") they fix the upper limit for this study of potential damage. The following hypothetical accidents, postulated as the maximum severity Category 4 accidents, are summarized in Table 7.3.

Postulated Mechanisms and Effects of Category 4 Accidents in Plants for Processing Beta-Gamma Emitters

Explosion-Fire in Cell - This accident is postulated to result from the explosion of a mixture of organic vapor or extraneous explosive gas in the cell air. The cell vessels are ruptured, the cell is destroyed, and the building is ruptured by debris from the cell. Twenty percent of the inventory of the cell is assumed released (5×10^4 curies) to the atmosphere as smoke or small particles. The maximum potential economic loss for such an accident involving Sr^{90} is $5,000,000,000 under washout conditions. Sunny-day conditions would result in a loss of only 1.5 million dollars.

Table 7.3. Hypothetical Maximum Severity Category 4 Accidents

Type of Plant	Isotope	Events	Assumed Fraction Release	Quantity Released	Off-Site Damage	
					Maximum (washout or inversion)	Minimum (Sunny day)
Beta-Gamma Isotope Processing	Sr^{90}	Explosion of organic vapor in cell which ruptures containment	20% of contents of cell	50,000 c	$5 x 10^9	$1.5 x 10^6
	I^{131}	Rupture of vessel containing isotope with complete release	100%	500 c I^{131}	$7.6 x 10^5	
	Kr^{85}			100,000 c Kr^{85}	None	
Fuel Fabrication Plant	Pu^{239}	Criticality excursion of 10^{21} fissions in carbide fuel storage area followed by CO explosion which ruptures containment	4% nonvolatile, 100% volatile of cell	4 kg Pu	$4 x 10^8	
	U^{233}			2 kg U^{233}	$2.5 x 10^6	
Alpha Isotope Processing	Po^{210}	Organic solvent explosion in cell which ruptures containment	20% of contents of cell	2 g	$7 x 10^8	
	Pu^{238}			2000 g	$5 x 10^{10}	
	Cm^{242}			4 g	$2 x 10^9	
	Cm^{244}			450 g	$1 x 10^{10}	

Release of Volatile Radioisotopes - Release of 100% of the volatile radioisotopes such as I^{131} and Kr^{85} is postulated to occur because of rupture of a single tank, with subsequent passage of the vapor or gas through the cell-ventilation system. Smoke, organic vapor, or steam are assumed to have made the I^{131} absorption beds in the ventilation system ineffective. While no losses are expected from release of 100,000 curies of Kr^{85}, the maximum potential economic loss from release of 500 curies of I^{131} is $760,000.

Postulated Mechanisms and Effects of Category 4 Accidents in Plants Fabricating Pu^{239} and U^{233} Fuel

Criticality Accident in a Storage Area for Carbide Fuel Followed by Fire and Explosion - The postulated Category 3 criticality accident (10^{21} fissions) in a carbide fuel storage area followed by a fire is postulated to be converted to a Category 4 accident by subsequent carbon monoxide explosions which first rupture the storage vault and then destroy the building. Experiments by W. E. Browning[3] indicate that pyrolytic-carbon-coated fuel in a graphite matrix may release about 4% of the fuel material and 20 to 100% of individual fission products under conditions comparable to these assumed. This postulated accident would result in a release of 4000 g of Pu^{239} or 2000 g of U^{233} as well as the fission products to the atmosphere as smoke. The maximum potential economic loss could be in the order of $400,000,000 from release of Pu^{239}, or 2.4 million dollars from the release of U^{233}. The loss in the Pu^{239} accident is due primarily to the Pu^{239} release; the loss in the U^{233} accident is due primarily to fission product release.

Postulated Mechanisms and Effects of Category 4 Accidents in Plants That Process Alpha Emitters

Fire-Explosion in Cell - Accidental formation of an explosive mixture of organic solvent in the cell air followed by ignition is postulated to cause this accident. Organic solvent may leak from a vessel and vaporize into the cell air. The accident is converted from the contained to the uncontained category with the assumption that the safeguards that maintain the cell air temperature below the flash point of the solvent and the devices that signal the approach to an explosive mixture fail simultaneously.

The effects of the accident are that the plugs in the roof of the cell are blown off, the building containment is ruptured, and the ventilation

systems cease to function. The explosion and a possible fire (organic solvents or ion exchange resins loaded with radioisotopes) in its aftermath are postulated to release fine powder or smoke that contains alpha emitters. All the radioactive material, whether finely divided powders or adsorbed on an ion exchange resin in a single cell, could conceivably be dispersed in the cell; 20% is assumed to be released to the atmosphere. The maximum potential economic losses are 49, 9.7, 1.7, and 0.68 billion dollars for accidents involving Pu^{238}, Cm^{244}, Cm^{242}, and Po^{210}, respectively.

Accidents More Serious Than Category 4

Although one might imagine accidents (e.g., release of entire radioactive inventory of a radiochemical facility) greater than those assumed in Category 4, we cannot conceive of any combination of circumstances by which this could happen. Therefore, no attempt was made to consider the damage consequences of any theoretical accident beyond Category 4. Category 4 is considered in this report not because it is ever expected to occur but because it provides an upper limit estimate of the damage under the worst possible circumstances.

REFERENCES (SECTION 7)

1. E. Lieberman, Union Carbide Nuclear Company, Sterling Forest, New York, personal communication (November 1962).
2. T. A. Butler, ORNL, personal communication with J. P. Nichols, ORNL (November 1962).
3. W. E. Browning, ORNL, personal communication with J. P. Nichols, ORNL (Jan. 17, 1963).

8. DISCUSSION OF RESULTS

In order to draw any conclusions from the figures developed in Section 7 one must be cognizant of the assumptions on which the results are based and the potential inaccuracies of the calculations.

Accuracy of Calculated Damage Figures

The values assumed for specific contamination and exposure ranges and the monetary values connected with these ranges, population density, meteorological calculations, and the amount of radioactive material released are the primary sources of error in the calculation of the maximum economic loss. The calculated costs as a function of contamination isopleths are probably accuracte for the average industrial site within plus or minus a factor of 5, although order-of-magnitude variation can be expected between an extremely remote area and a highly developed one. Errors in the personnel exposure ranges would not appreciably effect the results because the damages due to personnel exposure are, in general, minor compared to those from area contamination. For Category 4 accidents, the population density effects would tend to average out because these accidents would tend to involve distances of several hundred miles and, in general, area contamination is the major element of loss. The meteorological calculations of the dispersion and deposition of radioactive materials under the conditions of a major accident are probably accurate within a factor of plus or minus 2. The release fraction (20%) in a major accident is probably not high by more than a factor of 5 and is obviously not low by more than a factor of 5. The total of these errors indicates that the maximum economic losses are probably accurate for the average situation within plus or minus a factor of 50. It is more likely that the figures are an overestimation rather than an underestimation.

Comparison of Maximum Calculated Damage with Limits of Private Liability Insurance Available

The maximum private liability insurance currently available for any private plant in the atomic energy industry is sixty million dollars. A comparison of this figure with the results in Section 7 leads to the following conclusions:

1. Under the conditions assumed in this report presently available private insurance is sufficient to cover any accident which has more than a theoretical possibility of occurrence (Category 1, 2, or 3) in plants fabricating U^{233} or Pu^{239} fuel or processing and fabricating radioisotope sources.

2. For Category 4 accidents (those which are theoretically possible but incredible) the isotopes may be divided into the following categories:

(a) Those where the calculated damage from such as assumed release exceeded \$3 x 10^9 (i.e., where the calculated loss would exceed \$6 x 10^7 even if one assumes the calculations were a factor of 50 high) include Sr^{90}, Pu^{238}, and Cm^{244}.

(b) Those where the calculated damage from such an assumed release exceed sixty million dollars include: Co^{60}, Sr^{90}, Cs^{137}, Ce^{144}, Ir^{192}, Po^{210}, Pu^{238}, Pu^{239}, Cm^{242}, and Cm^{244}.

(c) Those where the calculated damage from such an assumed release was less than sixty million dollars include: Kr^{85}, Sb^{124}, I^{131}, Pm^{147}, Tm^{170}, and U^{233}.

(d) Those where the calculated damage from an assumed release of 100% of the assumed inventory (Table 2.3) would result in less than sixty million dollars include: Kr^{85}, I^{131} and Pm^{147}.

Major Elements in Calculating Damage Values

The following elements have a major effect in estimating damages and should be independently evaluated for specific situations:

Effect of Weather on Calculated Damage

The effect of various weather conditions on the calculated economic loss for the various damage categories is illustrated in Table 8.1, where the calculated values are presented for an assumed release of 50,000 curies of Sr^{90}. The calculated loss during conditions favorable for dispersion (sunny day) is lower by a factor of 2 x 10^3 than the calculated loss for the unfavorable conditions (washout and inversion). Inversion conditions maximize the loss from personnel exposure (Ranges A, B, and C). For the three isotopes with the largest calculated damage (Sr^{90}, Pu^{238}, or Cm^{244}) even under inversion conditions damage to personnel was only a small fraction of the total (<2%) with less than 100 people receiving Range A

Table 8.1. Effect of Weather on Calculated Economic Loss Due to a Hypothetical Accident Involving a Release of 50,000 Curies Sr^{90}

Loss Range	Potential Economic Loss During: Inversion	Washout	Sunny Day
I	$\$1.0 \times 10^9$	$\$1.6 \times 10^9$	$\$9.0 \times 10^5$
II	2.3×10^9	3.0×10^9	5.8×10^5
III	4.0×10^8	4.2×10^8	6.0×10^4
A	1.3×10^6		
B	1.0×10^7	1.6×10^4	2.8×10^4
C	5.0×10^7	1.6×10^5	5.2×10^4
Total	$\$3.8 \times 10^9$	$\$ 5 \times 10^9$	$\$1.6 \times 10^6$

exposures. Washout maximizes calculated damage from area contamination (Ranges I, II and III), but the effect is not significantly higher than the area contamination damage during inversion conditions.

Effect of Site

Since the dollar loss is primarily dependent of the value of buildings, land, and crops surrounding the site, it is obvious that the potential loss could vary tremendously depending on whether the site is, for example, in the Nevada desert or in the San Joaquin Valley. It is important, therefore, in applying this type of analysis to specific situations that the characteristics of the area surrounding the site be factored into the analysis rather than using the figures in this report which are for an average site.

Effect of Fractional Release

The fractional release in the case of an accident has an obvious effect on the potential damage. The fraction released assumed for the accidents postulated in this report is believed to be conservative (high). The research in release fraction and aerosol properties in reactor fuel

meltdown,[1,2,3,4] and the efficiency of reactor safeguards systems, has put reactor accident evaluation on a much more reasonable basis than was possible at the time WASH-740 (ref 5) was written. Similar improvement over the analyses in this report can be expected from research in release fractions, aerosol properties, effect of safeguards systems in radiochemical plant accident situations.

REFERENCES (SECTION 8)

1. ORNL Nuclear Safety Semiannual Report, ORNL-3319.
2. ORNL Nuclear Safety Semiannual Report, ORNL-3401.
3. ORNL Nuclear Safety Semiannual Report, ORNL-3483.
4. ORNL Nuclear Safety Semiannual Report, ORNL-3518.
5. Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants, WASH-740, USGPO, p 78 (Mar. 1957).

APPENDIX A

Appendix A contains graphs presenting the potential economic loss for 16 isotopes as a function of the amount released, the population distribution and the weather conditions. Table A-1 is an index of Figs. A-1 through A-30. The economic loss plots for Sr^{90} and mixed fission products from a criticality accident are found in Section 6, Figs. 6.1 through 6.4, pages 78-80 and 84.

Table A1. Index of Economic-Loss Plots in Appendix A

Isotope	Typical Population Distribution		Uniform Population Distribution, 100 people/mi^2	
	Figure	Page	Figure	Page
Co^{60}	A-1	110	A-2	111
Kr^{85}	A-3	112	A-3	112
Sb^{124}	A-4	113	A-5	114
I^{131}	A-6	115	A-6	115
Cs^{137}	A-7	116	A-8	117
Ce^{144}	A-9	118	A-10	119
Pm^{147}	A-11	120	A-12	121
Tm^{170}	A-13	122	A-14	123
Ir^{192}	A-15	124	A-16	125
Po^{210}	A-17	126	A-18	127
U^{233}	A-19	128	A-20	129
U^{235}	A-21	130	A-22	131
Pu^{238}	A-23	132	A-24	133
Pu^{239}	A-25	134	A-26	135
Cm^{242}	A-27	136	A-28	137
Cm^{244}	A-29	138	A-30	139

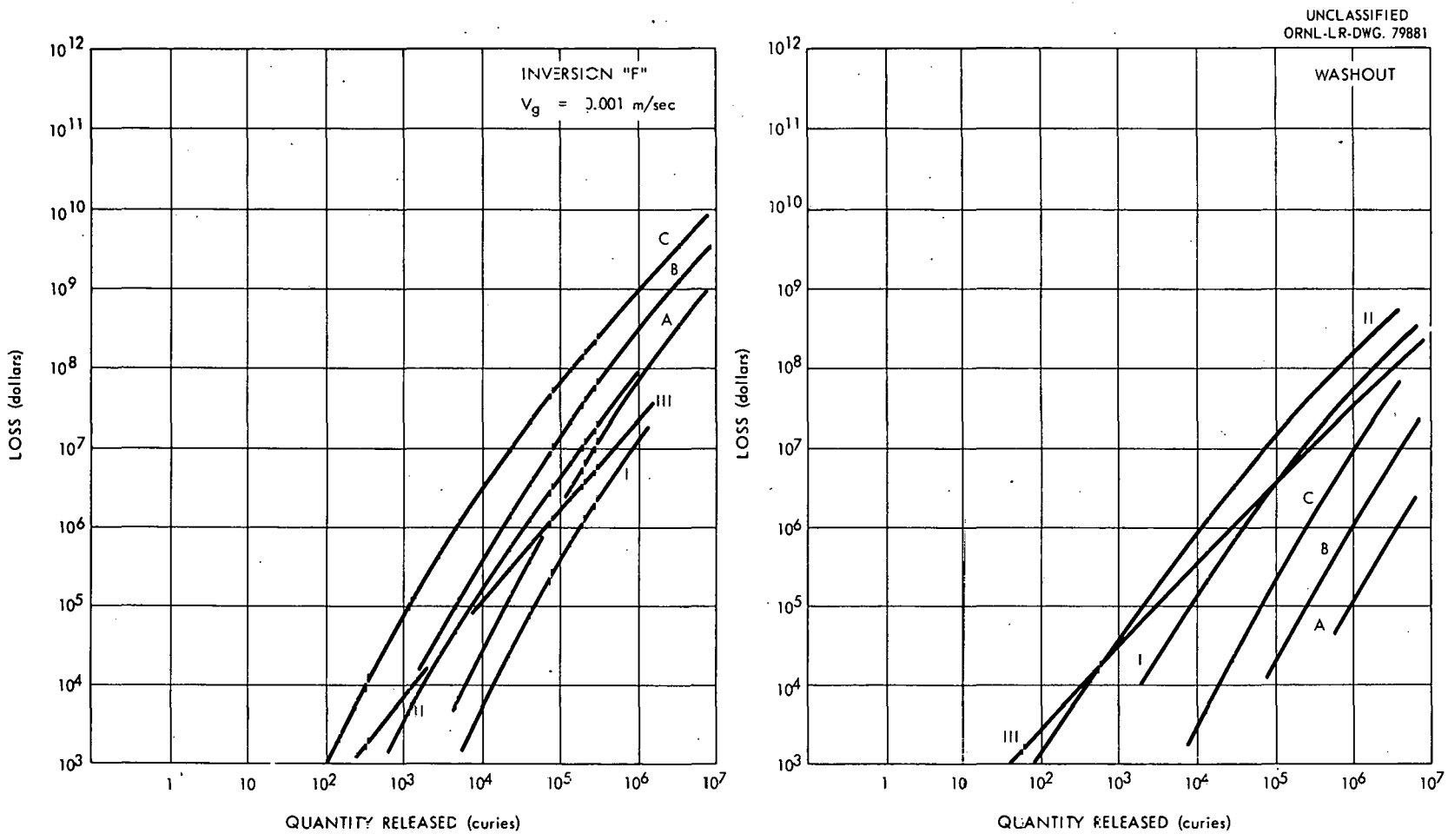

Fig. A1. Potential Economic Loss Resulting from Release of Co^{60}. Typical population distribution.

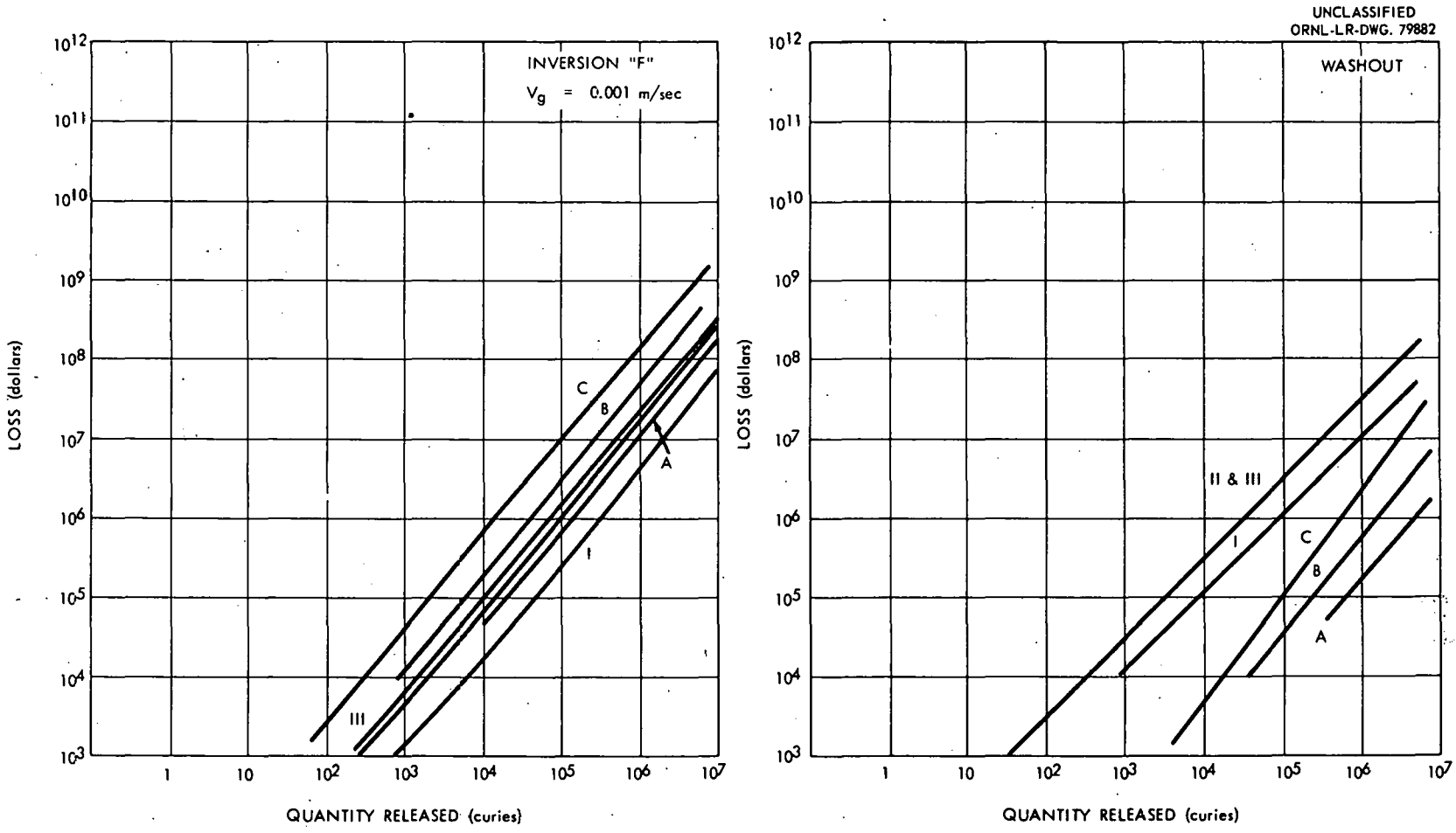

Fig. A2. Potential Economic Loss Resulting from Release of Co^{60}. One hundred persons per square mile.

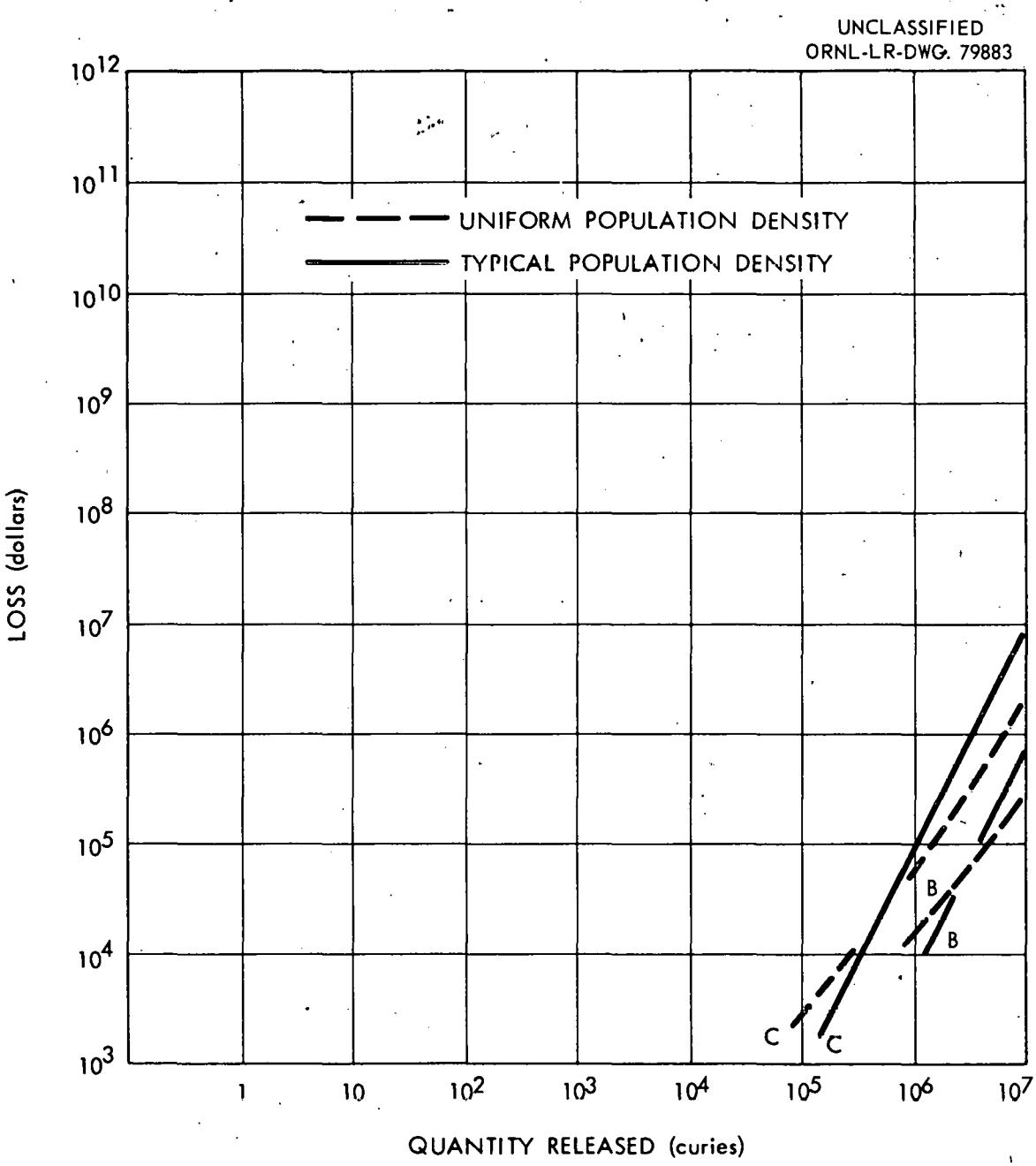

Fig. A3. Potential Economic Loss Resulting from Release of Kr^{85}.

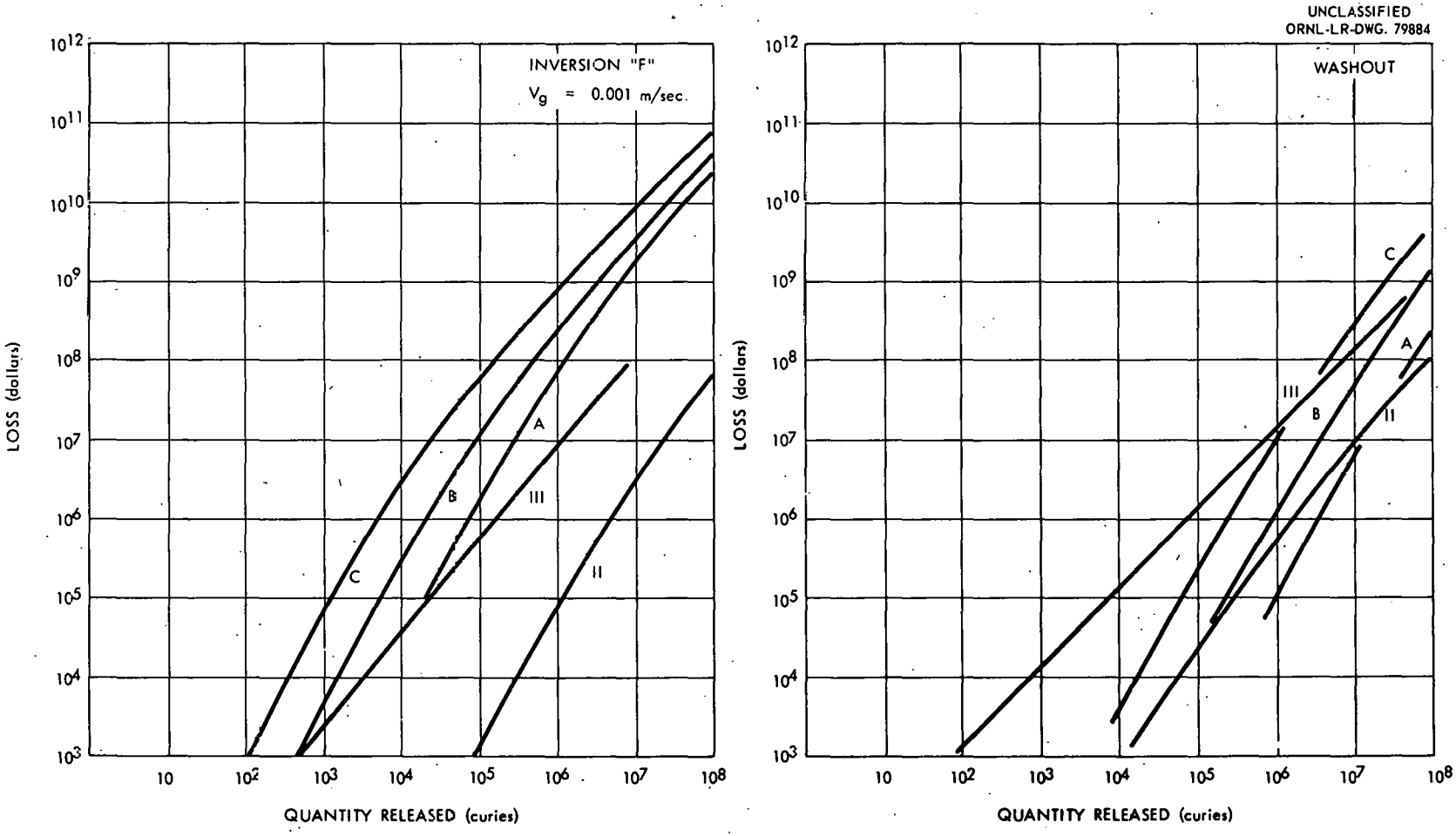

Fig. A4. Potential Economic Loss Resulting from Release of Sb^{124}. Typical population distribution.

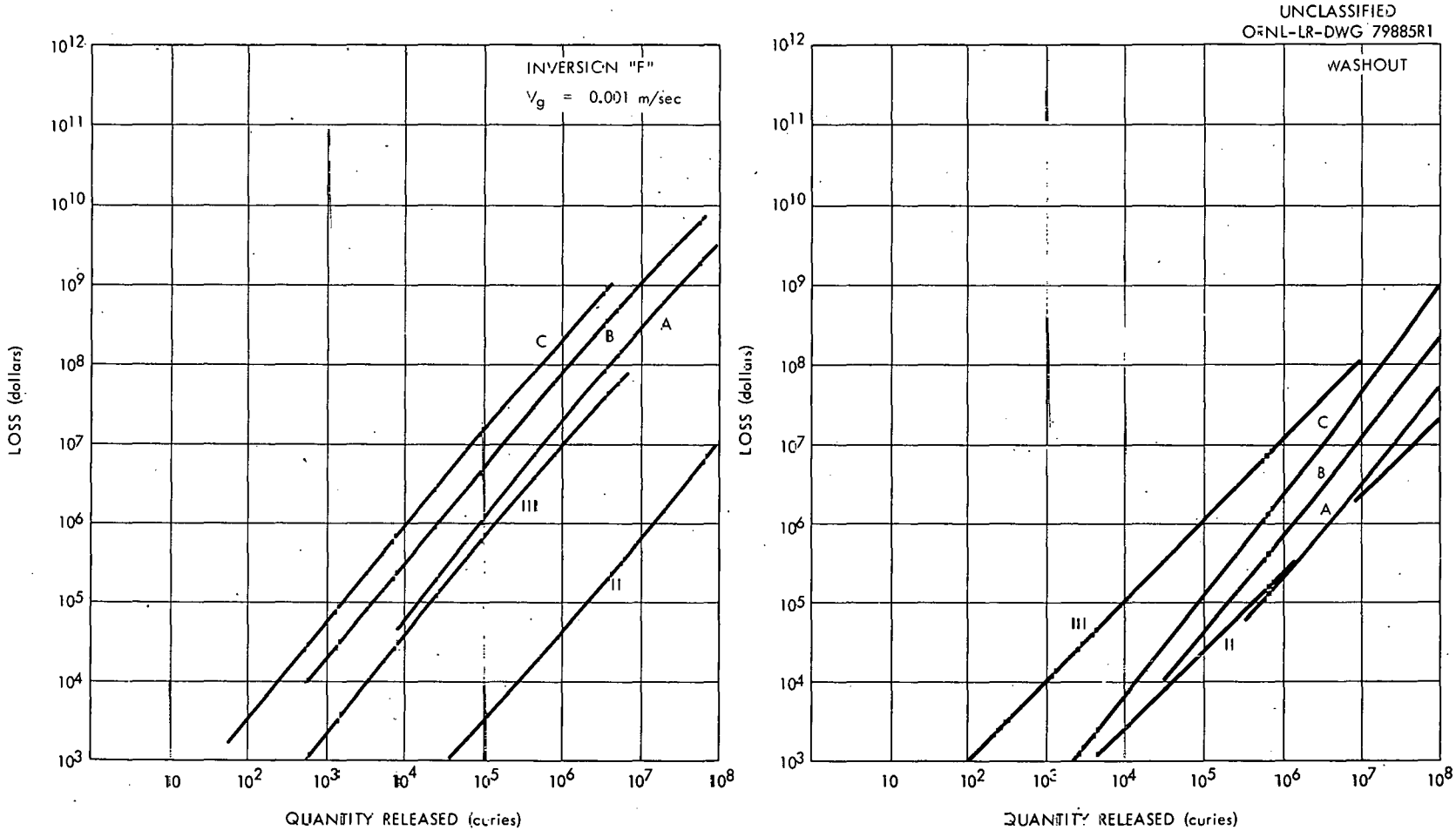

Fig. A5. Potential Economic Loss Resulting from Release of Sb^{124}. One hundred persons per square mile.

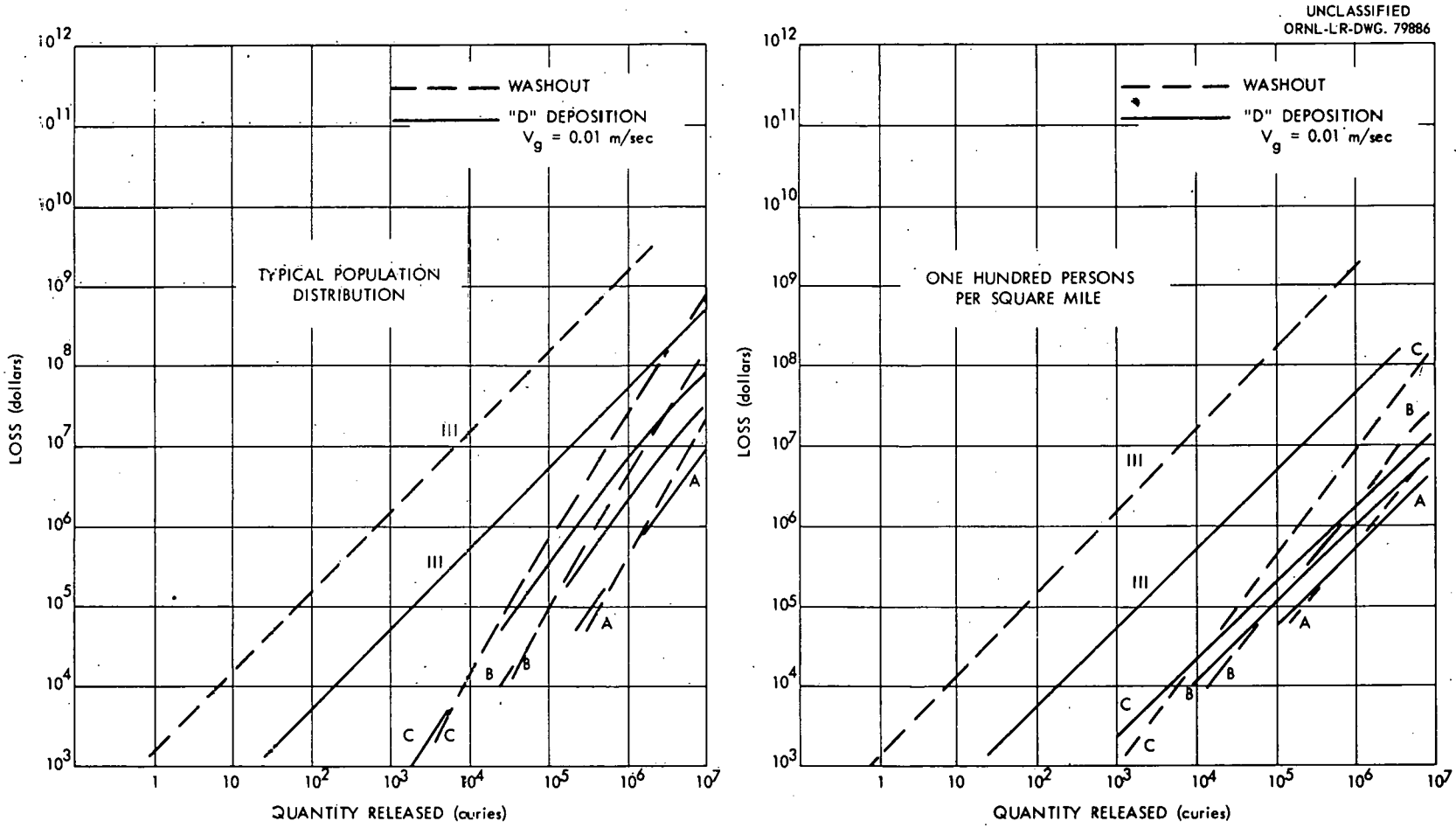

Fig. A6. Potential Economic Loss Resulting from Release of I^{131}. Typical population distribution.

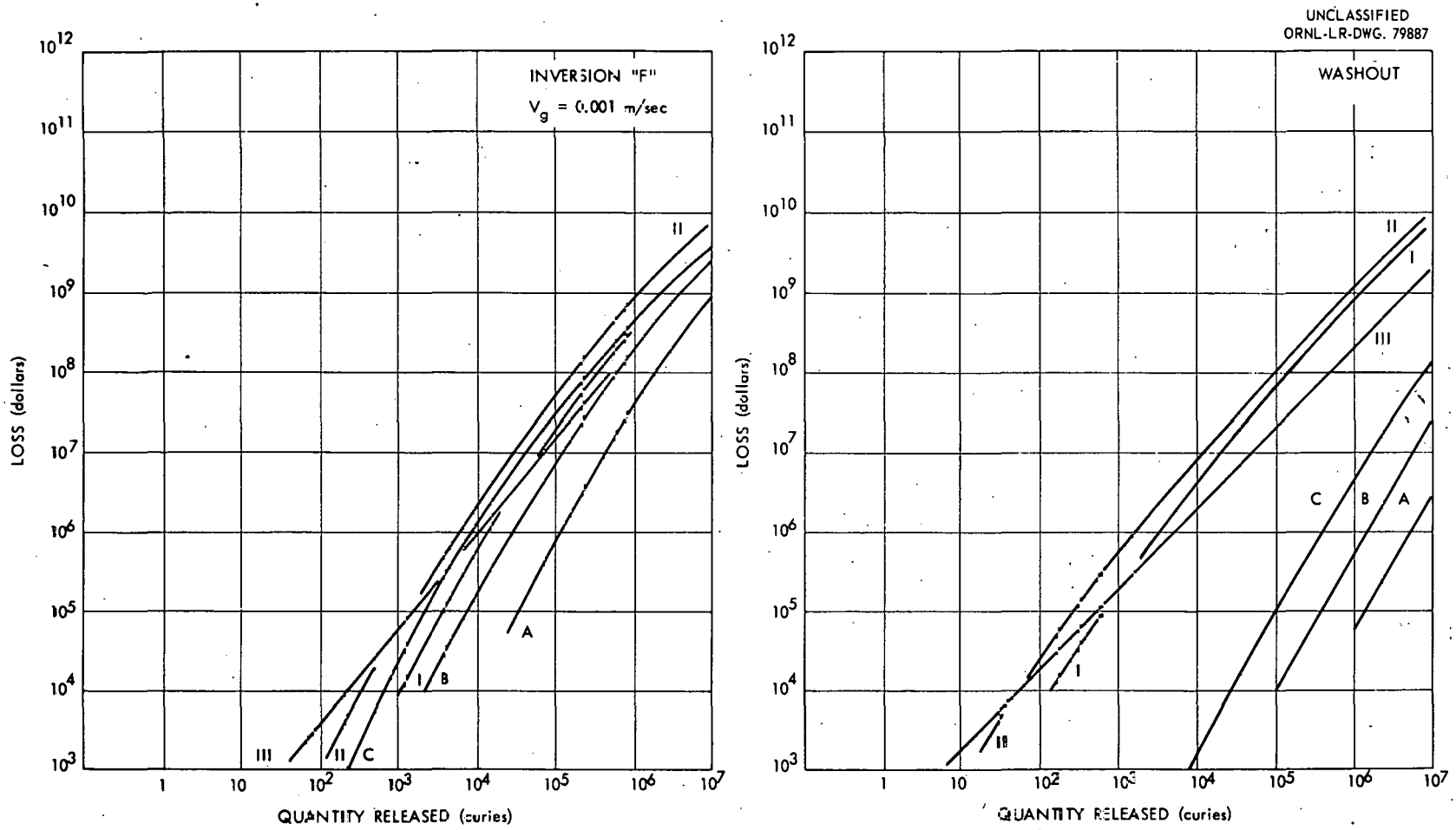

Fig. A7. Potential Economic Loss Resulting from Release of Cs^{137}. Typical population distribution.

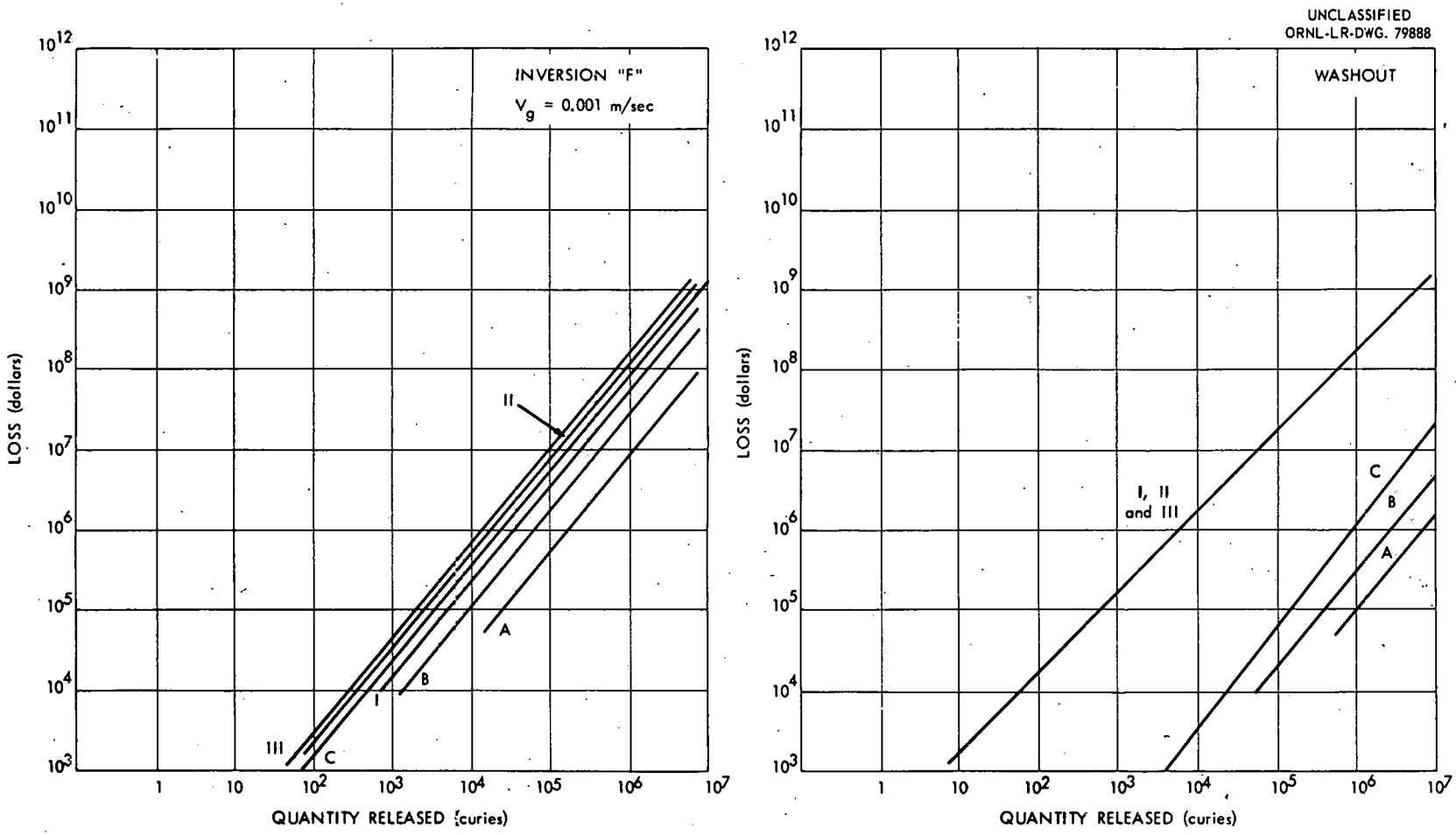

Fig. A8. Potential Economic Loss Resulting from Release of Cs^{137}. One hundred persons per square mile.

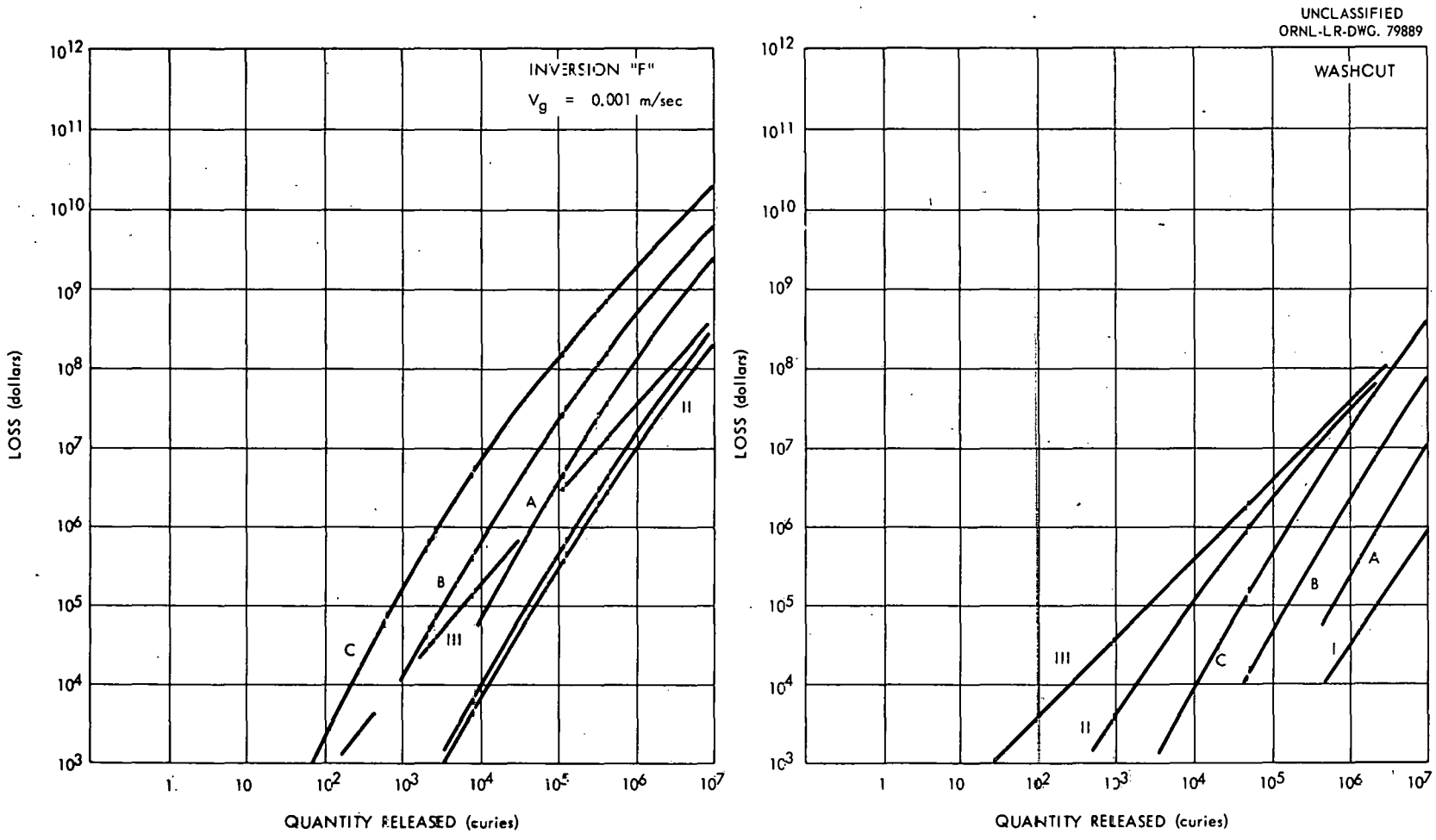

Fig. A9. Potential Economic Loss Resulting from Release of Ce^{144}. Typical population distribution.

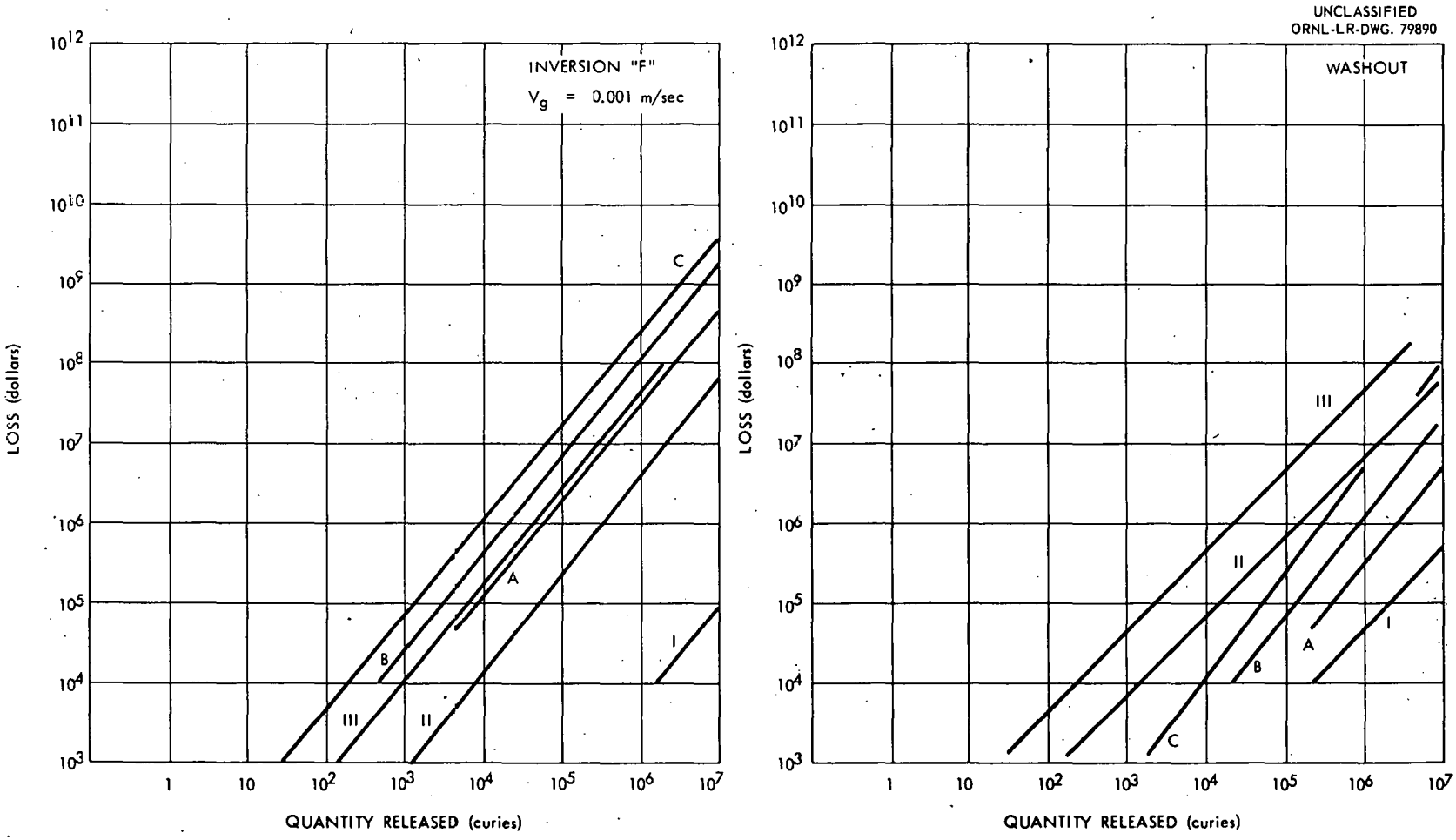

Fig. A1C. Potential Economic Loss Resulting from Release of Ce^{144}. One hundred persons per square mile.

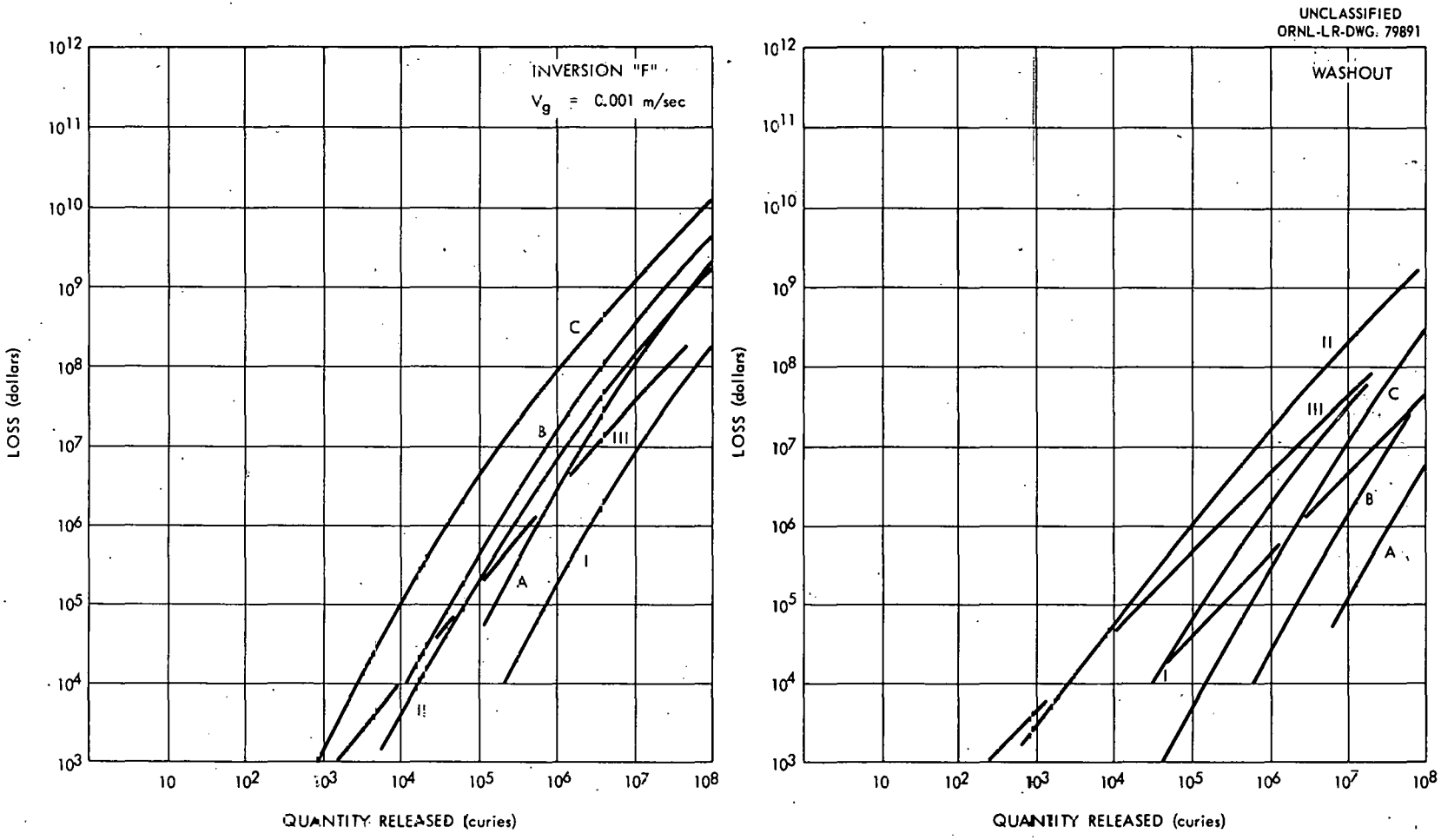

Fig. All. Potential Economic Loss Resulting from Release of Pm^{147}. Typical population distribution.

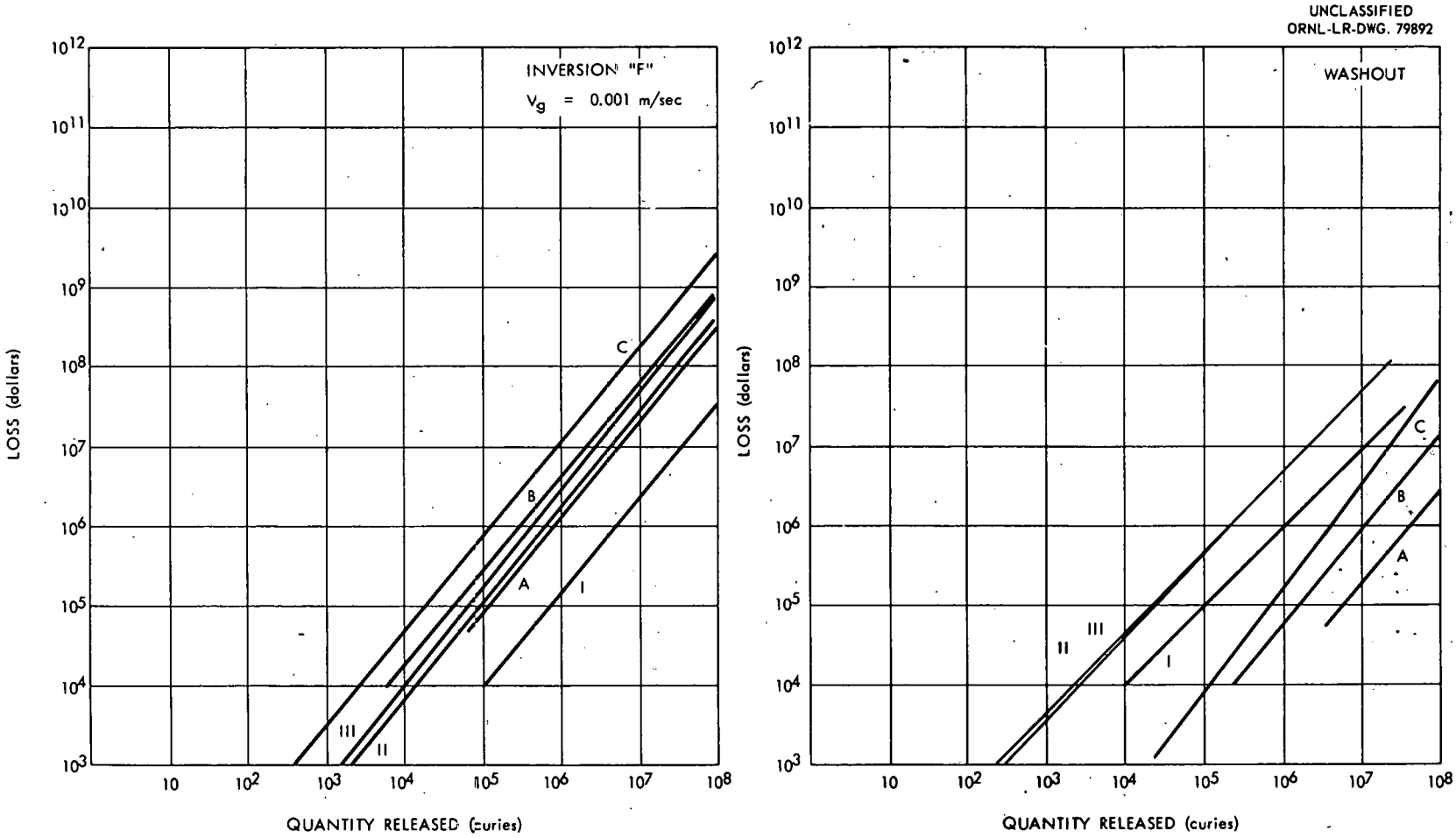

Fig. A12. Potential Economic Loss Resulting from Release of Pm^{147}. One hundred persons per square mile.

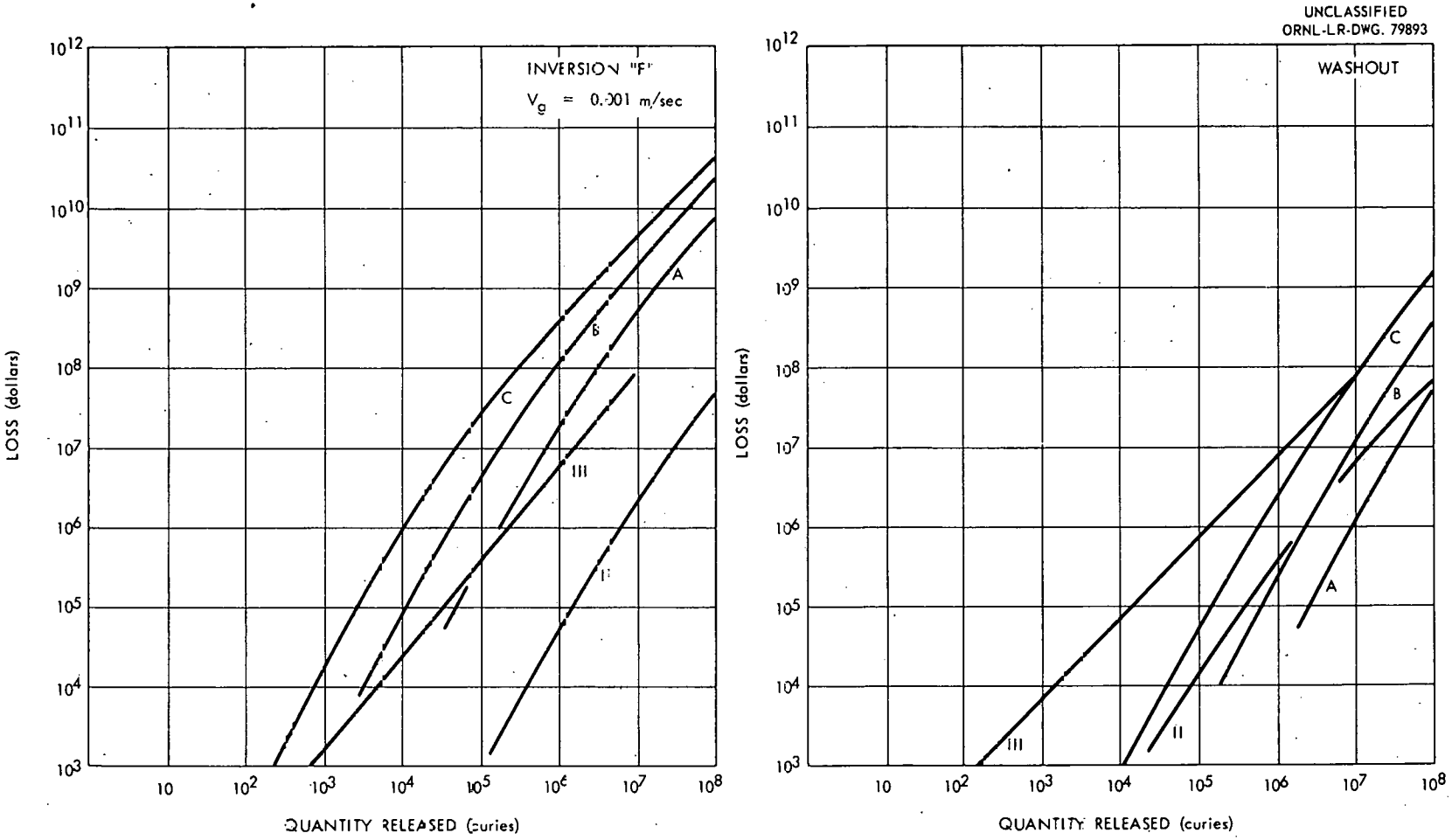

Fig. A13. Potential Economic Loss Resulting from Release of Tm^{167}. Typical population distribution.

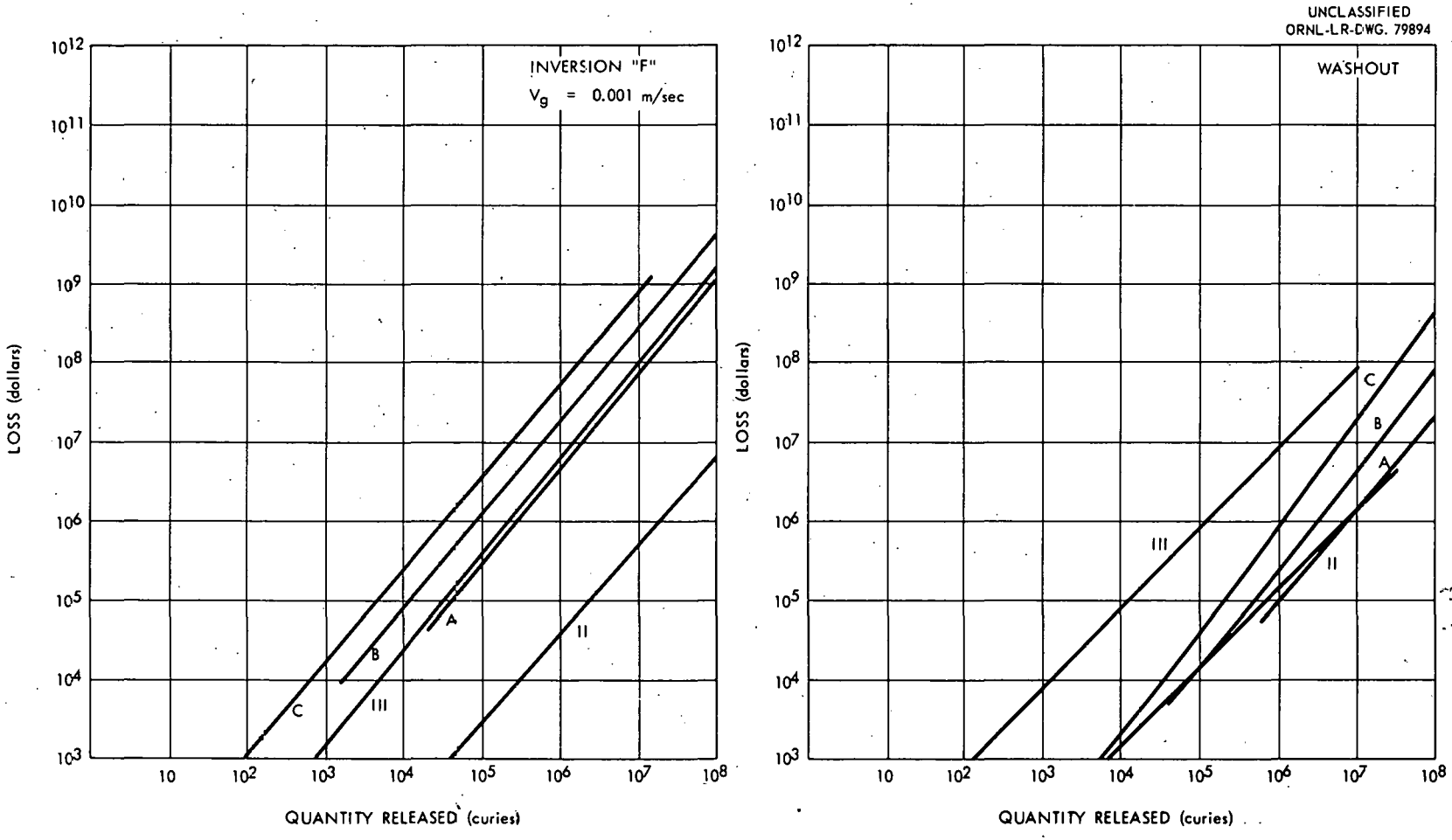

Fig. A14. Potential Economic Loss Resulting from Release of Tm^{170}. One hundred persons per square mile.

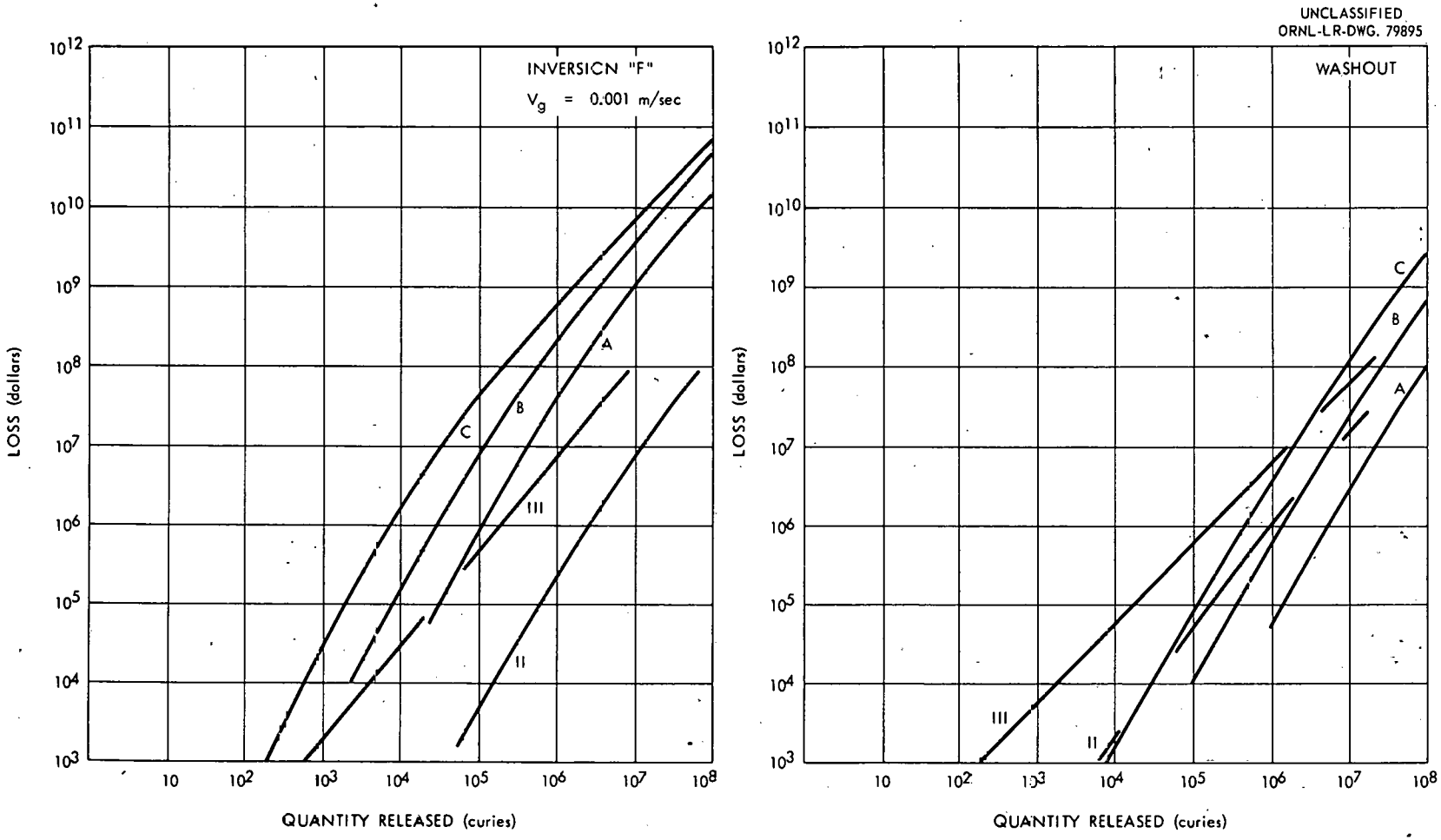

Fig. A15. Potential Economic Loss Resulting from Release of Ir^{192}. Typical population distribution.

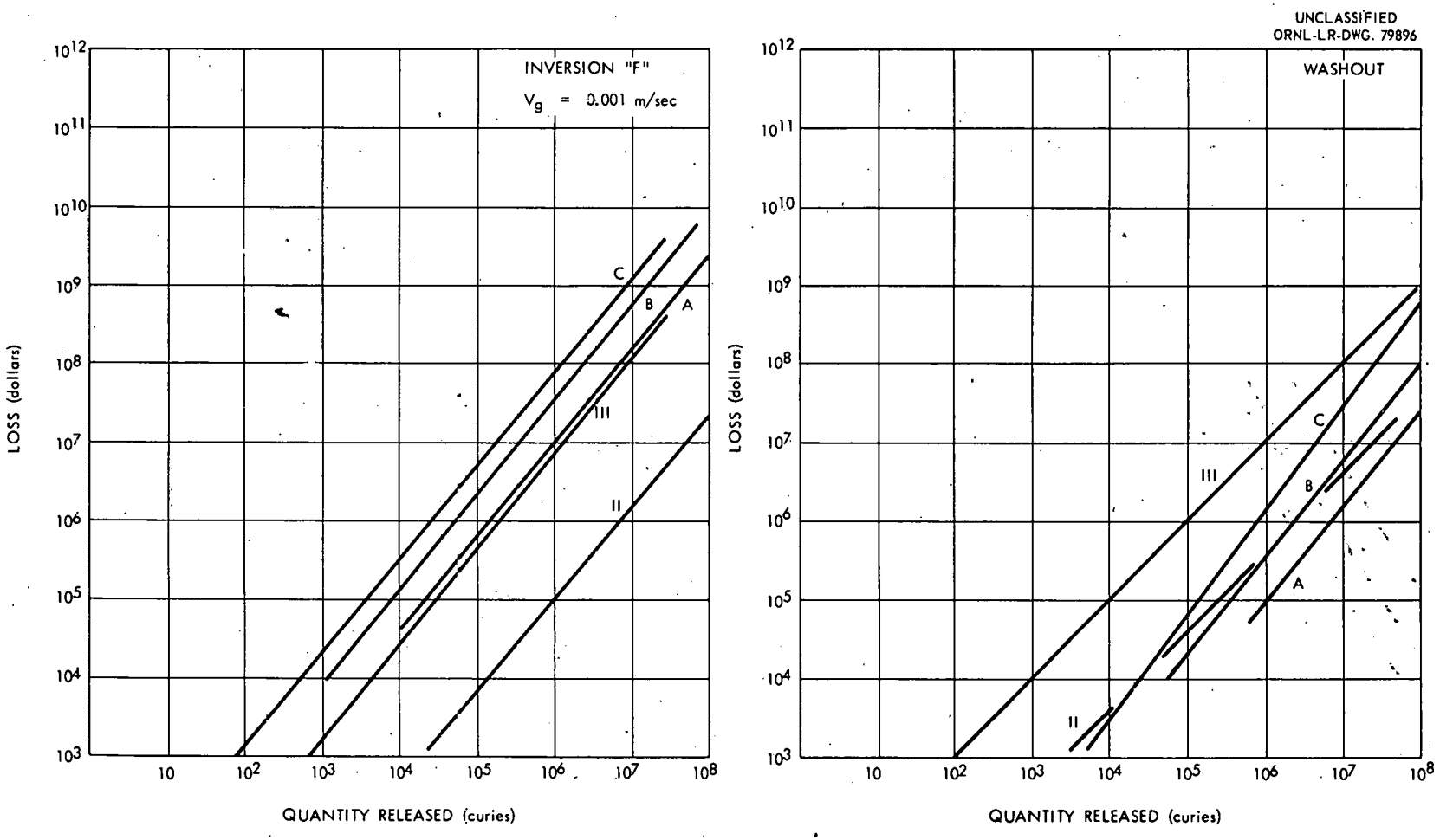

Fig. A16. Potential Economic Loss Resulting from Release of Ir^{192}. One hundred persons per square mile.

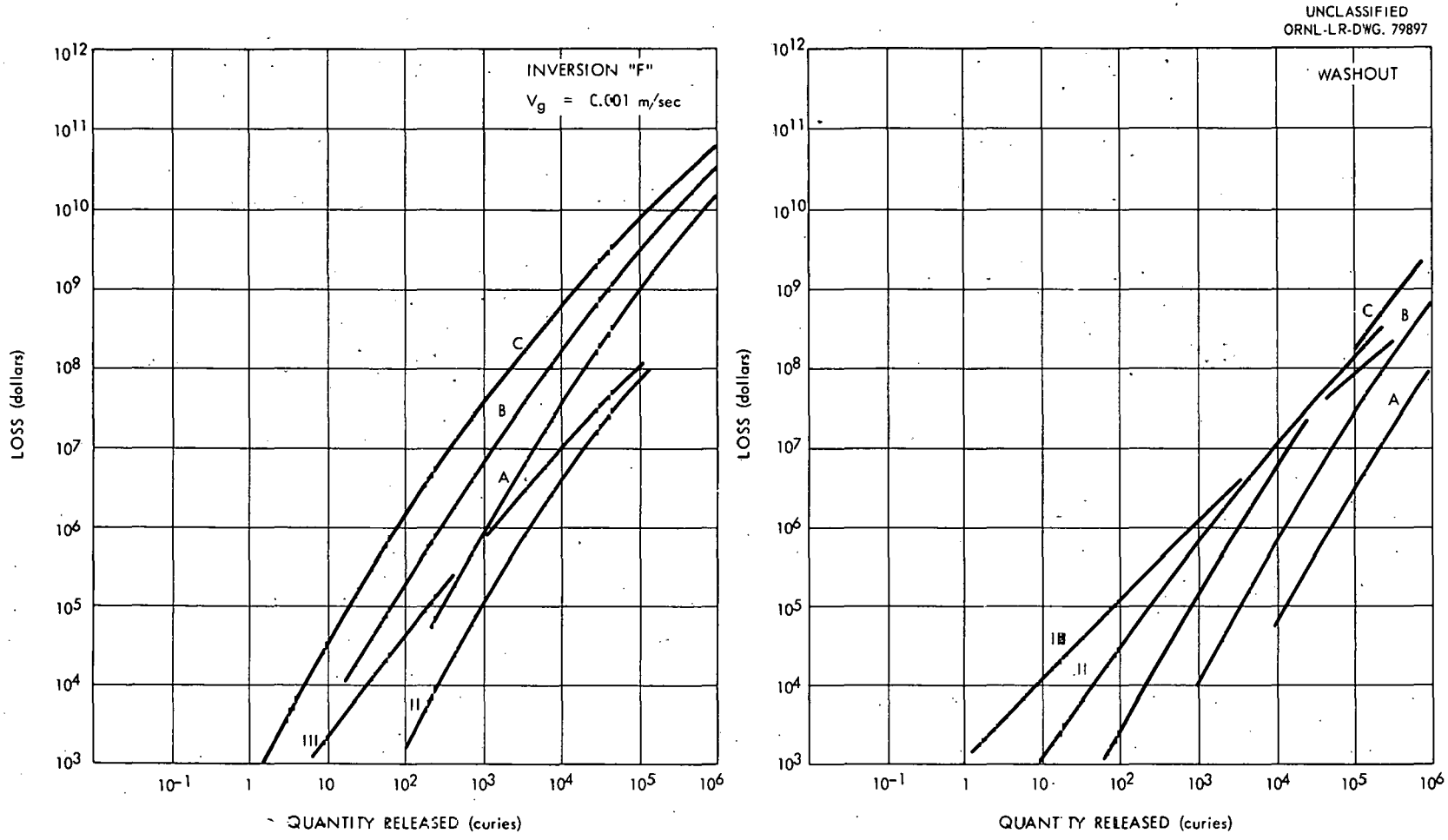

Fig. A17. Potential Economic Loss Resulting from Release of Po^{210}. Typical population distribution.

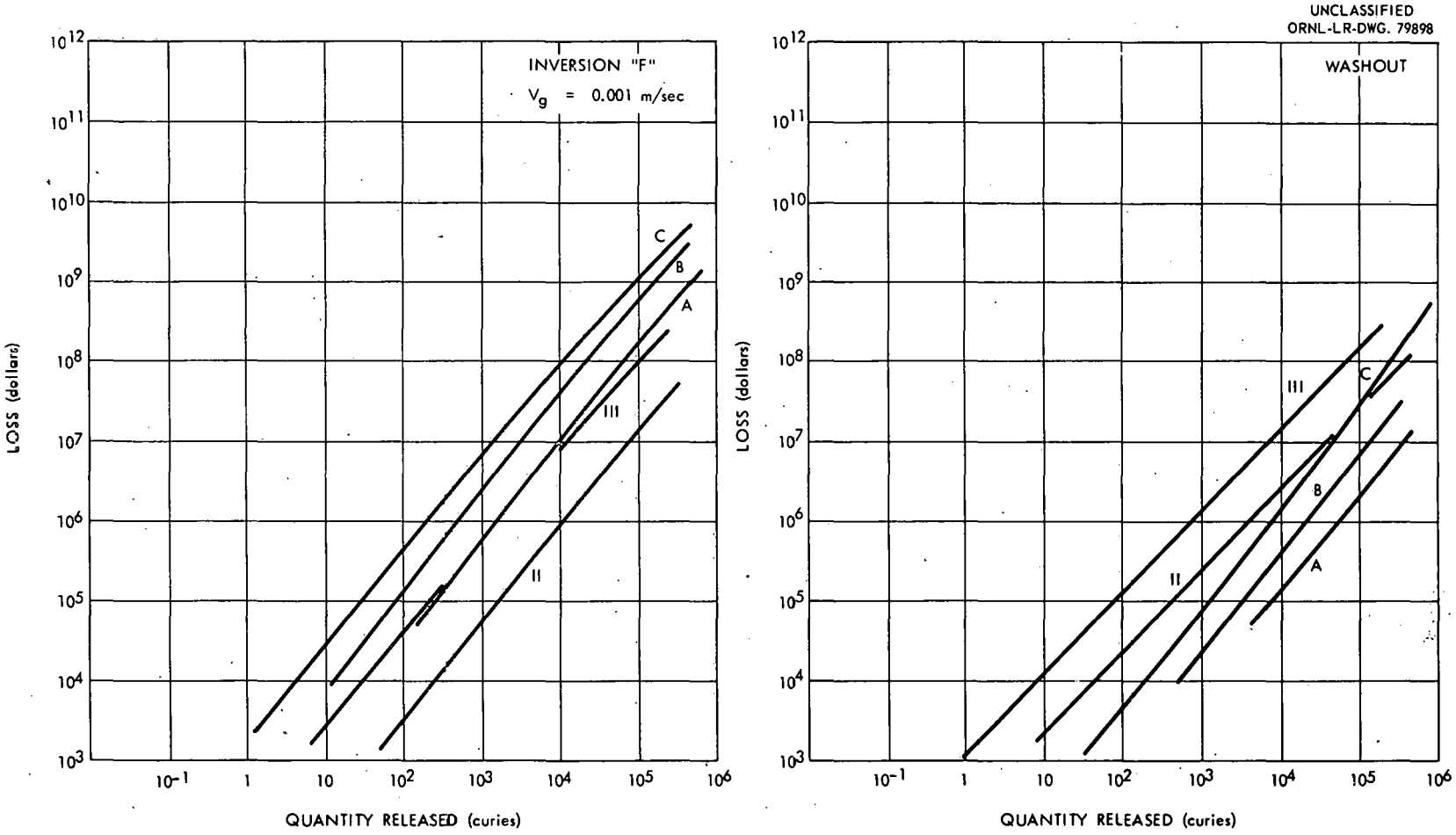

Fig. A18. Potential Economic Loss Resulting from Release of Po^{210}. One hundred persons per square mile.

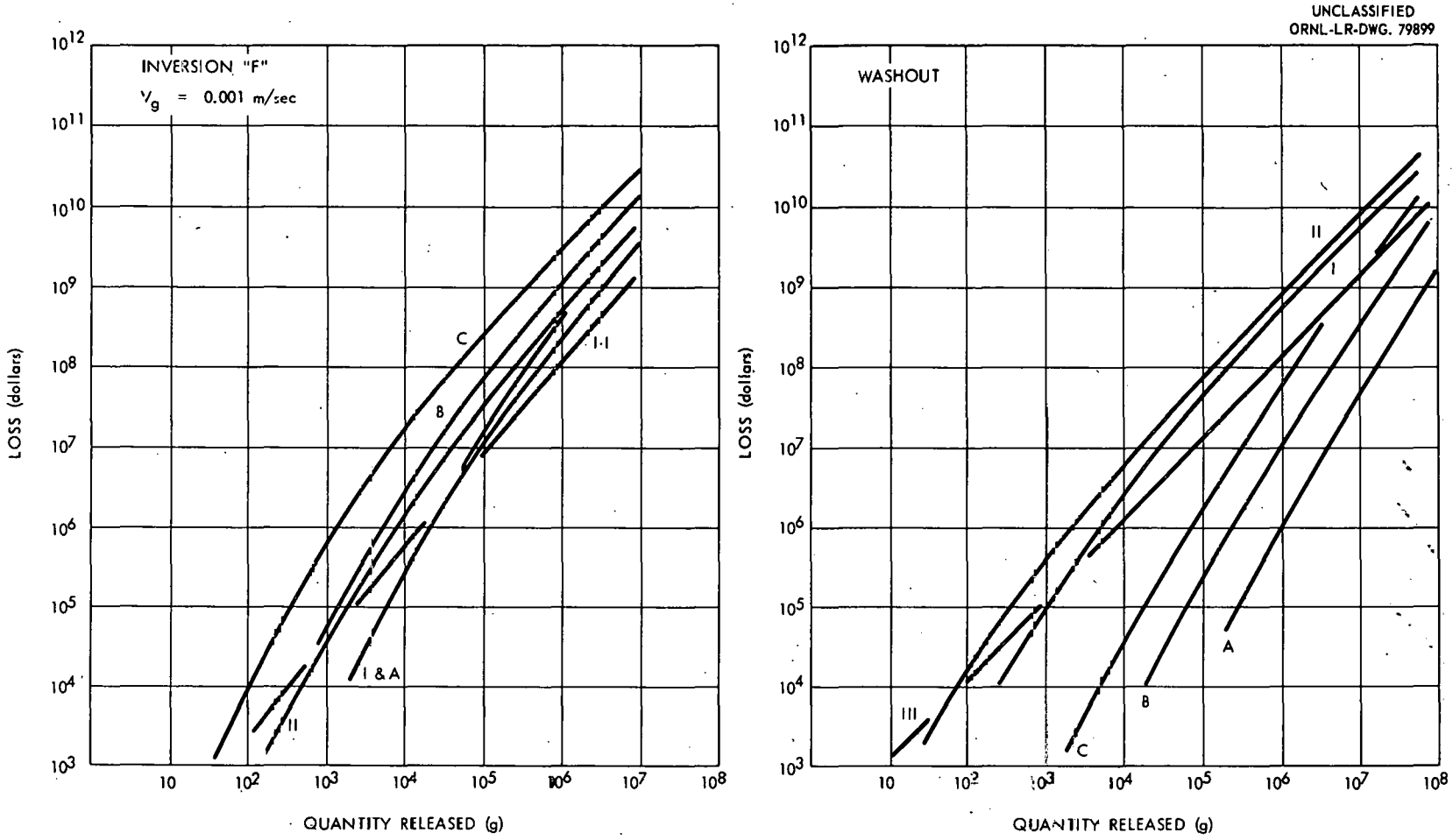

Fig. A19. Potential Economic Loss Resulting from Release of U^{233}. Typical population distribution.

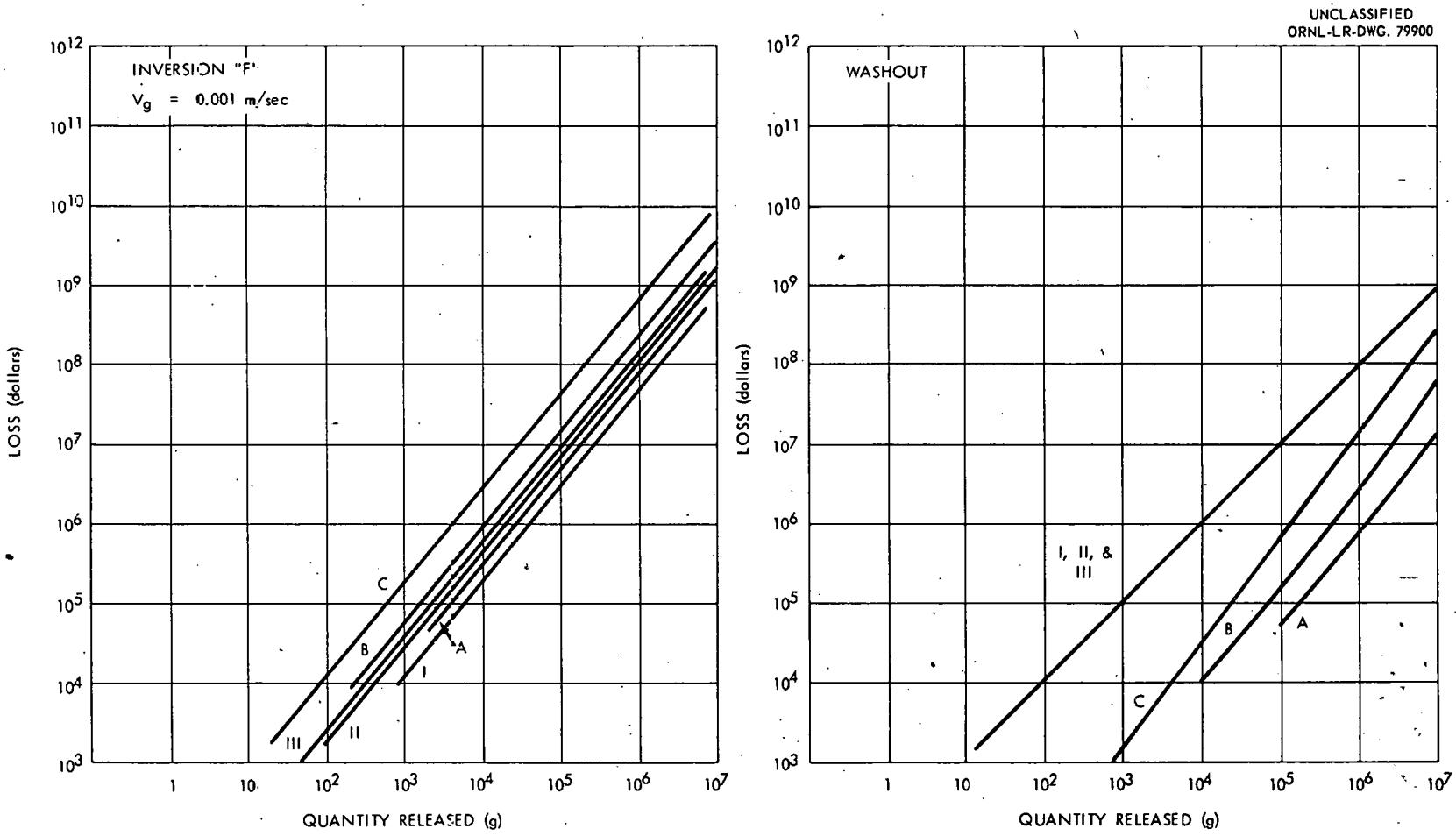

Fig. A20. Potential Economic Loss Resulting from Release of U^{233}. One hundred persons per square mile.

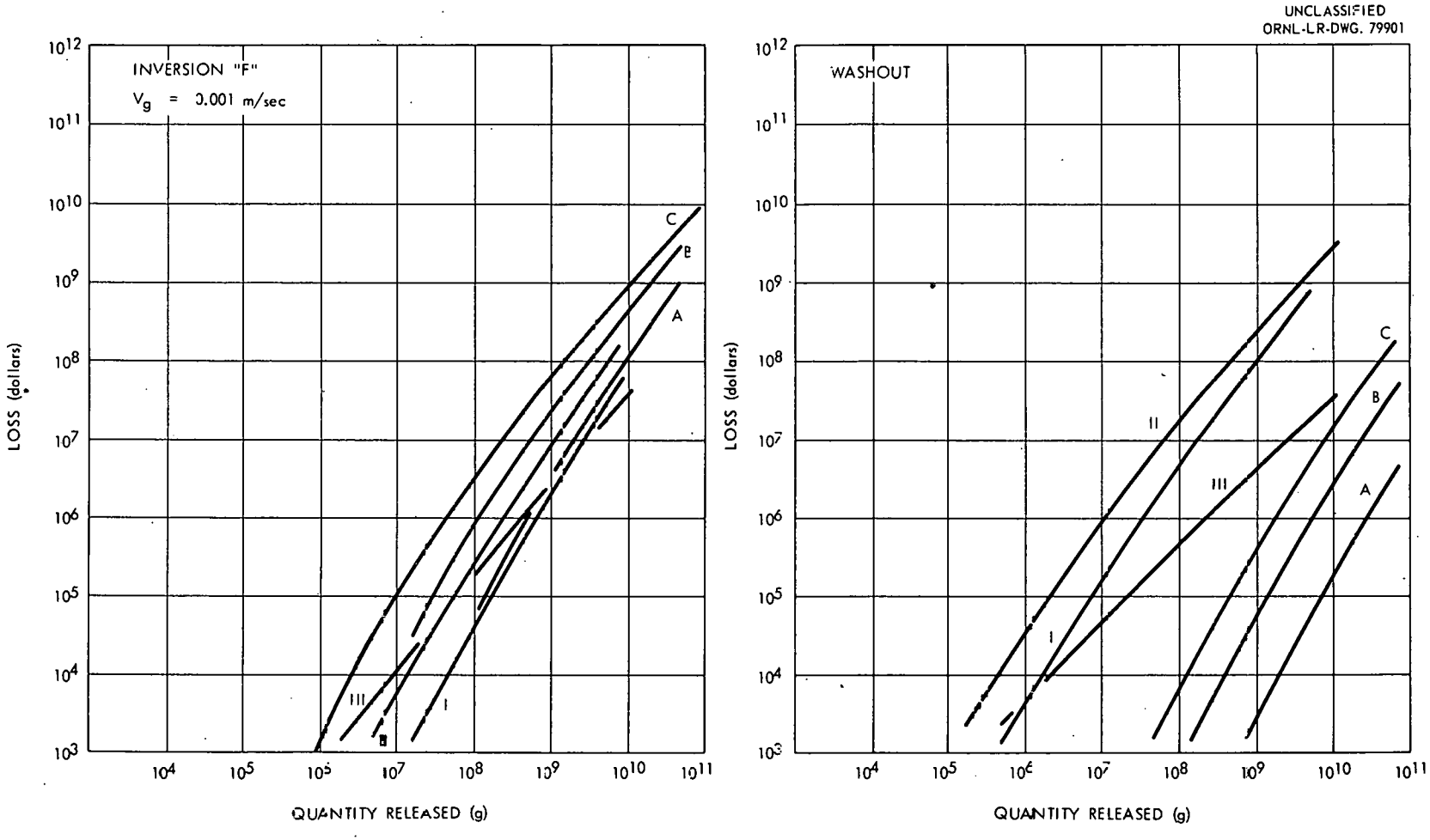

Fig. A21. Potential Economic Loss Resulting from Release of U^{235}. Typical population distribution.

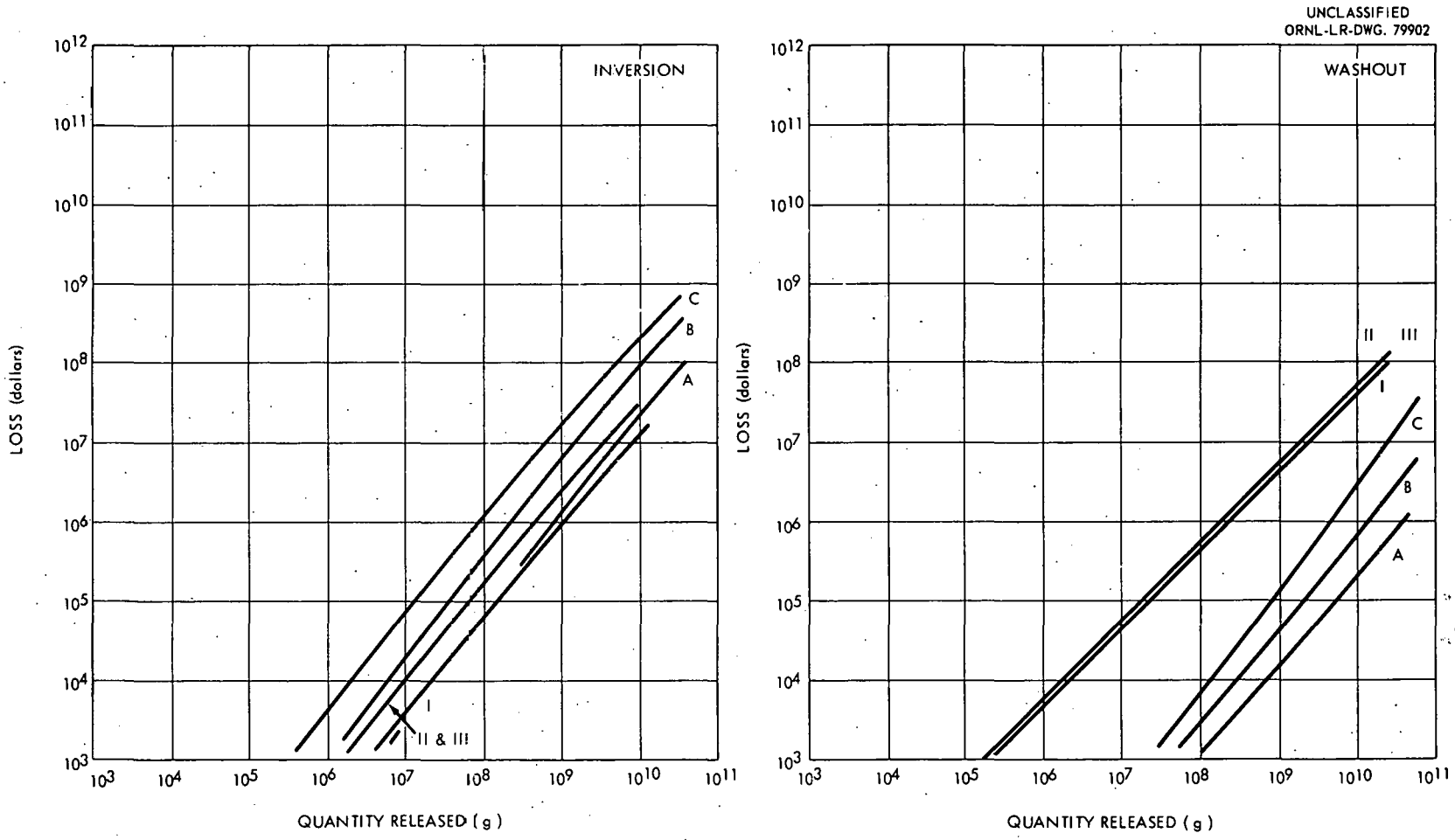

Fig. A22. Potential Economic Loss Resulting from Release of U^{235}. One hundred persons per square mile.

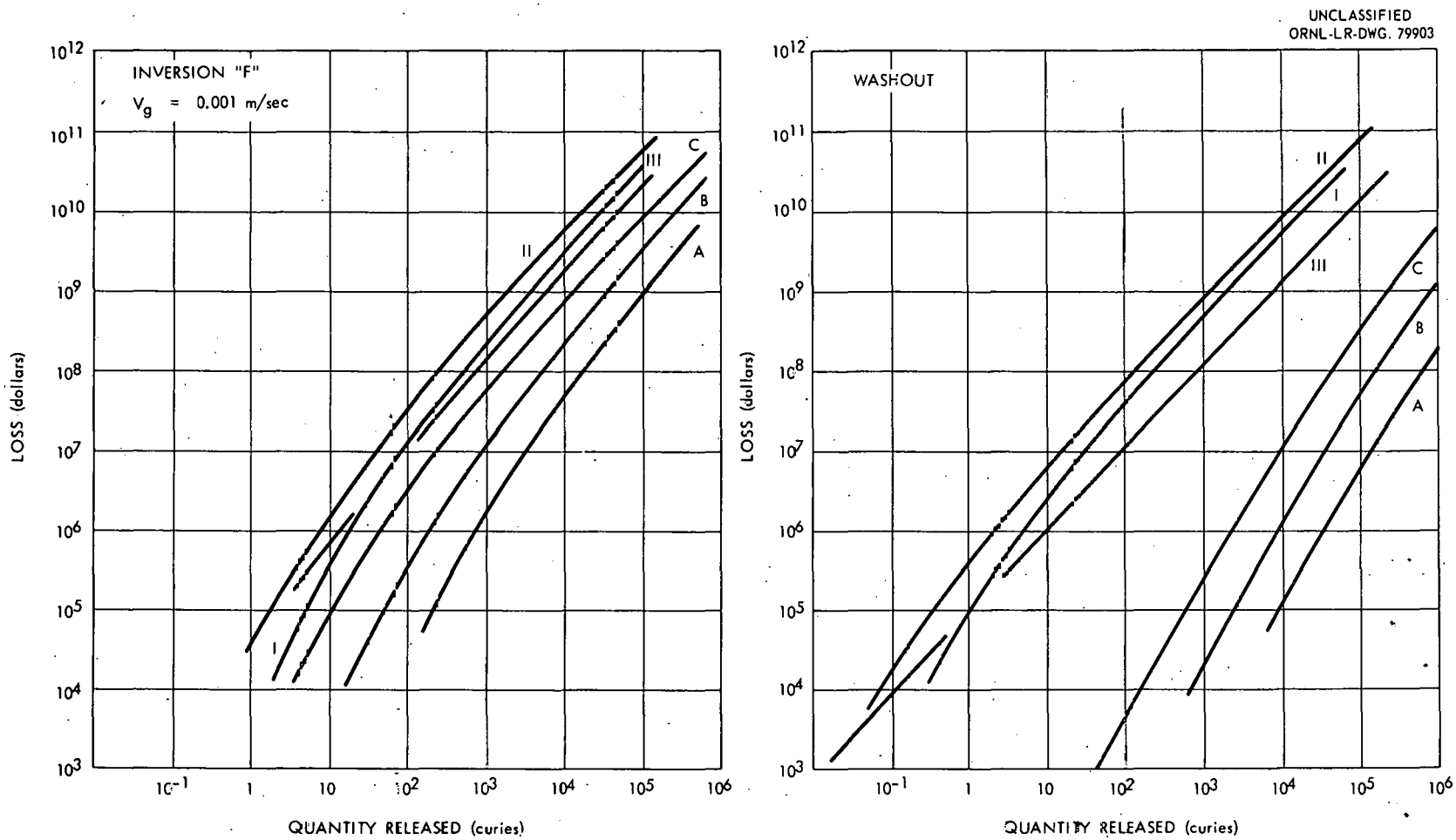

Fig. A23. Potential Economic Loss Resulting from Release of Pu^{238}. Typical population distribution.

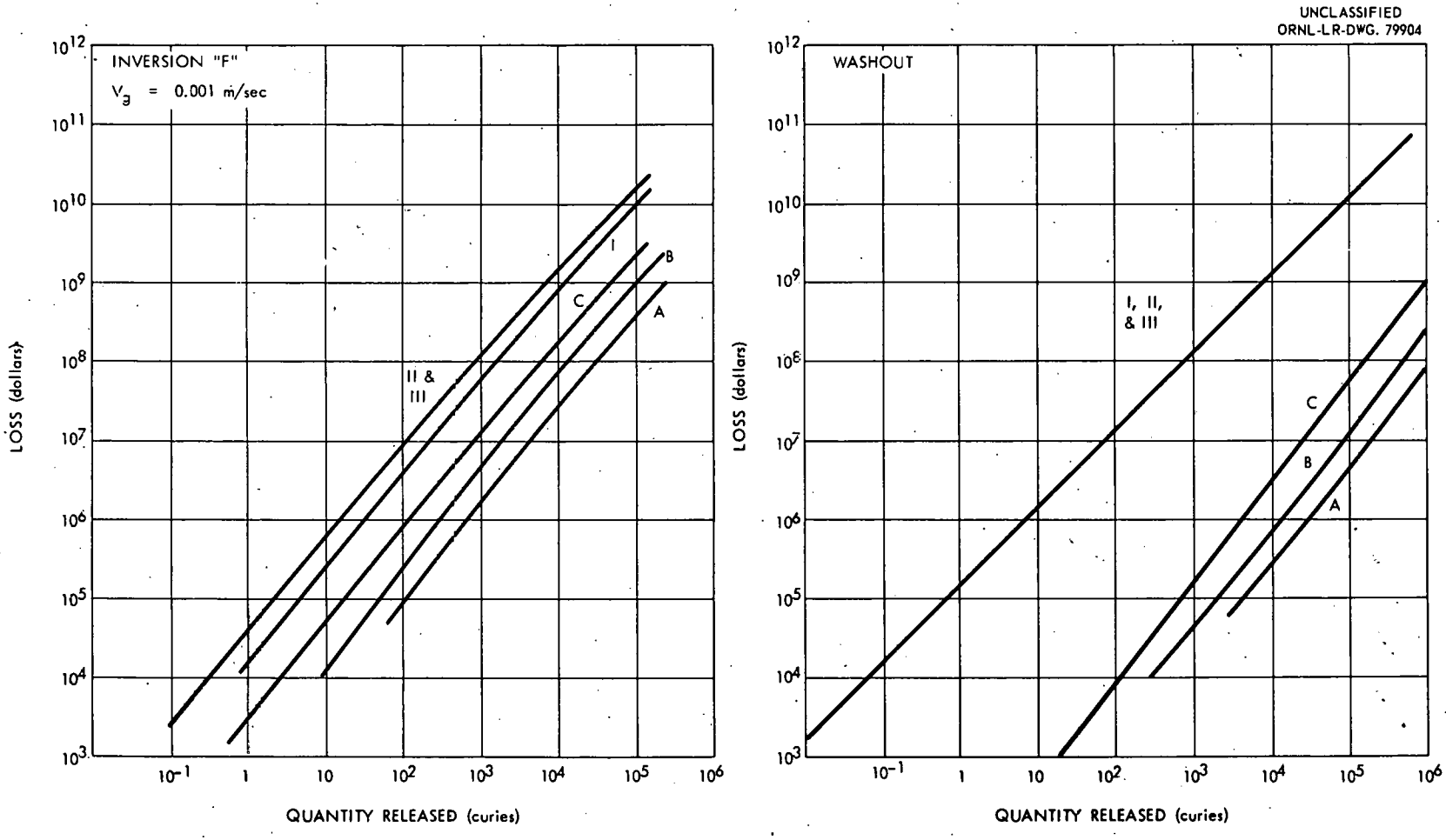

Fig. A24. Potential Economic Loss Resulting from Release of Pu^{238}. One hundred persons per square mile.

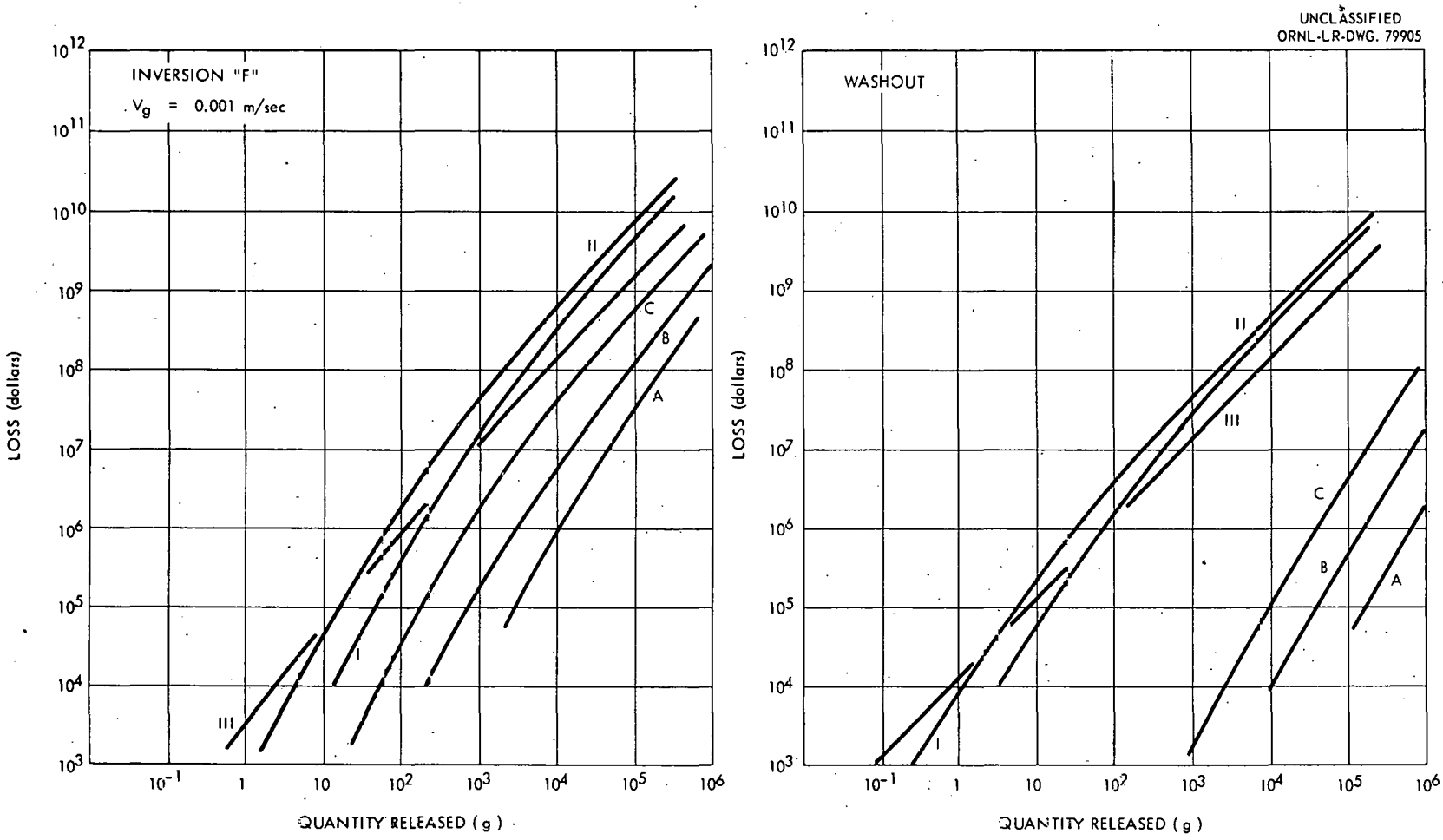

Fig. A25. Potential Economic Loss Resulting from Release of Pu^{239}. Typical population distribution.

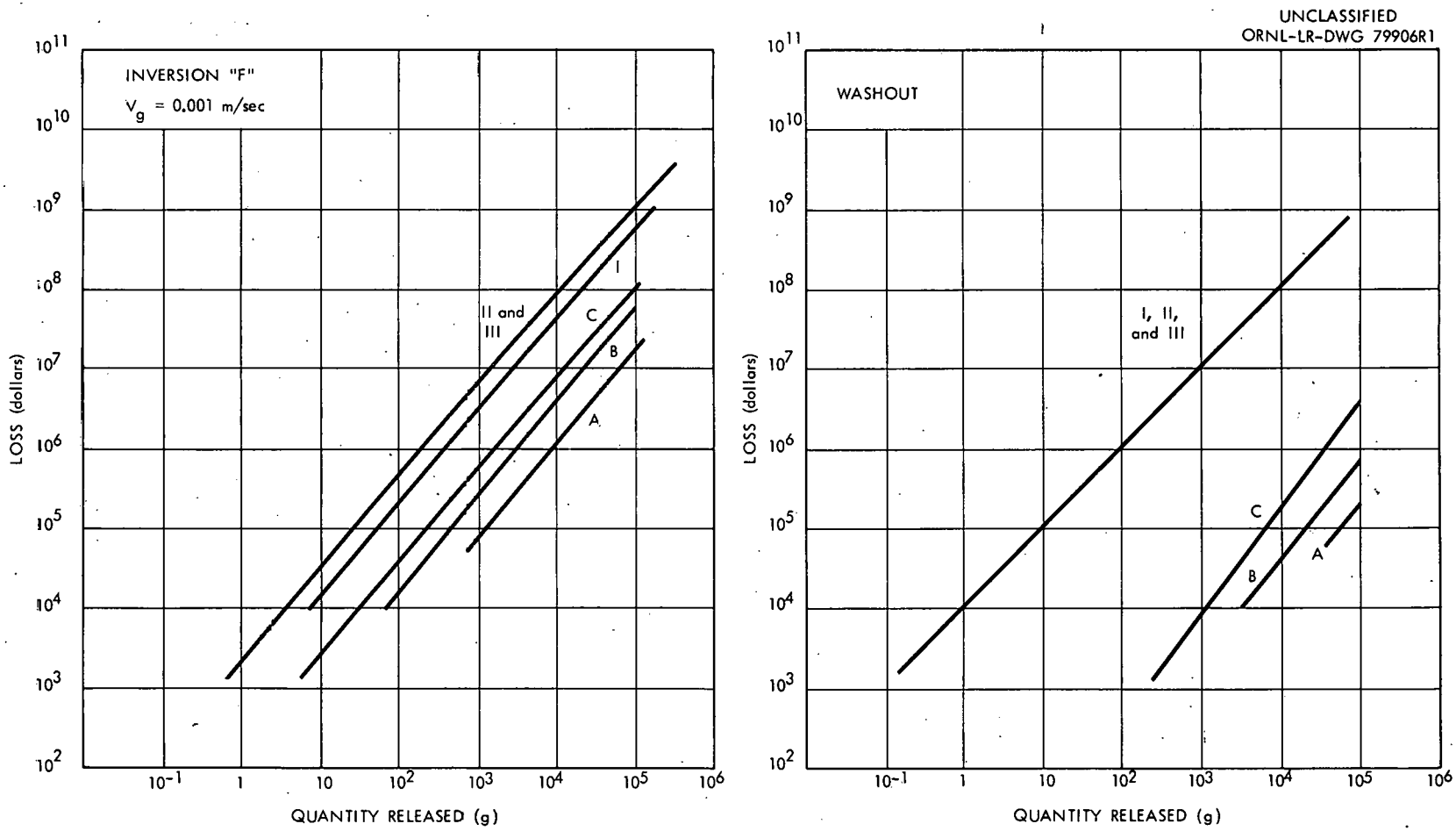

Fig. A26. Potential Economic Loss Resulting from Release of Pu^{239}. One hundred persons per square mile.

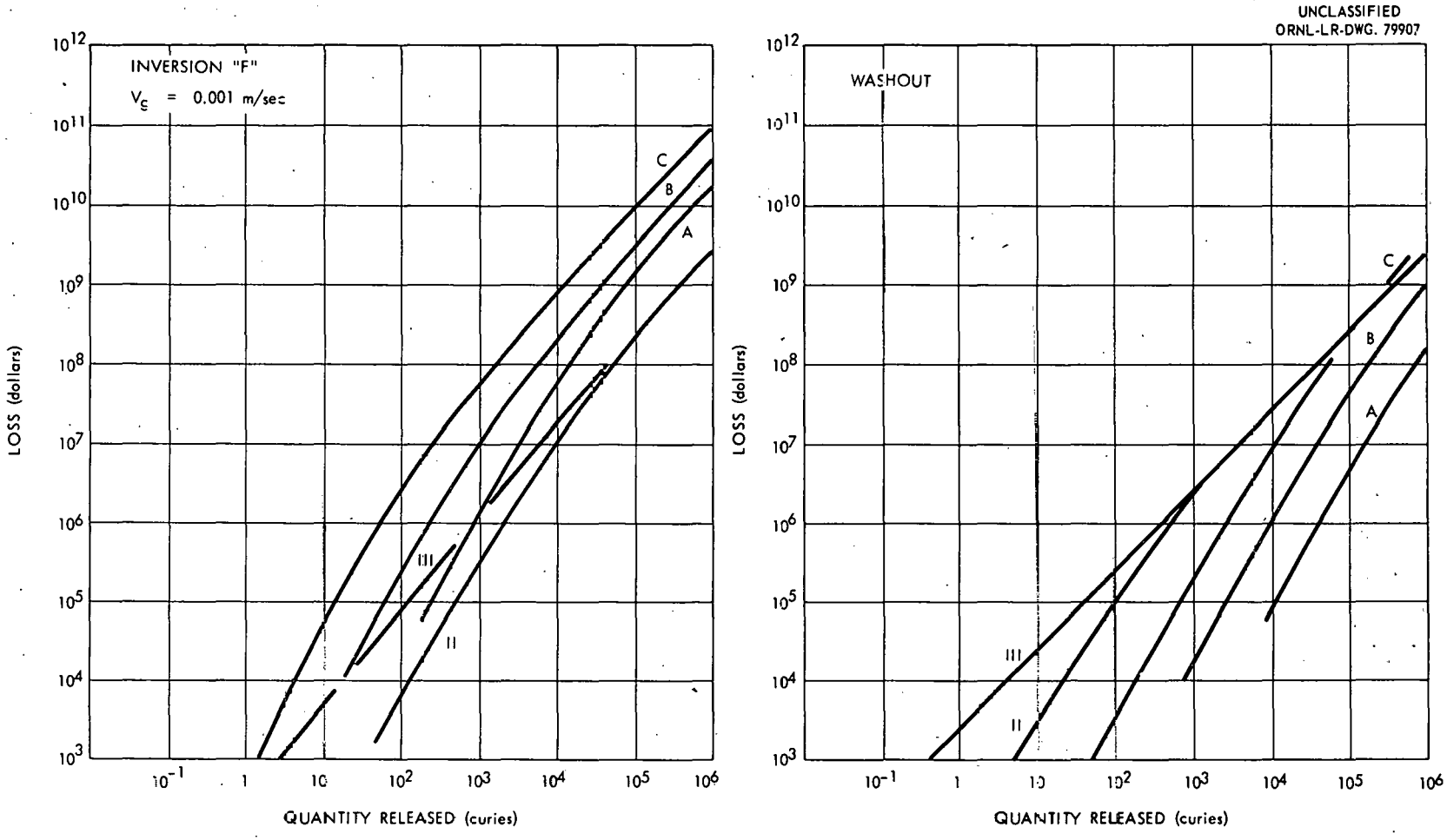

Fig. A27. Potential Economic Loss Resulting from Release of Cm^{242}. Typical population distribution.

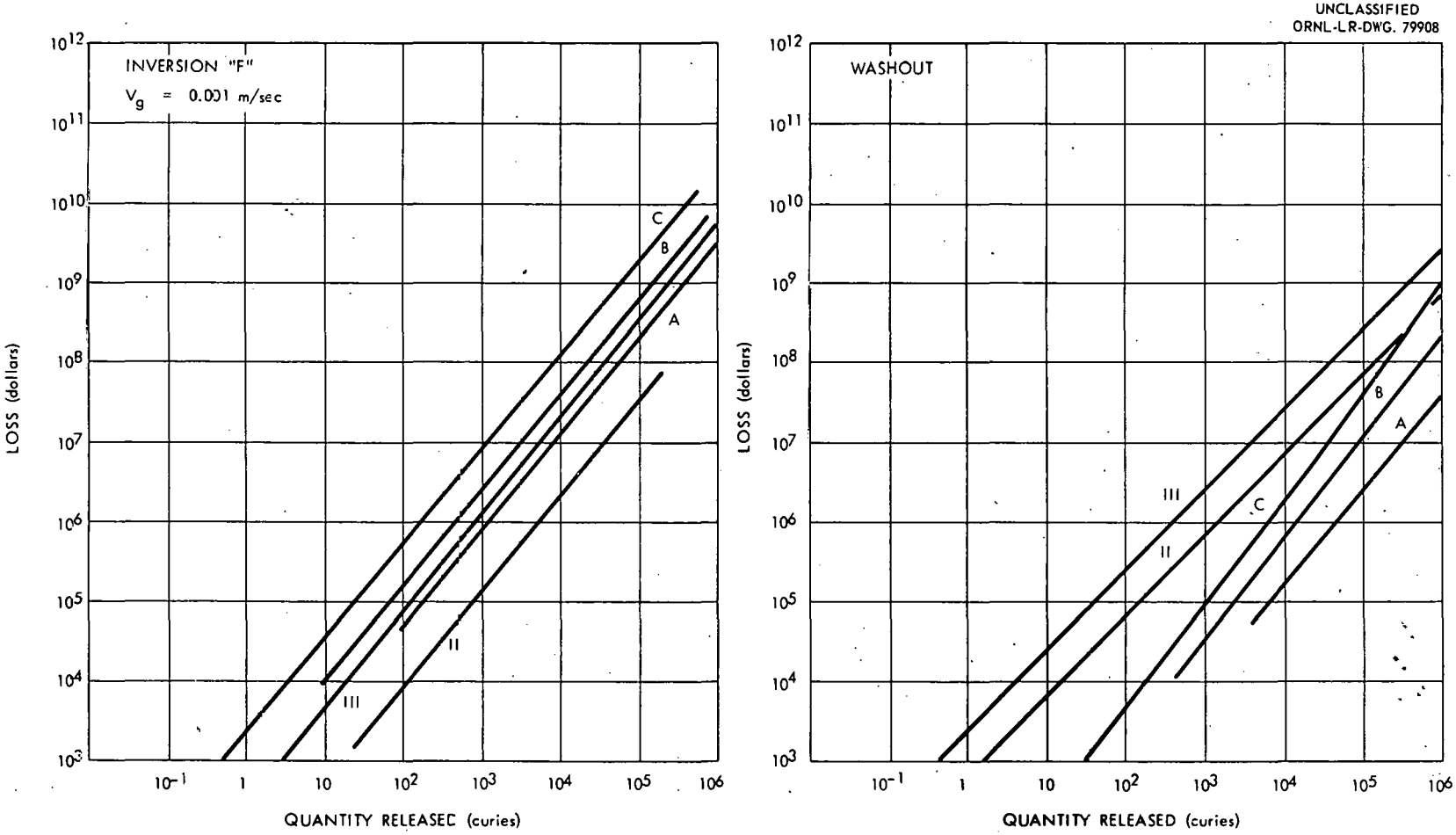

Fig. A28. Potential Economic Loss Resulting from Release of Cm^{242}. One hundred persons per square mile.

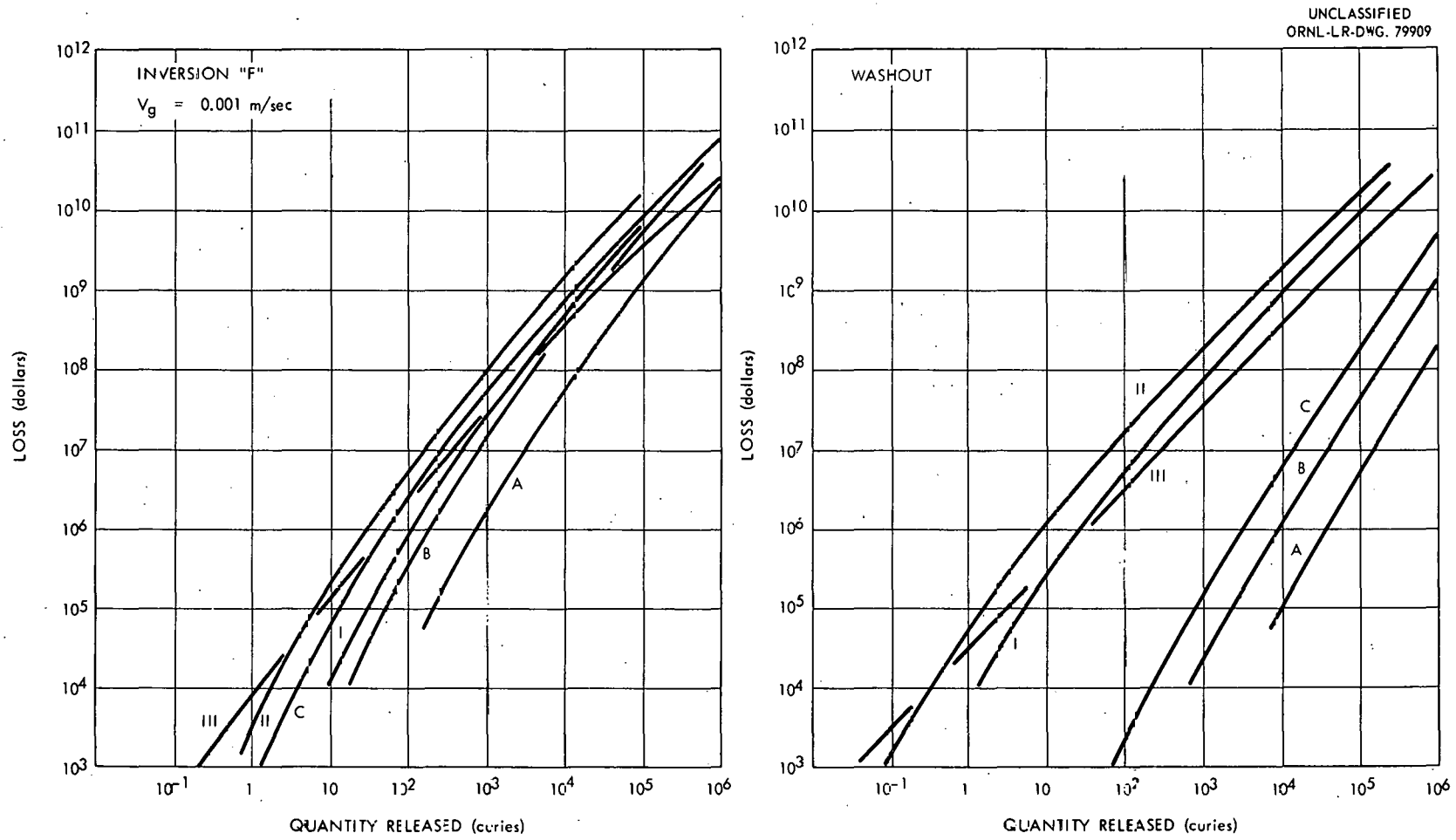

Fig. A29. Potential Economic Loss Resulting from Release of Cm^{244}. Typical population distribution.

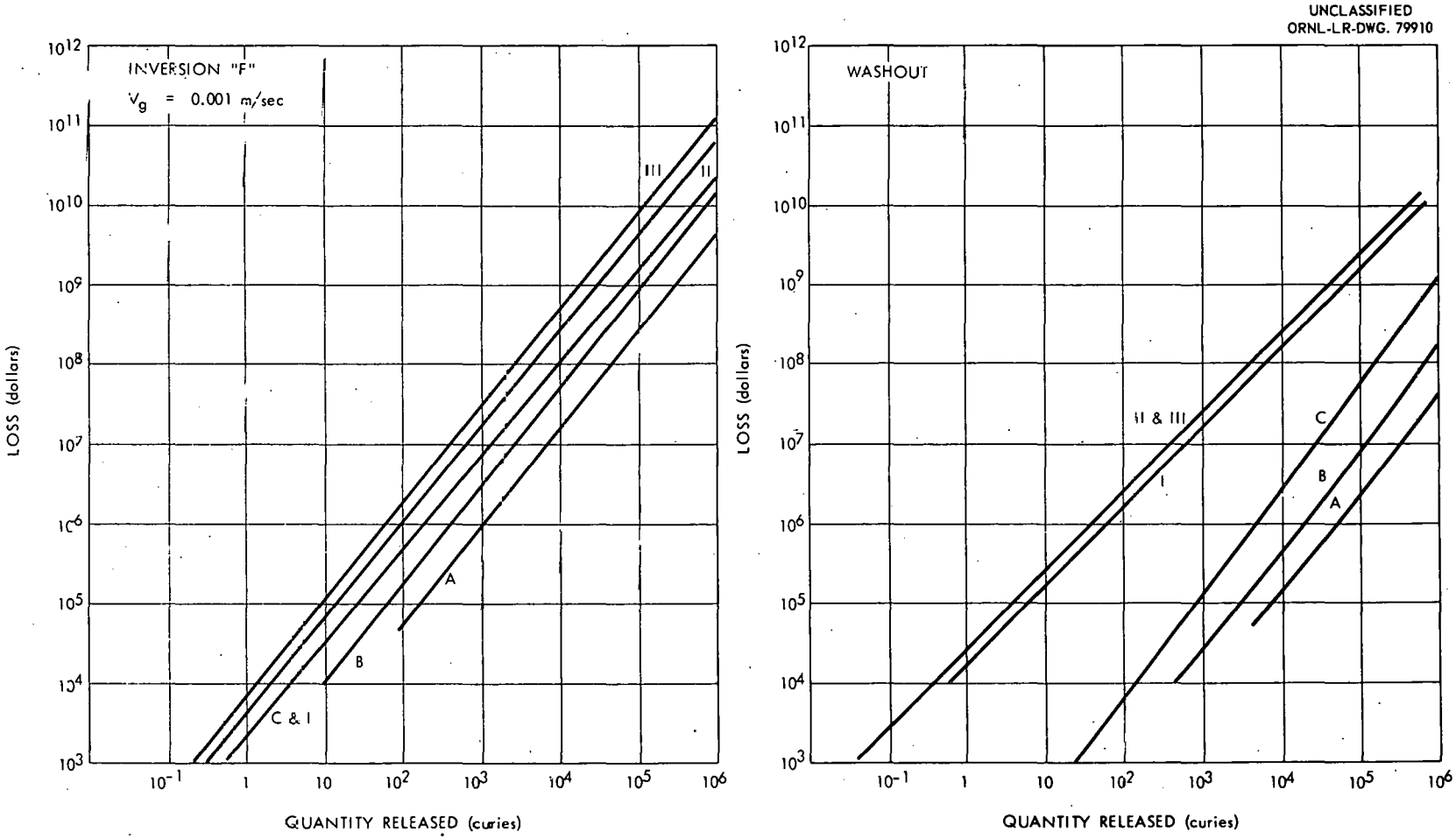

Fig. A30. Potential Economic Loss Resulting from Release of Cm^{244}. One hundred persons per square mile.