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ENVIRONMENTAL SCIENCES DIVISION

THE POTENTIAL USE OF CHERNOBYL FALLOUT DATA TO TEST AND EVALUATE THE PREDICTIONS OF ENVIRONMENTAL RADIOLOGICAL ASSESSMENT MODELS

Report to the U.S. Department of Energy
Office of Health and Environmental Research
from the
Interlaboratory Task Group on
Health and Environmental Aspects of the
Soviet Nuclear Accident

Prepared by the Committee on Model Validation

Environmental Sciences Division
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FOREWORD

A meeting of the Department of Energy's (DOE) Office of Health and Environmental Research (OHER) Biomedical and Environmental Program directors was held in Washington, D.C., on May 5, 1986, shortly after the April 26 accident at the Soviet Union's Chernobyl Nuclear Power Station. At that time there was relatively little information concerning the accident and a great deal of confusion both in the United States and throughout Europe and the rest of the world. One outcome of that meeting was an OHER-appointed task group on the Health and Environmental Consequences of the Soviet Nuclear Accident. The task group comprises staff members of the laboratories and universities conducting research programs for OHER. Dr. W. J. Bair, Battelle Northwest Laboratory, was selected to lead the task group.

Each of the four committees established within the task group was to direct its efforts to a specific task. These tasks were (1) to assess the resources at DOE/OHER laboratories that could assist the USSR as regards the Chernobyl accident, (2) to analyze data from the accident as a means of validating models used to predict the consequences of such accidents, (3) to provide information that will be useful to DOE and other organizations for emergency response (lessons learned), and (4) to analyze the health and environmental consequences of the accident at Chernobyl.

COMMITTEE MEMBERS

The findings of the Committee charged with the responsibility of using data from the Chernobyl accident to validate existing models are the subject of this report. The members of the Model Validation Committee are as follows:

C. R. Richmond, Chairman, Associate Director, Oak Ridge National Laboratory (ORNL)

F. O. Hoffman, Vice Chairman, Environmental Sciences Division, ORNL

B. G. Blaylock, Environmental Sciences Division, ORNL

K. F. Eckerman, Health and Safety Research Division, ORNL

P. A. Lesslie, Computing and Telecommunications Division, Martin Marietta Energy Systems

C. W. Miller, Illinois Department of Nuclear Safety

Y. C. Ng, Lawrence Livermore National Laboratory

J. E. Till, Radiological Assessments Corporation

This report represents work that was completed by the model validation committee in June of 1987. Since this time additional data have become available that could modify its conclusions. The publication of these early results is issued at this time to document the efforts of the committee and to demonstrate the potential use of Chernobyl fallout data for model testing.

Since completion of this document for the U.S. Department of Energy, the individual contributors to this report have been encouraged to continue with their investigations and to present their results in the open literature. The reader should take note of one publication that has already been prepared and which constitutes a supplement to the analysis presented herein on the transport of ^{137}Cs in terrestrial food chains. This publication, "A Comparison of Model Predictions and Observations of the Transfer of ^{137}Cs through the Air-Pasture-Cow-Milk Pathway" by Y. C. Ng and F. O. Hoffman, UCRL-96664 (December 1987), will appear in the proceedings of a workshop sponsored by the Commission of the European Communities and the U.S. Department of Energy on "Methods of Assessing the Reliability of Environmental Transfer Model Predictions", October 5-9, 1987, Athens, Greece.

OBJECTIVES AND SCOPE

The objectives of the Model Validation Committee were to collaborate with U.S. and foreign scientists to

- collect, manage, and evaluate data for identifying critical research issues and data needs to support an integrated assessment of the Chernobyl nuclear accident;

- test environmental transport, human dosimetric, and health effects models against measured data to determine their efficacy in guiding decisions on protective actions and in estimating exposures to populations and individuals following a nuclear accident; and

- apply Chernobyl data to quantifications of key processes governing the environmental transport, fate, and effects of radionuclides and other trace substances.

The scope of the Model Validation Committee was to

- establish working relationships with key foreign and U.S. scientists;

- collect, manage, and evaluate data obtained from reports and computer data banks;

- develop test questions based on the evaluation of available data for environmental transport, human dosimetric, and health effects models;

- challenge and test model predictions through their comparison with measured data;

- recommend model improvements that would support integrated environmental and health effects assessment models; and

- recommend further research issues.

As is often the case, the objectives and scope of the committee's effort changed as the task group's activities evolved. Our committee's major activity was restricted to a report on model validation. Some of

the data supplied by foreign colleagues were not used in this report; these data are, however, available as part of the Chernobyl data base we have assembled at ORNL.

The committee gratefully acknowledges the cooperation of many individuals associated with institutes and agencies throughout the world. We hope that this report, in turn, will be helpful to them. Special recognition must be given to F. Owen Hoffman, a pioneer in model validation activities, who assumed responsibility for preparing this report.

C. R. Richmond, Chairman
Model Validation Committee

PREFACE

On April 26, 1986, at 1:23 a.m., local time, an explosion occurred in Unit 4 of the Chernobyl Nuclear Power Station in the USSR. This event initiated extensive monitoring activities throughout the world, generating concerns, apprehensions, and discussions on the part of many governments. The communications media were left to speculate about the circumstances surrounding and possible implications of the accident because of the lack of information available from the USSR. Scientists throughout the world responded rapidly and admirably to the situation by collecting data and analyzing the accident, based on information brought to them via atmospheric fallout from Chernobyl. In fact, many of these activities preceded formal actions taken by scientific organizations.

Two years have passed since the accident at Chernobyl. Much information has now been accrued by many nations, part of which came out of an important meeting held at the International Atomic Energy Agency (IAEA) in August 1986, about four months after the accident. The Soviets presented a great deal of information at that meeting, to complement information already obtained by other governments. Much information is still needed, however, to help scientists throughout the world arrive at a better understanding of the accident and its subsequent impacts on man and the environment. Additional information will be extremely useful to all of us interested in validating the numerous models used in attempting to determine the consequences of a nuclear reactor accident. It is important that we learn as much as we can from this accident so that we can improve the accuracy and predictive capabilities of models used to assess impacts and to predict events. We must also realize that such information will be useful in other accident situations involving the widespread distribution of materials to man's environment, especially in those cases where the released materials cross international boundaries.

These model validation activities have been extremely useful in that we have been reminded to be more cognizant of information obtained in the past. For example, it has been known for many years that ^{137}Cs from nuclear weapons fallout tends to concentrate to a relatively high level in mushrooms within forested systems. Yet, many people were surprised to find high levels of ^{137}Cs in forest mushrooms as a result of the Chernobyl accident. In fact, had we in the scientific community been better prepared, mushrooms could have been used as a bioindicator of the deposition of ^{137}Cs from Chernobyl into foodstuffs. We might also have made better use of previously obtained information concerning the uptake of ^{131}I and its subsequent appearance in cheese and milk in goats as compared with cows. This kind of "early indicator" information might have provided useful input to decision makers faced with the problem of determining whether or not existing and predicted levels of radionuclides such as ^{131}I and ^{137}Cs were "acceptable."

A vast storehouse of metabolic and dosimetric information on ^{137}Cs exists which has been obtained worldwide from the testing of nuclear weapons in the atmosphere prior to the 1964 limited test ban treaty. The ^{137}Cs incorporated into the human body can be measured relatively easily using whole-body counting techniques. The minimum detectable activity for many whole-body counters is about 1 nCi for a 30-min counting period. Some of the more advanced whole-body counters can measure about 0.5 nCi within the body for a 15-min counting period. By repeatedly measuring activity levels in the same individual in advanced whole-body counters, it is possible to reach minimum detectable activities of several hundredths of a nanocurie. Thus, it will be important in the future to establish linkages among research establishments throughout the world that have the capability to detect small quantities of selected radionuclides in humans. There will almost certainly be interest in performing interlaboratory calibration studies for ^{137}Cs and other important radionuclides in anthropometric phantoms representing both adults and nonadults.

A great deal of information has been collected on the subject of predictive models during the decades following the introduction of

nuclear weapons testing in the atmosphere. These models also cover various scenarios, ranging from weapons accidents to failures at operating nuclear power stations. The models include the source term, atmospheric transport, terrestrial transport, aquatic transport, and, ultimately, the uptake of radionuclides by the general public, as well as an estimate of health effects in affected individuals and in select populations. Despite the large number of data available, this report shows that for two of the most studied radionuclides, ^{131}I and ^{137}Cs, the existing models overpredict these materials in cow's milk by amounts that should make us more concerned than complacent. It is also clear that for other radionuclides, the state of our knowledge is less precise and more needs to be learned.

Much remains to be done in the area of model validation. We believe international efforts such as those undertaken by the Biospheric Model Validation Study (BIOMOVS) are particularly important and should be strongly supported. Other activities, such as the IAEA's effort to develop a coordinated research program to calculate radiation dose per unit intake of radioactivity to the general public, should be fully supported.

C. R. Richmond, Chairman
F. O. Hoffman, Vice Chairman
B. G. Blaylock
K. F. Eckerman
P. A. Lesslie
C. W. Miller
Y. C. Ng
J. E. Till

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ACKNOWLEDGMENTS

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Special acknowledgment goes to Mr. David Norwood, U.S. Environmental Protection Agency (EPA), Montgomery, Alabama, who provided us with the EPA data base on Chernobyl measurements and who facilitated telecommunications to ORNL.

In addition, we gratefully acknowledge the numerous individuals who unselfishly provided us with unpublished data, in particular, Mr. Harry Koehler, Dr. Eric Wirth, and Prof. J. Schwibach, Federal Health Office, Neuherberg, Federal Republic of Germany (FRG); Dr. Arnulf Matting, Federal Ministry for Environment, Nature Protection, and Reactor Safety, FRG; Dr. Frances Fry, National Radiological Protection Board, United Kingdom; Dr. Lief Moberg and Mr. Gunnar Johansson, National Institute for Radiation Protection, Stockholm, Sweden; Ms. Ulla Bergström, Studsvik Energieteknik AB, Nyköping, Sweden; Dr. Illka Savolainen and Dr. Gordon Linsley, International Atomic Energy Agency, Vienna, Austria; Dr. X. Chen, Laboratory of Industrial Hygiene, Peoples Republic of China; Dr. O. Matsuaka, National Institute of Radiological Sciences, Japan; and Mr. I. Lauren Larsen and Mr. James S. Eldridge of ORNL.

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EXECUTIVE SUMMARY

From April 26 to May 6, 1986, approximately 2000 PBq (50 MCi) of radioactivity (excluding noble gases) were released to the atmosphere as a result of the accident at Chernobyl. Consequently, radioactivity was detected in environmental samples throughout the northern hemisphere. The immediate concern after the accident was with the monitoring of radioactivity to assess potential harm to human health. Later, it became evident that the extensive amount of data developed from monitoring Chernobyl fallout could provide an opportunity to test or "validate" the predictive capability of mathematical models used to assess the environmental and health consequences of radioactivity and other trace substances. This report examines the potential use of Chernobyl data in testing the predictions of mathematical models currently used to assess the environmental and health consequences of releases of radioactivity. Included also in this report are general conclusions and specific recommendations concerning (1) further model testing, (2) further use of the Environmental Protection Agency (EPA) data base, and (3) recommendations for accident assessment. The appendix to the report contains a bibliography of reports and other information on the Chernobyl accident that have been obtained by the Oak Ridge National Laboratory.

Although an enormous amount of data was collected as a result of the Chernobyl accident, the quality of the data currently available for model testing is less than ideal. In some cases, this committee was able to perform preliminary testing of model predictions. In other cases, only recommendations could be made about the potential uses of data that may become available in the future. The examples given in this report concerning the use of Chernobyl fallout data for model testing are preliminary and have been presented for illustrative purposes only. Additional data will be required to confirm the results of these analyses and to explain the discrepancies between model predictions and observations. Such data may become available through

international model testing programs such as BIOMOVS (International Biospheric Model Validation Study) and the recent cooperative research agreement between the U.S. Department of Energy, Office of Health and Environmental Research, and the Commission of the European Communities on the validation of terrestrial and aquatic food chain models.

The subjects of the models tested and the significant results or recommendations follow.

ANALYSIS OF SOURCE TERMS DERIVED FROM ENVIRONMENTAL DATA

Considering the limitations associated with available data and the large distances from the source at which environmental measurements were made, model predictions of the source terms appear to have been remarkably consistent. No current estimate can be considered a reference source term, including estimates provided by the USSR. It is apparent that scientists currently lack a clear and consistent rationale for what environmental measurements should be taken, how much measurement should be used to derive source terms, and which format should be used to report source term information.

TESTING ATMOSPHERIC TRANSPORT MODELS

It is difficult to test atmospheric transport model predictions as initially applied to the Chernobyl reactor because published results have already been calibrated with environmental monitoring data. Ideally, the application of these models should be tested based on predictions made before the arrival of Chernobyl fallout using accurate information for model input. Therefore, the only practicable test is a comparison of predicted air concentrations, ground depositions, and exposures near the Chernobyl reactor with data provided by the USSR. Unfortunately, there are obvious limitations associated with data received from the USSR thus far.

TESTING MODEL PREDICTIONS OF THE TRANSFER OF ^{131}I OVER THE AIR-PASTURE-COW-MILK PATHWAY

Radiological assessment models selected in this report overpredicted the transfer of ^{131}I from air to pasture vegetation and from pasture vegetation to cow's milk. The extent of this overprediction is approximately one to two orders of magnitude for several locations in Europe and the United States. The overpredictions are apparently the result of the combined effect of overestimating wet and dry deposition and the transfer from the cow's diet into milk. Contrary to generally accepted assumptions, the results for this accident imply that direct inhalation by humans of airborne ^{131}I may have been an important source of thyroid exposure for adult members of the population.

MODEL PREDICTIONS OF ^{137}Cs IN TERRESTRIAL FOOD CHAINS

The reference model selected for this analysis tended to overpredict the transfer of ^{137}Cs from air to pasture vegetation and from pasture vegetation to cow's milk. As with the prediction of ^{131}I transfer, the extent of the overprediction is approximately one to two orders of magnitude for several locations in Europe and the United States. Also, as with ^{131}I, the overpredictions produced by the model appear to be the result of a combination of processes, including overestimation of vegetation interception and retention of wet deposition and overestimates of the milk transfer coefficient for ^{137}Cs.

ANALYSIS OF ^{137}Cs IN AQUATIC ECOSYSTEMS

Analysis of data on ^{137}Cs in aquatic systems suggests that this radionuclide was transported more rapidly through runoff from the terrestrial system than is generally anticipated. Assessment models

consider the transfer of radionuclides from the terrestrial system to aquatic systems to be of minor importance with respect to potential radiological exposure. However, data in the United Kingdom, Sweden, and Finland indicate that fish in freshwater lakes were among the food products most highly contaminated with ^{137}Cs. Thus, the consumption of fish from lakes could define the critical population subgroup for radiation exposure assessment.

ESTIMATES OF HUMAN EXPOSURE

An illustrative analysis of current dosimetric models indicates reasonable agreement for ^{131}I, but further information is needed, particularly for ^{137}Cs, before any definitive conclusions can be made. *In vivo* data should be analyzed further to examine the range and variability of observed levels in humans resulting from the contamination event. Such an analysis could define the characteristics of critical groups in the population and thus contribute significantly to the interpretation of *in vivo* measurements from acute contamination of the environment.

DESCRIPTION AND ANALYSIS OF THE EPA DATA BASE ON CHERNOBYL FALLOUT IN THE UNITED STATES

This report presents the first published analysis of EPA monitoring data on Chernobyl fallout in the United States. Concentrations of ^{131}I in milk, rain, and air indicate that the highest exposures of individuals most probably occurred in the states of Idaho, Oregon, and Washington. Ratios of time-integrated concentration between milk and air for ^{131}I are comparable to those in Europe and are overpredicted by current radiological assessment models analyzed in the report. The maximum concentrations in milk were also well within those predicted by the ARAC system at Lawrence Livermore National Laboratory, and the upper limit estimates are not expected to be exceeded.

1. INTRODUCTION

From April 26 to May 6, 1986, approximately 2000 PBq (50 MCi) of radioactivity (excluding noble gases) were released to the atmosphere as the result of the accident at Chernobyl. Consequently, radioactivity was detected in environmental samples throughout the northern hemisphere. The immediate concern after the accident was with the monitoring of radioactivity to assess potential harm to human health. Later, it became evident that the extensive number of data developed from monitoring Chernobyl fallout could provide an opportunity to test or "validate" the predictive capability of mathematical models used to assess the environmental and health consequences of radioactivity and other trace substances.

Numerous national and international groups have been charged with the primary task of evaluating the radiological impact on human health. The focus of this report, however, is on the potential use of data that have become available on Chernobyl fallout measurements to test the predictions and parameter values assumed by present mathematical models applied in radiological assessments. In addition to model testing, the available data from Chernobyl fallout measurements are being used to critique currently accepted assumptions about the source term and the environmental behavior and fate of radionuclides discharged from a major reactor accident.

In some cases, this committee has obtained enough information to permit preliminary tests of model predictions. In other cases, only recommendations could be made about the potential use of data that may become available in the future. Most of the data received by this committee have been obtained from reports and informal communications issued by scientists from foreign countries and international organizations. In a very few cases, data were obtained from the open literature.

A major source of data on the extent of Chernobyl fallout in the United States was made possible through a computer link established between The Oak Ridge National Laboratory and the U.S. Environmental Protection Agency at Montgomery, Alabama. The majority of this

information, however, has been obtained through recent attendance at international symposia and workshops that have either been held specifically to discuss or have included sessions about the Chernobyl accident. Much of this information was received only during the latter part of 1986.

This report includes the possibilities for using Chernobyl data to test predictions of source terms, based on environmental measurements, and also evaluates estimates of reactor releases that were reported before publication of the Soviet report to the International Atomic Energy Agency. In addition, predictions of current terrestrial food chain models for ^{131}I and ^{137}Cs are analyzed. This analysis is followed by an evaluation of current assumptions about the importance and behavior of radionuclides in aquatic systems following an accidental release and a comparison of the environmental exposure pathways for ^{131}I and ^{137}Cs. The pattern of Chernobyl fallout in air, rain, and milk within the United States and the potential usefulness of these data for model validation are described. Finally, recommendations are made concerning future use of Chernobyl fallout data for testing and improving the state of the art in the assessment of accidental releases of radionuclides.

In the future, we expect the quality of information available on Chernobyl fallout to increase as individual investigators complete their evaluations of samples, initially reported to satisfy the emergency requirements of a postaccident situation. As a result of this inevitable process of review and reevaluation, errors associated with some of the data reported thus far will undoubtedly be revealed. Thus, the conclusions and recommendations in this report could change, depending on the extent and nature of future reports on measurements of Chernobyl fallout.

2. THE USE OF CHERNOBYL DATA FOR TESTING MODEL PREDICTIONS AND ASSUMPTIONS

2.1 A COMPARISON OF PREDICTED SOURCE TERMS WITH DATA PROVIDED BY THE SOVIET UNION

2.1.1 Introduction

In making decisions to minimize risk to exposed persons and in estimating the health impact of the accident, the source term must be well characterized. Errors in the assumed release will be transferred throughout the risk assessment in a linear fashion. Characteristics of the source term were perhaps even more important in this accident because the temperature of the release, the time profile of the release, and the presence of other constituents such as particles and steam affected the level to which material ascended in the atmosphere and even the physical and chemical form of many radionuclides. If the source term is poorly characterized, uncertainties of risk estimates may increase significantly. The objective of this part of the study is to compare estimates of the source term that were based on field measurements of radionuclides made during the Chernobyl accident to those reported by the Soviet Union in their official documentation of the accident (USSR 1986).

The lack of source term data posed significant problems to authorities outside the Soviet Union in attempting to make decisions to minimize risk to persons exposed to the radioactive fallout in the European sector and to predict the long-range implications of globally dispersed activity. Before and during the accident, little was known outside the Soviet Union regarding the operating history of the Unit 4 reactor, the events leading up to the accident, or the fraction of the radionuclide inventory being released. Therefore, scientists had to derive a source term using calculations based on measurements of radioactive substances in the environment, often at great distances from the accident site.

Consequently, source term estimates were made that included such information as the reactor's operating history, the type and quantity of radionuclides released, physical and chemical characteristics of the radionuclides, energy associated with the release, and temporal distribution of emissions. This evaluation of source term estimates uses the data reported by the USSR (USSR 1986) and the International Nuclear Safety Advisory Group (INSAG) analysis of these data (IAEA 1986) as a basis for comparison to source term estimates reported by other agencies. Source term data discussed in this report are limited to information published before the USSR released their formal report on the accident at the International Atomic Energy Agency (IAEA) experts' meeting in Vienna, Austria, on August 25-29, 1986.

It is emphasized that, even with the Soviet data and INSAG's analysis of these data, considerable uncertainty still remains in the accident source term. In addition, the data reported by both of these groups contain numerous inconsistencies and gaps. Therefore, it may be incorrect to assume that the information reported in these documents defines the "reference source term" for the purposes of validating source term estimates reported by other investigators.

The preliminary comparisons in this analysis are intended to serve merely as an example of how source term estimates and their method of derivation and reporting varied between different investigators. Indeed, it must be stressed that, before any comprehensive validation of source term estimates can be made, an intensive effort must be undertaken to accurately characterize the releases of radionuclides from the Chernobyl reactor.

2.1.2 Reference Source Term

According to the Soviet report, two independent calculations were made of the total activity released to the environment. The initial estimate was derived from aerial exposure measurements that were plotted on isodose curves and converted to deposited activity. From an analysis of these data, it was estimated that 300 to 520 PBq of activity were

deposited on the ground as of June 26 and that the total amount that had been discharged and deposited within the 30-km zone up to June 26 had been approximately 740 PBq. Extrapolation of this figure led the Soviets to conclude that the total activity released to the environment during the accident did not exceed 1900 PBq (excluding noble gases), which is less than 3 to 4% of the existing core inventory.

The Soviets made another estimate of the source term using aerial gamma spectrometry to determine radionuclide deposition. Although they did not specifically state the amount of activity deposited using this technique, they concluded that the data supported the 740-PBq value calculated using the isodose method.

The spectrum of radionuclides emitted from the reactor was determined using measurements of airborne radionuclides and samples of soil taken in the nearby region. The Soviet report concludes that the composition of the fission products released (with the exception of the volatile cesium, iodine, and tellurium) was similar to that in the core before the accident. Further, they concluded that small amounts of actinides were released and that the majority (>90%) of the transuranic activity deposited in the region around the reactor was from ^{242}Cm. Analysis of fallout samples showed the presence of "hot particles" primarily enriched in radionuclides of one type, such as cesium or cerium.

The Soviet report describes the release of radionuclides from the core as taking place in four major stages. The first stage, which occurred immediately after the accident began, was the mechanical release of materials from the core resulting primarily from the explosion in the reactor. The spectrum of radionuclides released was similar to that in the core but enriched in the volatile elements iodine, tellurium, and cesium. Approximately 25% of the total activity released from the core (excluding noble gases) escaped during this period.

In the second stage, which occurred after the initial release on April 26 and up to May 2, emission of radioactivity continued to

decline over a 5-d period, ultimately reaching a daily activity release rate that was about one-sixth of the value of the initial release. During this time, measures were being taken to stop the fire and contain the radionuclides. The spectrum of nuclides was similar to their composition in the fuel, and they were emitted as finely dispersed material being exhausted with the hot air and other reactor material escaping with the burning graphite.

The third stage occurred between May 2 and 5 and was characterized by a rapid increase in the discharge of radionuclides, which was initially dominated by the escape of volatile elements followed by a spectrum of radionuclides, again resembling that of the core inventory. No definitive explanation has been developed for this part of the source term; however, the Soviet report suggests it was created by a disintegration of uranium dioxide pellets caused by decay heating of the fuel to a temperature in excess of 2000°C. This gradual rise in temperature resulted in further release of fission products along with other reactor materials escaping with burning graphite. The total activity discharged on a daily basis gradually increased to a value that was about 70% that of the initial release.

The final stage of the source term, starting on May 6, was characterized by a rapid decrease in the escape of fission products and termination of the source term altogether.

Little information was given in either the Soviet report or the INSAG report regarding the physicochemical composition of the source term. It was noted that chemical forms of the aerosolized materials were quite variable and that the particle sizes of aerosols evolved from the core were in the range of less than one micron to tens of microns.

Both the Soviet report and the INSAG report lack clarity in defining the initial core inventory at the time of the accident and the percentage of each radionuclide estimated to have escaped containment and entered the environment. Table 2.1-1 summarizes the source term as taken from the Soviet and INSAG reports. The reported error for these

Table 2.1-1. Summary of Chernobyl reference source term

A	B	C	D	E	F	G	H	I	J
Radionuclide	Half-life (d)	Initial discharge on 26 April[a] (PBq)	Activity discharged by 6 May[a] (PBq)	Percent of core inventory discharged by 6 May[a] (%)	Initial core inventory corrected to 6 May[b] (PBq)	Initial core inventory corrected to time of the accident (PBq)	Activity ratio in core relative to ^{137}Cs corrected to 6 May	Activity ratio in core relative to ^{137}Cs at time of accident	Activity ratio relative to ^{137}Cs taken from WASH-1400[c]
^{85}Kr	3930	5.6	19	100	33	33	0.11	0.11	0.12
^{89}Sr	53	9.3	81	4	2000	2300	6.9	8.0	20
^{90}Sr	10,200	0.56	8.1	4	200	200	0.69	0.69	0.79
^{95}Zr	65.5	17	140	3.2	4400	5000	15	17	32
^{99}Mo	2.8	17	110	2.3	4800	72,000	17	250	34
^{103}Ru	39.5	22	120	2.9	4900	6000	17	21	23
^{106}Ru	368	7.4	59	2.9	2000	2000	6.9	7	5.3
^{131}I	8.05	170	270	20	1300	3300	4.5	11	18
^{132}Te	3.25	150	48	15	320	3300	1.1	11	26
^{133}Xe	5.27	190	1700	100	1700	7200	5.9	25	36
^{134}Cs	750	5.6	19	10	190	190	0.65	0.66	1.6
^{137}Cs	11,000	11	37	13	290	290	1.00	1.00	1
^{140}Ba	12.8	19	160	5.6	2900	5200	10	18	34
^{141}Ce	32.5	15	100	2.3	4400	5600	15	19	32
^{144}Ce	284	17	89	2.8	3200	3300	11	11	18
^{238}Pu	31,500	0.0037	0.030	3	1.0	1.0	0.0035	0.0035	0.012
^{239}Pu	8,900,000	0.0037	0.026	3	0.85	0.85	0.0029	0.0029	0.0045
^{239}Np	2.35	100	44[d]	3.2	140	3600	0.49	12	350
^{240}Pu	2,400,000	0.0074	0.037	3	1.2	1.2	0.0041	0.0041	0.0045
^{241}Pu	4800	0.74	5.2	3	170	170	0.59	0.59	0.72
^{242}Cm	164	0.11	0.78	3	26	27	0.090	0.09	0.11

[a] Taken from USSR (1986). Reported accuracy is ±50%.

[b] WASH-1400 (USNRC 1975).

[c] Taken from IAEA (1986).

[d] Note inconsistency in reported data with May 6 value being less than April 26 value.

data is ±50%. Column G in Table 2.1-1 is the core inventory corrected to the time of the accident using the inventory data listed in column F. Columns H and I show the ratio of radionuclides in the core relative to ^{137}Cs for May 6 and the time of accident initiation. Column J shows the ratio of radionuclides in the core relative to ^{137}Cs for a core inventory taken from WASH-1400 (USNRC 1975). Although the WASH-1400 core inventory used to derive the ratios in column J may not precisely reflect the inventory of the Chernobyl Unit 4 reactor, it does provide a reasonable check of the Soviet data term and gives some perspective regarding the abundance of these radionuclides relative to ^{137}Cs for a typical core. For example, a comparison of the ratios in columns I and J indicates good agreement (with the exception of ^{99}Mo and ^{239}Np). Molybdenum-99 appears to be too high while ^{239}Np appears to be too low in the Soviet data term. The rationale for these discrepancies is not clear and should be investigated.

2.1.3 Examples of Methods Applied by Scientists Outside the USSR to Estimate the Chernobyl Source Term Using Environmental Measurements

In the absence of Soviet information characterizing radionuclide emissions from the Chernobyl reactor during the accident, scientists outside the USSR used environmental measurements to assemble a picture of the accident as it was taking place. These estimates were often made using data that were collected at great distances from the source. The following discussion illustrates techniques that were applied to characterize the source term during the early stages of the accident.

2.1.3.1 Determining the Operating History of the Chernobyl Reactor

Reactor effective full power hours before the accident

The ratio of ^{137}Cs to ^{134}Cs in the environment can yield insight regarding the operation period of a reactor before an accident.

The shorter-lived ^{134}Cs ($T_{1/2}$ = 2.5 years) is an activation product [$^{133}Cs(n,\gamma) \rightarrow {}^{134}Cs$] and builds up in the core during power level operations. The longer-lived ^{137}Cs ($T_{1/2}$ = 30 years) is a fission product with a relatively high fission yield of 5.9%. For a typical reactor, the ratio of these isotopes varies as a function of the effective full power hours operation of the reactor as shown below (Kuhn et al. 1986):

Operation time (d)	$^{137}Cs/^{134}Cs$ ratio
1	12,000
10	220
30	37
100	7
360	1.83
720	0.8
1100	0.5

The operation period of the reactor can be approximated by fitting the ratio determined from measurements in the environment to radionuclide inventory data as a function of time. In the case of the Chernobyl reactor, the mean value of the $^{137}Cs/^{134}Cs$ was about 2, leading scientists to estimate that the reactor had been in operation approximately 400 d before the accident (Devell et al. 1986).

Although the $^{137}Cs/^{134}Cs$ ratio may provide approximate estimates of reactor operation time, the uncertainty of result using this method must be considered. For example, it is known that the startup date of the Chernobyl-4 reactor was December 1983 and that prior to the accident it had been in operation for about 875 d. A model with a 400-d operating period that results in a correct $^{137}Cs/^{134}Cs$ ratio obviously has a specific power that is a factor of 2 too high for Chernobyl-4. Such a model, if run for the correct operating period, would overestimate the unsaturated nuclides by about a factor of 2.

Time of shutdown before the accident

The ratio of $^{133}I/^{131}I$ in the environment also provides information about the history of operation of a reactor before an accident. The half-life of ^{133}I is 0.85 d while that for ^{131}I is 8 d. The steady-state ratio of $^{133}I/^{131}I$ in the core is about 2.14. This ratio decreases after shutdown because of the more rapid decay of the ^{133}I. Therefore, a plot of the $^{133}I/^{131}I$ ratio based on environmental measurements as a function of time can be extrapolated back to the steady-state ratio of 2.14 to give the approximate time of shutdown of the reactor.

Again, the uncertainty associated with this method must be considered. The load factor in the days prior to reactor shutdown may significantly affect estimates of the shutdown. The Chernobyl-4 reactor was not at steady-state prior to the accident since power had been reduced from 100% to 50% 22 h before the accident. Whatever the $^{133}I/^{131}I$ ratio at the time of power reduction, its value at the time of the accident would have been about 25% lower. Unless there were compensating factors, the time-of-accident estimate would have been off by several hours. Therefore, caution must be used in applying this method since not accounting for changes in power level could lead to significant errors in estimates of the time of shutdown.

Core temperatures based on physical characteristics of particulate aerosols and ratios of radionuclides

Based on an analysis of airborne particulate radionuclides, assumptions can be made regarding temperatures in the core during the accident. For example, the more volatile radionuclides such as iodine, cesium, and tellurium would be expected to be released at lower temperatures than nonvolatile, more refractory elements such as ruthenium, cerium, and neptunium. Therefore, the concentration of less-volatile radionuclides relative to cesium could indicate the temperature attained in the core. For example, a ratio of strontium

(b.p. = 1384°C) to cesium (b.p.= 690°C) in the environment equal to that existing in the core at the time of the accident would suggest that the core reached a high enough temperature to volatilize both the strontium and the cesium.

An electron microscopy analysis of the physical characteristics of particulates released to the atmosphere also yields insight regarding temperatures in the core during the accident. Spherical particles of ruthenium approximately 1 μm in diameter were found in Sweden. As a result, it was assumed that, because the melting point of ruthenium is 2500°C, at least part of the reactor core reached this temperature.

2.1.3.2 Determining the Amount of Radioactivity Released to the Environment

The most critical part of the source term is the total activity of radionuclides released to the environment during the accident. Quantities can be estimated using meteorological models coupled with isotopic ratios and measurements of radionuclides in air and deposited on the ground surface. Environmental concentrations can be extrapolated back to the source of the accident using atmospheric transport models to estimate a source term for the accident. Generally, the greater the distance from the source, the more uncertain are the source term estimates. In the case of the Chernobyl accident, errors in the estimate of source term at large distances were compounded due to the differential deposition of individual radionuclides downwind from the site. However, in addition to distance, other factors can influence the reliability of source term estimates using this approach. For example, release height can be strongly influenced by thermal and physical phenomena occurring during the accident, thus significantly affecting the atmospheric modeling of radionuclide transport. In the case of Chernobyl, the lack of data needed to determine the effective height of release of the radioactive cloud significantly affected the reliability of source term estimates.

2.1.4 Discussion of Selected Source Term Estimates

As examples of source terms available for validation, Table 2.1-2 summarizes data published by several agencies before release of the Soviet and IAEA reports (USSR 1986, IAEA 1986). Sections 2.1.4.1 through 2.1.4.4 discuss these source terms.

2.1.4.1 Laylavoix et al. (1986)

In the absence of quantitative data on the core inventory at the time of the accident and the fraction of the core released during the accident, the French (Laylavoix et al. 1986) initially assumed that the core inventory was similar to that of a 1000-MW(e) pressurized water reactor having a burnup of 11,000 MWd/t. This appeared to be a reasonable assumption based on the ratio of $^{137}Cs/^{134}Cs$ found in the environment compared with that of pressurized water reactor (PWR) fuel with this burnup. The balance of activity in the core allowing for a 2-d decay period is shown in Table 2.1-2. Based on deposition measurements immediately after the accident, the French proposed that 22 PBq of ^{131}I had been discharged on April 26 and dispersed toward Scandinavia and that 22 PBq of ^{131}I had been dispersed on April 27 toward Europe. An additional ^{131}I source term was dispersed toward Byelorussia; however, this was not estimated because of the absence of data. They further concluded in this early report that approximately 2 to 3% of the core inventory was released during the accident.

Cogne (1986) published a second report that revised earlier estimates of the source term based on additional analysis of environmental samples. Further measurements yielded a consistent $^{137}Cs/^{134}Cs$ ratio of 1.9, supporting earlier assumptions that the burnup of fuel had been between 9,000 and 12,000 MWd/t. Cogne used cesium measurements reported by other agencies along with trajectory models to estimate the fraction of cesium in the core that was released. They concluded that, during the first 2 d of the accident,

Table 2.1-2. Summary of selected source terms for the Chernobyl accident that were reported before August 1986

	Laylavoix (1986), Cogne (1986)			Devell et al. (1986)		
	Qualitative source term estimates			Qualitative source term estimates		
	—9,000- 12,000-MWd/t fuel burnup —2-3% core inventory released			—400-d operation time for reactor —Reactor shutdown at 19:00 on April 25 —Part of core reached 2500°C		
	Quantitative source term estimates			Quantitative source term estimates		
Radionuclide	Initial core inventory, 2-d cooling (PBq)	Activity released (PBq)	Inventory released (%)	Initial core inventory (PBq)	Activity released (PBq)	Inventory released (%)
^{85}Kr	11					
^{89}Sr	3200					
^{90}Sr	81					
^{95}Zr	4800					
^{99}Mo						
^{103}Ru	3600	61	1.5	1400		
^{106}Ru	480	31	1.5			
^{131}I	2300			1000		
^{132}Te	2500	23	1	1400		
^{133}Xe	5200					
^{134}Cs	59	37	20			
^{137}Cs	93	57	20	52		
^{140}Ba	4400					
^{141}Ce	4800	0.11	0.025	1800		
^{144}Ce	2200	0.78	0.025			
^{238}Pu	0.63	0.000030	0.03			
^{239}Pu	0.063	0.00026	0.03			
^{239}Np	21,000			19,000		
^{240}Pu	0.41	0.00037	0.03			
^{241}Pu	110	0.052	0.03			
^{242}Cm	3.0	0.0078	0.03			

Table 2.1-2. (continued)

Radionuclide	Dickerson and Sullivan (1986)			WASH-1400 (PWR-2)		
	Qualitative source term estimates			Qualitative source term estimates		
	-Initial release followed by subsequent releases -Significant meltdown possible -50% of volatiles escaped			-Initial release over first 30 min, then remainder of release at relatively low rate		
	Quantitative source term estimates			Quantitative source term estimates		
	Initial core inventory (PBq)	Activity released (PBq)	Inventory released (%)	Initial core inventory (PBq)	Activity released (PBq)	Inventory released (%)
^{85}Kr		[a]		21	19	90
^{89}Sr				3500	210	6
^{90}Sr		0.001-0.07		140	8.1	6
^{95}Zr		0.037-2.6		5600	22	0.4
^{99}Mo				5900	120	2
^{103}Ru	4100	81	2			
^{106}Ru				930	19	2
^{131}I	3000	1500[b]	50	3100	2200	70
^{132}Te				4400	1300	30
^{133}Xe		[a]		6300	4400	90
^{134}Cs				280	140	50
^{137}Cs	220	110[c]	50	170	89	50
^{140}Ba				5900	360	6
^{141}Ce				5600	22	0.4
^{144}Ce				3100	13	0.4
^{238}Pu				2.1		
^{239}Pu				0.78		
^{239}Np				61,000	240	0.4
^{240}Pu				0.78		
^{241}Pu				130		
^{242}Cm				19		

[a] 3700- to 7400-PBq total activity released on 50% of total inventory.

[b] 1200 PBq first day, then 59 PBq/d for days 2 through 6.

[c] 89 PBq first day, then 4.5 PBq/d for days 2 through 6.

approximately 20% of the cesium escaped with an uncertainty factor of 4 (i.e., 5 to 80%). For iodine, they concluded that a slightly lower fraction had escaped. This lower iodine fraction was the result of either greater retention of this element in the installation or greater depletion of iodine during environmental transport. Release fractions for other elements were also reported as follows:

Tellurium	7%
Ruthenium	1-2%
Lanthanides	0.01-0.04%
Actinides	0.02-0.04%

2.1.4.2 Devell et al. (1986)

Very soon after the accident began, Devell et al. (1986) predicted characteristics of the source term. A hypothetical core inventory for a 3000-MW(t) reactor was derived using several different environmental measurements. Based on the ratio of $^{137}Cs/^{134}Cs$ in deposited fallout of approximately 2, they concluded that the source of contamination was from a nuclear reactor rather than a nuclear weapons test and that the reactor had been in operation for 400 d.

Electron microscopy of hot spots on air particulate samples revealed particles that were about 1 μm in diameter. The uniform shape of some particles indicated that melting had occurred in part of the core and that in at least part of the core temperatures of approximately 2500°C were achieved. This conclusion was further explained by a higher-than-expected proportion of volatile elements in the fallout, suggesting both high temperatures and the failure of containment.

Based on the ratio of $^{133}I/^{131}I$, the time of reactor shutdown was estimated to be about 19:00 on April 25. This assessment was made by extrapolating the $^{133}I/^{131}I$ ratio back to its steady-state value of 2.14, as described earlier.

2.1.4.3 Dickerson and Sullivan (1986)

Dickerson and Sullivan (1986) estimated the extent and magnitude of airborne radionuclides released during the accident. Measurements of radionuclides were coupled with atmospheric dispersion model calculations to develop a scenario for source term.

Initial calculations were based on air concentrations and ground deposition measurements from Scandinavia for about 15 fission products. Of these, ^{131}I and ^{137}Cs were selected for the source term estimates because data on these radionuclides were the most readily available.

The source was divided into a lower cloud and an upper cloud. The lower-level cloud was assumed to be produced over a period of 6 d as a result of heat from the burning fire. The upper-level cloud was assumed to be produced by one or more of the following: explosions followed by a hot fire for several hours, convective activity associated with thunderstorms near the reactor site, or lift over a warm front located between Chernobyl and the Baltic Sea.

Of the released material, 80% was assumed to be in the upper-level cloud and 20% in the lower-level cloud. The inventory in the reactor at the time of the accident and the amount of activity released during the first 6 d of the accident are shown in Table 2.1-2. A total of 1500 PBq of ^{131}I and 110 PBq of ^{137}Cs were assumed to be released over the 6-d period. Initial reports in the Dickerson and Sullivan source term suggested that, because of the presence of ^{140}Ba and ^{95}Zr in Sweden, a significant meltdown could have occurred and that a majority of the volatiles had escaped the core during the first 24 h. Additional measurement data subsequent to the initial reports suggested that approximately 50% of the volatiles had escaped.

2.1.4.4 USNRC (1986)

The U.S. Nuclear Regulatory Commission (NRC) (USNRC 1986) developed an approximate source term for the accident by comparing

environmental measurements with computer-generated plots of dose versus distance for a number of WASH-1400 (USNRC 1975) release categories and for a variety of meteorological conditions. Because few measurements were reported in the vicinity of the accident where the curves could be applied directly, the curves were extrapolated to longer distances for which dose measurement data were available. Comparison of these data suggested that a release similar to the PWR-1, PWR-2, or PWR-3 categories could have been involved. For this analysis, the PWR-2 scenario was selected for comparison. The following paragraphs contain a description of the PWR-2 accident taken from WASH-1400.

This category is associated with failure of core-cooling systems and melting concurrent with the failure of containment spray and heat removal systems. Failure of the containment barrier would occur through overpressure, releasing a substantial fraction of the containment atmosphere in a puff over a period of about 30 min. Because of the sweeping action of the gases generated during containment vessel melt-through, the release of radioactive material would continue at a relatively low rate thereafter. The total release would contain approximately 70% of the radioiodines and 50% of the alkali metals present in the core at the time of the release. As in the PWR-1 release category, the high temperature and pressure within containment at the time of containment failure would result in a relatively high release rate of sensible energy from containment.

The NRC (USNRC 1986) found reasonably good agreement between estimates of quantities of radionuclides released based on environmental measurements and those predicted using WASH-1400 for the PWR-1 and PWR-2. Reasonable agreement was also noted for the PWR-3 case, considering the uncertainties associated with this method of source term estimation.

In its source term estimate, the NRC cautioned that release scenarios and time dependence of radionuclide release magnitudes for Chernobyl are not necessarily expected to follow patterns similar to the light-water reactor (LWR) release categories of WASH-1400 because of design differences. When the NRC source term for Chernobyl was

developed (as of May 7, 1986), the absence of actinides other than ^{237}Np in environmental measurements was noted. This suggested that a melting of the core may not have occurred as described in the PWR-2 scenario.

2.1.5 Analysis of Source Term Estimates Compared with the Soviet Data

Scientists have estimated that the majority of the initial release and probably major fractions of subsequent releases appear to have been injected at heights in the range 300 to 1500 m (USNRC 1986). Further, some initial fraction of the release must have injected relatively high into the atmosphere for it to have been been transported at levels of 6000 to 9000 m and to reach the west coast of the United States by May 5, 1986. In view of the Soviet description of the accident, these assumptions were probably correct.

It is important to note that, according to the Soviet data, ratios of released activity are substantially different from those existing in the core inventory. This suggests that, in some areas of the core, temperatures may have been high enough to release nonvolatile elements but that in other parts of the core these elements were contained. This nonuniformity in core temperature during the accident leads to the conclusion that the derivation of source terms based on isotopic analysis of environmental samples was misleading.

The absence of ^{90}Sr in environmental measurements outside the Soviet Union was particularly noteworthy. According to the Soviet report, the ratio of $^{90}Sr/^{137}Cs$ in the core was approximately 0.71 and the ratio in the released activity was about 0.22. However, the reported ratio of $^{90}Sr/^{137}Cs$ in environmental samples of approximately 0.01 leads to the conclusion that either the core did not reach a high enough temperature to release strontium or, once released, the strontium preferentially deposited closer to the reactor.

Regarding specific amounts of radionuclides released during the accident, it is again emphasized that the Soviet source term used as the basis for comparison lacks clarity in its description and remains

highly uncertain. It is possible that the source terms considered in this study may in fact represent more accurate estimates of radionuclides released than those reported in the Soviet report. Further, it must be noted that since the Soviet data are apparently based on measurements of radionuclides deposited within the USSR, their accounting of the percentage of total inventory of radionuclides released may underestimate what actually occurred. A more in-depth analysis of the Soviet data is needed to determine the significance of this problem.

Therefore, conclusions drawn when comparing the Soviet data with that of other investigators must be judged with extreme caution since each of the source term estimates in this study may indeed not represent the same entities. This lack of uniformity between methods of derivation of source terms and the manner in which the data were reported seriously affected our ability to compare results. With this disparity in mind, several preliminary observations can be made about quantities of individual radionuclides released to the environmental during the accident as reported by scientists outside the USSR.

Considering the lack of data with which to work, source terms reported by Laylavoix (1986) and Cogne (1986) for ^{103}Ru, ^{106}Ru, ^{132}Te, ^{134}Cs, and ^{137}Cs were within a factor of about 2 of the releases reported by the Soviets through May 6. Reported amounts of releases of actinides were underestimated by about two orders of magnitude. Dickerson and Sullivan (1986) overestimated the amounts of ^{131}I and ^{137}Cs released by factors of about 5 and 3, respectively. They also correctly determined an initial release followed by subsequent releases over a prolonged period of time.

The ratio of $^{137}Cs/^{134}Cs$ ranged from about 1.7 to 2.4. The ratio of released activity given in the Soviet report was 2.0, while the ratio for core activity was 1.54. It is not readily apparent why this disparity between $^{137}Cs/^{134}Cs$ ratios in the environment and in the core exists. According to the Soviet report, the reactor had been in operation since December 1983, although the number of days at power

since initial startup was not clear. The assumption by Devell et al. (1986) that the core had been in operation for approximately 400 d before the accident based on the $^{137}Cs/^{134}Cs$ ratio was apparently an underestimate. Nevertheless, the use of $^{137}Cs/^{134}Cs$ ratios in environmental samples to determine the period of operation of the reactor before the accident is an excellent method to apply in the absence of other data.

The PWR-2 data were selected for comparison in Table 2.1-2 because they appear to reasonably describe the Soviet source term on a nuclide-specific basis. Note, however, the disparity between reported values for ^{131}I and ^{132}Te, which are significantly higher in WASH-1400. Assuming that the Soviet source term is correct, this disparity points out that predictions of these nuclides in reactor accident source terms remain highly uncertain. Nevertheless, the closeness of the WASH-1400 source term to the Soviet source term is noteworthy, especially in view of its 1975 publication date.

2.1.6 Conclusions

This section of the report discussed methods used to derive source terms for the Chernobyl nuclear accident and the feasibility of validating source term estimates using the Soviet data and the IAEA analysis of the Soviet data as reference points. Several conclusions are summarized in the following paragraphs.

Validation of source terms estimated by scientists outside the USSR is complicated by weaknesses and inconsistencies in the Soviet data used as the "reference source term" for this analysis. Examples are the unusual amounts of ^{99}Mo and ^{239}Np reported in the source term. In addition, the Soviet report and the IAEA analysis of the Soviet report are not entirely clear in their description of events leading up to the accident or the event as it was taking place. Bearing this in mind, several statements can be made regarding the validation of source terms reported by agencies outside the USSR before details of the accident were released.

Source terms that were reported in the literature lacked consistency both in methods used for their derivation and in specific data published, thus making direct comparisons difficult. If these reports had been more uniform, a more accurate description of the event could probably have been formulated while the accident was taking place, thus providing a sounder basis on which to make decisions for estimating public health impact.

Considering the poor quality of data with which scientists outside the Soviet Union had to work, estimates of releases from Chernobyl were remarkably good, not only in predicting the quantities and temporal distribution of radionuclides released, but also in characterizing the operating history of the Chernobyl reactor. The French (Laylavoix et al. 1986, Cogne 1986) provided a comprehensive timely description of the source term. Devell et al. (1986) provided a reasonably accurate description of the reactor's operating history and predicted at least a partial melting of the core. Dickerson and Sullivan (1986) accurately determined an initial release followed by subsequent releases over a period of time. It is most interesting to note that WASH-1400 (USNRC 1975) provided a very good estimate of the source term, especially in view of its 1975 publication date.

The most important conclusion of this analysis, however, is that methodologies must be improved to derive source term estimates from environmental measurements. A key lesson learned from Chernobyl is that, regardless of where an accident may happen, data characterizing emissions from the nuclear reactor may not be available because of inaccessibility of the area or a lack of information communicated by authorities at the scene. Transformation of environmental measurements into source terms may be the only mechanism available to quantify releases. Based on this review of source term estimates for Chernobyl from an international spectrum of agencies, it is apparent that we currently lack a clear and consistent rationale for what environmental measurements should be taken, how environmental measurements should be used to derive source terms, and which format should be used in reporting source term information.

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2.2 COMPARISON OF OBSERVED AND PREDICTED DOSES NEAR THE CHERNOBYL PLANT

This section briefly reviews efforts that were made to estimate doses to persons living near the Chernobyl plant and compares these estimates with the doses reported by the authorities of the Soviet Union for persons living in the town of Pripyat'.

2.2.1 Dose Estimates from Specific Models

2.2.1.1 ARAC Studies

On Monday, April 28, 1986, the Department of Energy (DOE) requested that the Atmospheric Release Advisory Capability (ARAC) (Dickerson et al. 1985) located at Lawrence Livermore National Laboratory begin dose calculations related to the Chernobyl accident (Dickerson and Sullivan 1986, Gudiksen 1986, Gudiksen and Lange 1986). ARAC used the PATRIC model (Lange 1978a) to estimate inhalation and immersion doses, primarily from ^{131}I and ^{137}Cs, throughout Europe and North America. A modified version of the three-dimensional MATHEW/ADPIC code system (Sherman 1978, Lange 1978b) was used to estimate doses near the Chernobyl plant. In their calculations, the ARAC models assumed that no mitigation efforts were used to reduce the doses to persons. Also, during the course of the many calculations made by ARAC, the model results were integrated with all available measurements from outside the Soviet Union in an attempt to better define the source term used and thus improve results of the modeling efforts.

ARAC estimated doses of up to 30,000 μSv within 10 km of the plant, 3000 μSv in the range 10 to 100 km, and 300 μSv in the range 100 to 200 km or more from the plant (Gudiksen and Lange 1986). These doses are the committed effective dose equivalent to an adult resulting from inhalation of contaminated air.

2.2.1.2 MESOS Model

The MESOS model, which was developed specifically to study potential transfrontier consequences of nuclear accidents, was used to estimate the dispersion of Chernobyl radionuclides across Europe (ApSimon and Wilson 1986). The source term initially input into this puff trajectory model was based on source terms used in the Reactor Safety Study (USNRC 1975). The radionuclides ^{133}Xe, ^{131}I, and ^{137}Cs have been studied. The maximum whole-body dose from a 24-h exposure to the passing plume of ^{133}Xe was estimated to be something greater than 24 μSv. Doses were not presented for the other radionuclides considered in this study.

2.2.1.3 MLAM Model

Staff from Pacific Northwest Laboratory used a different puff trajectory model, MLAM, to calculate trajectory paths for hourly releases from Chernobyl during the first 3 d after the accident (Davis et al. 1986). It was assumed that the radionuclides were released in the form of a vertical line source in nine layers above the surface of the earth ranging in height from 300 to 5500 m. These trajectories were combined with an estimate of the source term and appropriate dose models to estimate inhalation doses from ^{131}I and external whole-body doses from exposure to both the passing plume and the contaminated ground.

2.2.1.4 NRC Study

Staff of the NRC estimated the severity of the Chernobyl accident on the basis of radioactivity measurements made outside the Soviet Union (USNRC 1986). The NRC staff compared doses measured in Sweden, Finland, and elsewhere with doses calculated for various accident scenarios in the Reactor Safety Study (USNRC 1975) to obtain an estimate of the Chernobyl source term. They concluded that this accident was best described by the PWR-1 or PWR-2 class of accident.

The NRC staff estimated doses near the Chernobyl reactor by extrapolating from the Finnish and Swedish measurements. They assumed that atmospheric dispersion between the release and observation points was inversely proportional to the distance between these points raised to the 1.5 power. They further assumed that deposition and radioactive decay processes acted to remove material from the plume during transport. As a result of these assumptions, the NRC staff estimated that within 1 mile of the Chernobyl plant a 1-d exposure could result in a dose of more than 1 Sv to the whole body resulting from immersion in noble gases and a dose of more than 40 Sv to the thyroid resulting from the inhalation of ^{131}I (Table 2.2-1).

2.2.2 Vienna Report by the Soviet Union

It seems clear from their report in Vienna that, during the initial days of the Chernobyl accident, Soviet authorities were more concerned with halting the accident and mitigating its effects than in providing a detailed analysis of exposures and doses to persons living around the plant. In their Vienna report (USSR 1986), Soviet authorities provided detailed descriptions of the medical problems of workers initially exposed during the accident, as well as exposure readings at selected points of interest around the plant that were used in making evacuation decisions. More-detailed environmental measurements evidently were not taken until several days after the accident began, and these measurements were used more to help in estimating the source term than in estimating doses.

Within 10 d of initiation of the accident, 135,000 people living within 30 km of the Chernobyl plant were evacuated. Of this number, 45,000 were evacuated from the town of Pripyat' on the afternoon of April 27, the second day of the accident. Soviet authorities estimate that, as a result of their mitigation efforts, few persons initially living within this 30-km zone received a dose in excess of .25 Sv from external sources (i.e., the passing plume and the contaminated ground). The dose to the thyroid glands of children living in population centers within the 30-km zone was estimated to range between

Table 2.2-1. Comparison of reported potential doses to persons living in Pripyat' for 1-d exposures following the Chernobyl accident

Pathway	Radionuclide	Exposed organ	Amount	Model references
		USSR data		
External, cloud	NS[a]	Whole body	0.1–0.15 Gy	(USSR 1986)
External, ground				
Gamma	NS	Whole body	0.01–0.05 Gy	
Beta	NS	Skin	0.1–0.2 Gy	
Inhalation	^{131}I	Thyroid	0.015–0.25 Gy	
		Model predictions		
Inhalation	^{131}I	Adult thyroid	0.1–1 Sv[b]	ARAC
Inhalation	^{137}Cs	Whole body	0.001 Sv	(Gudiksen 1986)
External, cloud	^{133}Xe	Whole body	>24 μSv[c]	MESOS (ApSimon and Wilson 1986)
Inhalation	^{131}I	Adult thyroid	11 Sv	MLAM (Davis et al. 1986)
Inhalation	NS	Whole body	0.10 Sv	NRC
External, cloud	NS	Whole body	0.10 Sv	(USNRC 1986)
External, ground	NS	Whole body	0.45 Sv	
External, cloud	Noble gases	Whole body	>1 Sv[d]	
Inhalation	^{131}I	Thyroid	>40 Sv[d]	

[a]NS = radionuclide not specified.

[b]Two contours closest to Chernobyl.

[c]Value given for maximum grid square.

[d]Estimated for locations within 1.8 km of the Chernobyl plant; Pripyat' is located approximately 3 km from the plant.

0.25 Sv and 2.5 Sv because of the inhalation of radioiodine. Overall, however, radioiodine in milk is assumed to be the greatest source of internal exposure to these persons because non-Pripyat' evacuees consumed local food products, including milk, for 9 to 10 d following the beginning of the accident.

2.2.3 Comparisons for Pripyat'

Radiation exposures to persons in the town of Pripyat' were a major concern to Soviet authorities from the beginning of the accident. This town of approximately 45,000 people is located only 3 km west of the Chernobyl plant. During the initial releases from the accident, the plume is thought to have passed directly over this town. As soon as the severity of the accident became apparent to officials, the residents of Pripyat' were told to restrict their outdoor activities and remain indoors as much as possible. Evacuation of the town began at 2:00 on the afternoon of April 27.

Estimates of the dose received by residents of Pripyat' before their evacuation have been made on the basis of a number of different measurements. Because of the early concern of officials, multiple measurements of the exposure resulting from ground surface contamination were made in Pripyat' during the first days following the accident. Soviet authorities estimate that actual doses received from ground contamination were 2 to 5 times lower than expected from street exposure, including the effects of sheltering. Thyroid dose estimates were made on the basis of the radioiodine burden measured in 208 evacuees from Pripyat'. The primary source of radioiodine exposure to these persons was found to be inhalation. Doses resulting from exposure to the contaminated plume passing overhead were estimated on the basis of exposure measurements made in the plume itself.

Dose estimates made for the citizens of Pripyat' are the most detailed doses presented in the Soviet report (USSR 1986). As a result, an attempt has been made to compare doses predicted by various modeling efforts with those reported for Pripyat'. A direct comparison of such doses is difficult, however, because the models generally

present dose contours for a region of dose estimates for a relatively large grid square rather than dose estimates for particular location such as Pripyat'. Table 2.2-1 shows the doses reported by the Soviets for Pripyat' and the best 1-d doses that could be gleaned from the various dose estimates considered for that same location. The MLAM seems to overpredict the thyroid dose, but the overhead plume and ground contamination dose estimates are in reasonably good agreement with those reported by the Soviets. The ARAC thyroid doses are also in reasonably good agreement with the Soviet doses. The ARAC staff expected their near-in doses to be low, possibly by as much as an order of magnitude (Gudiksen 1986). However, the lack of consideration of mitigation efforts in the model may have helped compensate for other uncertainties in the calculation. Direct comparison of the Soviet doses with calculated doses does not appear possible because of the lack of resolution in the other estimates.

2.2.4 Conclusions

The use of Chernobyl data for validating predictions made by atmospheric transport models is limited because published results have been "tuned" to available environmental monitoring data obtained after the plume had reached locations of concern. Despite this severe limitation, model predictions can be compared with information published by the USSR during the summer of 1986. This section has provided only an initial examination of the models and techniques used to estimate near-in doses associated with the Chernobyl accident. For example, many of the modelers considered in this review probably have available more-detailed near-in dose estimates that might be more appropriately compared with the reported Pripyat' doses. Also, the plume trajectories calculated by the atmospheric transport models considered could be compared with trajectories estimated from a detailed synoptic analysis of the global weather patterns, which guided the transport of material in the atmosphere during the course of the accident. It must also be noted again that at least some of the models used to predict near-in doses were "tuned" during the course of the calculations on the basis of (1) monitoring data from outside the

Soviet Union, (2) increased information on the dynamics of the plume release, and (3) improved meteorological information. The comparisons considered in this section are not necessarily representative of the doses that were predicted when the first news of the Chernobyl accident was reported outside the Soviet Union.

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2.3 TESTING PREDICTIONS OF THE BEHAVIOR OF CHERNOBYL FALLOUT IN TERRESTRIAL SYSTEMS

2.3.1 The Use of Chernobyl Fallout To Test Model Predictions of the Transfer of Radioiodine over the Air-Pasture-Cow-Milk Pathway

Among the most important exposure pathways for evaluation of the radiological consequences of nuclear power reactors and atmospheric testing of nuclear weapons is the transfer of ^{131}I from the atmosphere to fresh milk. Airborne ^{131}I in the elemental form, for example, is expected to produce human thyroid doses via ingestion of contaminated cow's milk that are two to three orders of magnitude higher than would occur solely from direct inhalation when cows are grazing on contaminated pasture (Hoffman 1973a, 1973b; 1975).

The Chernobyl accident resulted in atmospheric dispersion of ^{131}I on a global scale and has thus provided an opportunity to collect data for testing model predictions of this exposure pathway for a variety of locations. The consequence of this large-scale release was that ^{131}I was detected in environmental samples throughout the northern hemisphere.

2.3.1.1 Data Acquisition and Evaluation

For this analysis, data have been selected from international symposia and individual reports received from Italy (Sandroni 1986), France (CEA 1986, Robeau and Wartenberg 1986), Belgium (CEN/SCK 1986), Germany (GRS 1986, Bonka 1986, Winkelmann 1986, GSF 1986, KFA 1986, Kühn et al. 1986, Händl and Pfau 1986), and the United States (Larsen et al. 1986, Bondietti and Brantley 1986, Eldridge 1986, EPA 1986, Schell et al. 1986). Data have thus been summarized for 11 different locations and have been analyzed for time-integrated concentrations of ^{131}I in above-ground air, pasture vegetation, and cow's milk, and for the total deposition per unit ground area. It is important to note that the data have been selected for conditions in which cows were grazing on pasture at the time of the arrival of Chernobyl fallout.

2.3.1.2 Time-Integrated Concentrations

Time-integrated concentrations derived from selected Chernobyl fallout measurements in Europe and the vicinity of Oak Ridge, Tennessee, are presented for ^{131}I in air, vegetation, and milk in Table 2.3-1 and Figs. 2.3-1, 2.3-2, and 2.3-3, respectively. The time-integrated concentrations estimated for Oak Ridge are substantially less than those estimated for European locations. The difference between Oak Ridge and European concentrations is about two to three orders of magnitude. An exception is the low time-integrated air concentration reported for Grenoble, France.*

The Grenoble air concentration is only ten times higher than the value for Oak Ridge. It is also an order of magnitude lower than other European locations reporting air concentrations representative of all physicochemical forms of ^{131}I. However, by comparison, the time-integrated concentration of ^{131}I in milk for Grenoble is similar to milk concentrations obtained for six other European locations. The Grenoble air concentration is therefore suspect.

2.3.1.3 Concentration Ratios

To facilitate a comparison of model predictions and Chernobyl data, time-integrated concentrations in milk and pasture were divided by the time-integrated concentration of ^{131}I in air. This normalization gives a concentration ratio that shows the time-integrated concentration in milk and pasture that would occur as the result of a given time-integrated concentration in air (with units of m^3/L and m^3/kg, respectively). Note that these ratios encompass all mechanisms of ^{131}I uptake by dairy cows, including inhalation of contaminated air and consumption of contaminated drinking water, as

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*Recent information received from N. Parmentier of the CEA, Paris, confirms that air concentrations reported for Grenoble should be increased by a factor of 4 to account for the total amount of ^{131}I in air.

Table 2.3-1. Time-integrated concentrations and concentration ratios (CR) obtained from Chernobyl fallout data on ^{131}I

Location	Time-integrated concentrations			Concentration ratios			References
	Air (Bq d/m^3)	Vegetation[a] [kBq d/kg (dry)]	Milk (kBq d/L)	$CR_{milk/air}$ (m^3/L)	$CR_{veg/air}$ [m^3/kg (dry)]	$CR_{milk/veg}$ [kg (dry)/L]	
Oak Ridge, United States	1.2E-01	2.8E-01	1.4E-02	1.2E+02	2.3E+03	5.1E-02	Larsen et al. 1986, Eldridge 1986
Grenoble, France	1.8E+00*	no value	9.0E-01	5.1E+02*	no value	no value	Robeau and Wartenberg (1986)
Ispra, Italy	1.6E+01	1.7E+02	1.6E+00	1.0E+02	1.1E+04	9.4E-03	Sandroni (1986)
Cadarache, France	2.1E+01	2.9E+01	2.0E+01[b]	9.6E+02[b]	1.4E+03	6.6E-01[b]	Robeau and Wartenberg (1986)
Geel, Belgium	2.2E+01[c]	3.0E+01	1.4E+00	6.2E+01	1.4E+03	4.5E-02	CEN/SCK (1986)
Mol, Belgium	2.2E+01[c]	2.1E+01	9.8E-01	4.5E+01	9.5E+02	4.6E-02	CEN/SCK (1986)
Aachen, Federal Republic of Germany (FRG)	3.0E+01	9.6E+01	2.2E+00	7.4E+01	3.3E+03	2.3E-02	Bonka (1986)
Jülich, FRG	3.0E+01	5.0E+01	1.4E+00	4.5E+01	1.7E+03	2.8E-02	KFA (1986)
Baden, FRG	3.5E+01	1.5E+02	7.6E-01	2.2E+01	4.2E+03	5.2E-03	GRS (1986)
Westphalia, FRG	4.1E+01	8.1E+01	5.9E-01	1.4E+01	2.0E+03	7.2E-03	GRS (1986)
Bavaria, FRG	7.6E+01	7.4E+02	4.1E+00	5.5E+01	9.8E+03	5.5E-03	GRS (1986)
Geometric mean	1.3E+01	4.5E+01	8.3E-01	6.3E+01[d]	2.7E+03	1.7E-02[d]	
Geometric standard deviation	6.8E+00	8.0E+00	4.6E+00	2.7E+00	2.3E+00	2.6E+00	

[a] Values reported in fresh weight have been converted to dry weight assuming 20% dry matter for pasture vegetation.

[b] Milk samples obtained from goats.

[c] The total amount of ^{131}I in air was estimated from reported particulate ^{131}I concentrations using the author's assumption that the particulate fraction was approximately one-third of the total.

[d] Excludes the values for goat's milk.

*Recent information received from N. Parmentier of the CEA, Paris, confirms that air concentrations reported for Grenoble should be increased by a factor of 4 to account for the total amount of ^{131}I in air. Thus the $CR_{milk/air}$ for Grenoble should be reduced by the same factor.

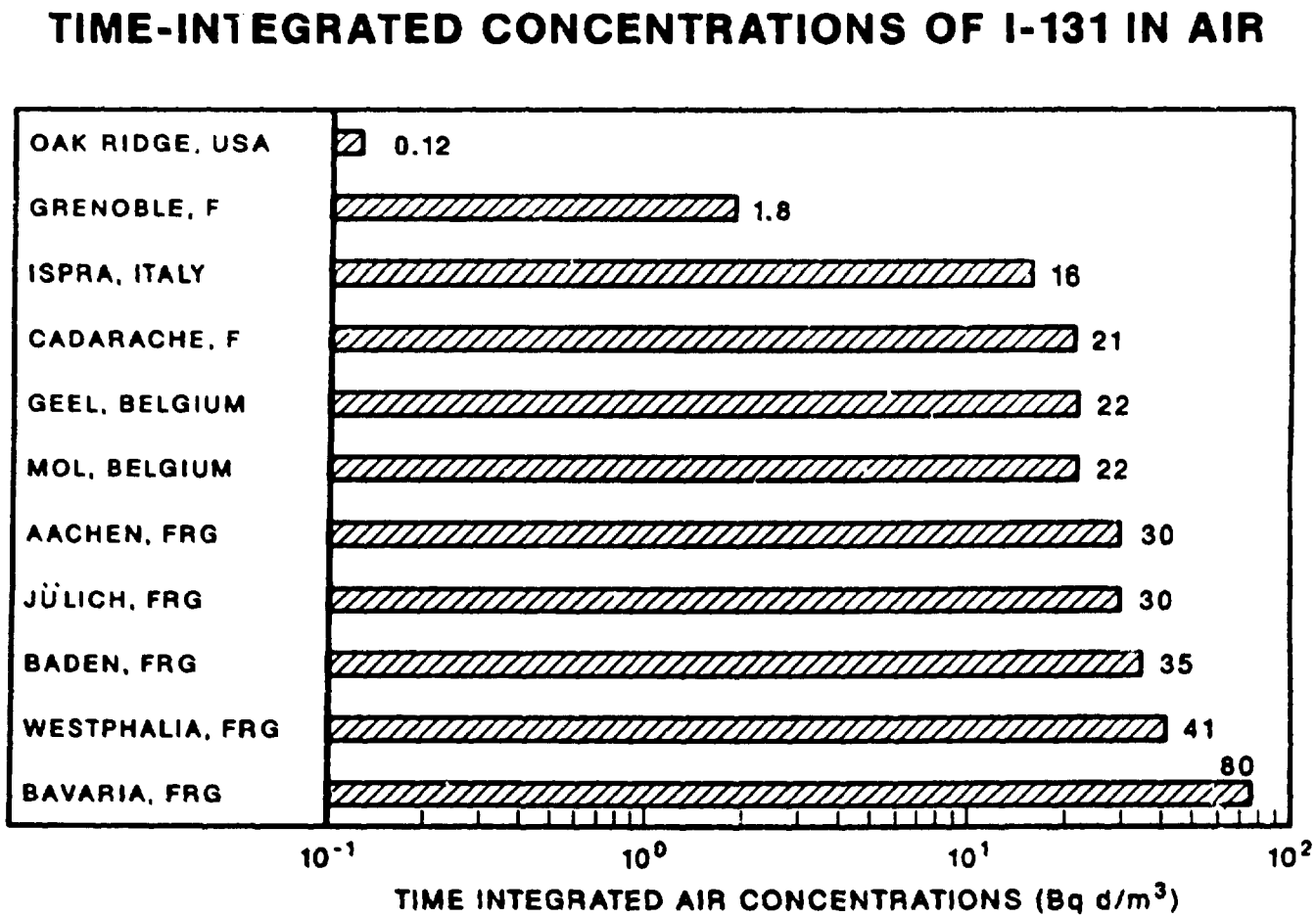

Fig. 2.3-1. Time-integrated concentrations of ^{131}I in air obtained from Chernobyl fallout data (Bq d/m^3).

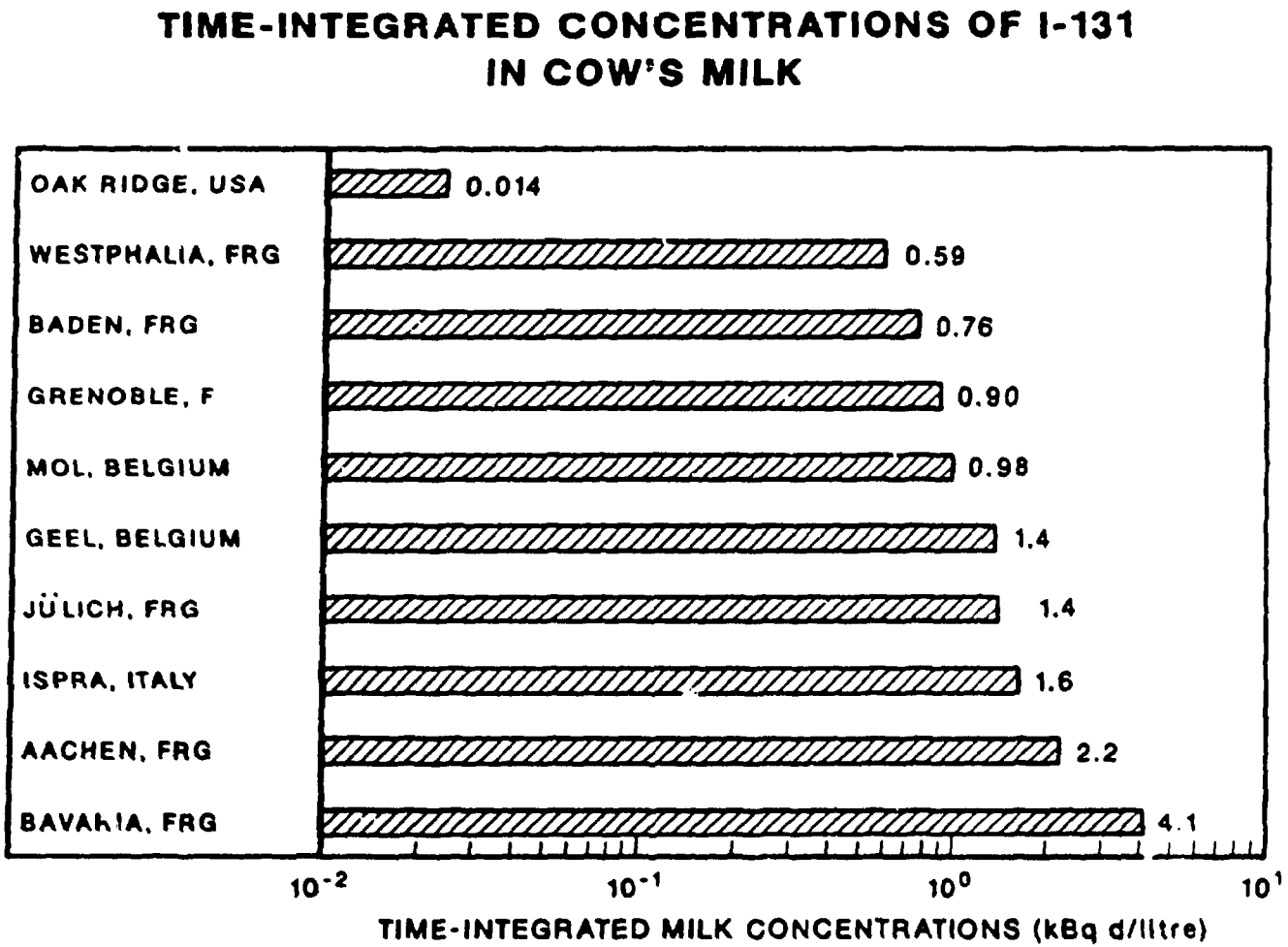

Fig. 2.3-2. Time-integrated concentrations of ^{131}I in cow's milk obtained from Chernobyl fallout data (kBq d/m^3).

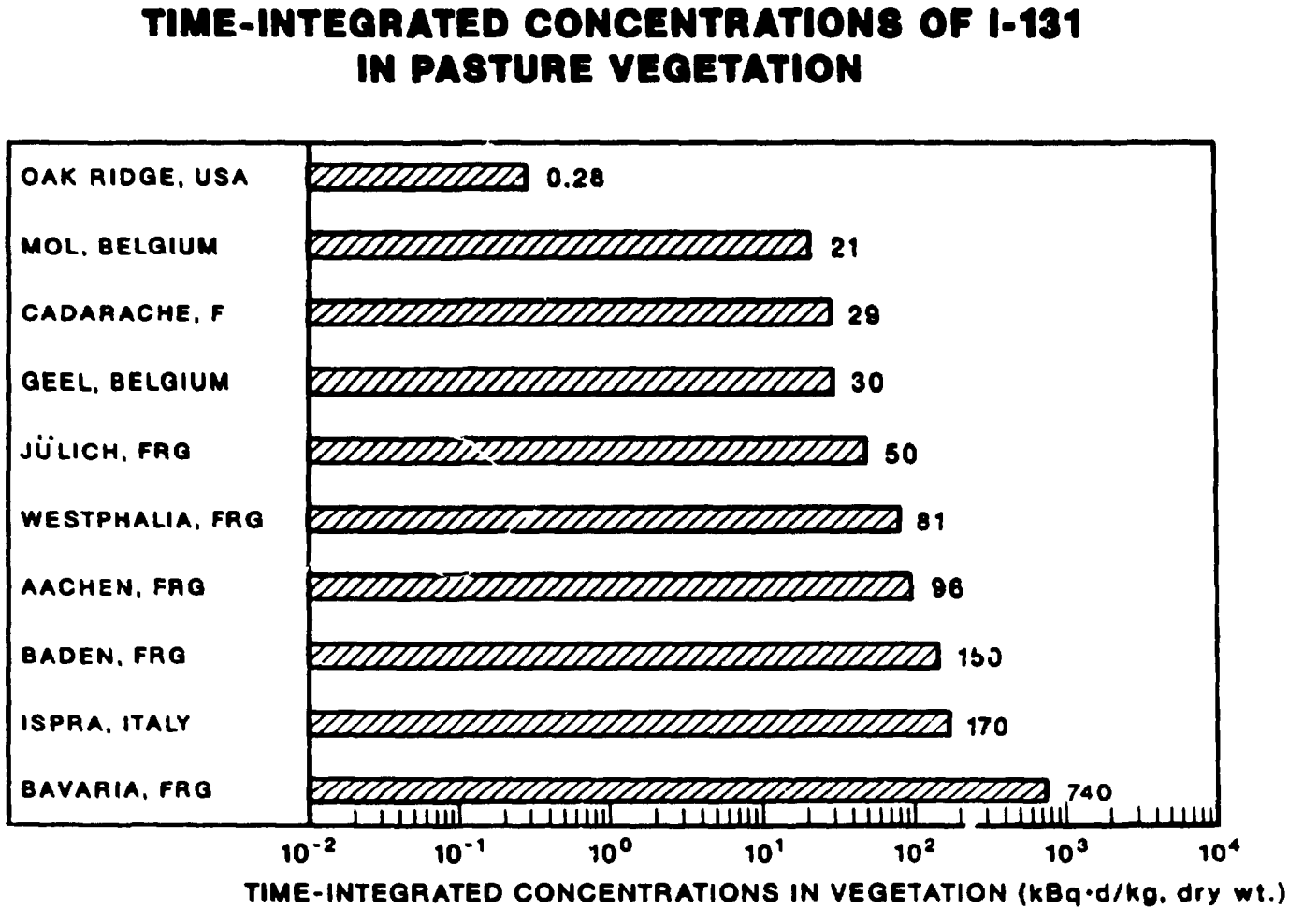

Fig. 2.3-3. Time-integrated concentrations of ^{131}I in pasture vegetation obtained from Chernobyl fallout data [kBq d/kg (dry)].

well as the ingestion of contaminated soil and pasture vegetation. The ingestion of pasture vegetation by dairy cows is expected to be the dominant contributor to ^{131}I in milk, assuming that the animal's diet is composed of at least 10% contaminated pasture forage and that the animal's drinking water is not obtained from cisterns.

Once the time-integrated concentrations in vegetation and milk obtained from Chernobyl data are normalized by the time-integrated concentration in air, the differences between Oak Ridge and Europe are no longer apparent. Iodine-131 concentration ratios for vegetation to air ($CR_{veg/air}$), milk to air ($CR_{milk/air}$), and milk to vegetation ($CR_{milk/veg}$) vary within one order of magnitude for all locations (Table 2.3-1 and Figs. 2.3-4 through 2.3-6), excluding the values of $CR_{milk/air}$ for Grenoble and values of goat's milk $CR_{milk/air}$ and $CR_{milk/veg}$ for Cadarache, France. The high value for Cadarache is not atypical for goat's milk. Throughout Europe, the highest concentrations of ^{131}I in milk were obtained from goats or sheep.

The high $CR_{milk/air}$ concentration ratio for Grenoble may be the result of suspect data obtained for ^{131}I concentrations in air*. Note that, with the exception of the value for Grenoble, the values of $CR_{milk/air}$ obtained for other European locations and for Oak Ridge are not appreciably different than values estimated from the Environmental Protection Agency's (EPA's) data base on Chernobyl fallout for numerous other sites in the United States and Canada (Lesslie and Durfee 1986).

2.3.1.4 Selection of Models

All models selected for testing in this report have been developed for predicting equilibrium concentrations in vegetation and milk as a result of a continuous release of ^{131}I. However, for an accidental release, the infinite time integral of concentrations in air, pasture

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*Recent information received from N. Parmentier of the CEA, Paris, confirms that air concentrations reported for Grenoble should be increased by a factor of 4 to account for the total amount of ^{131}I in air. Thus the $CR_{milk/air}$ for Grenoble should be reduced by the same factor.

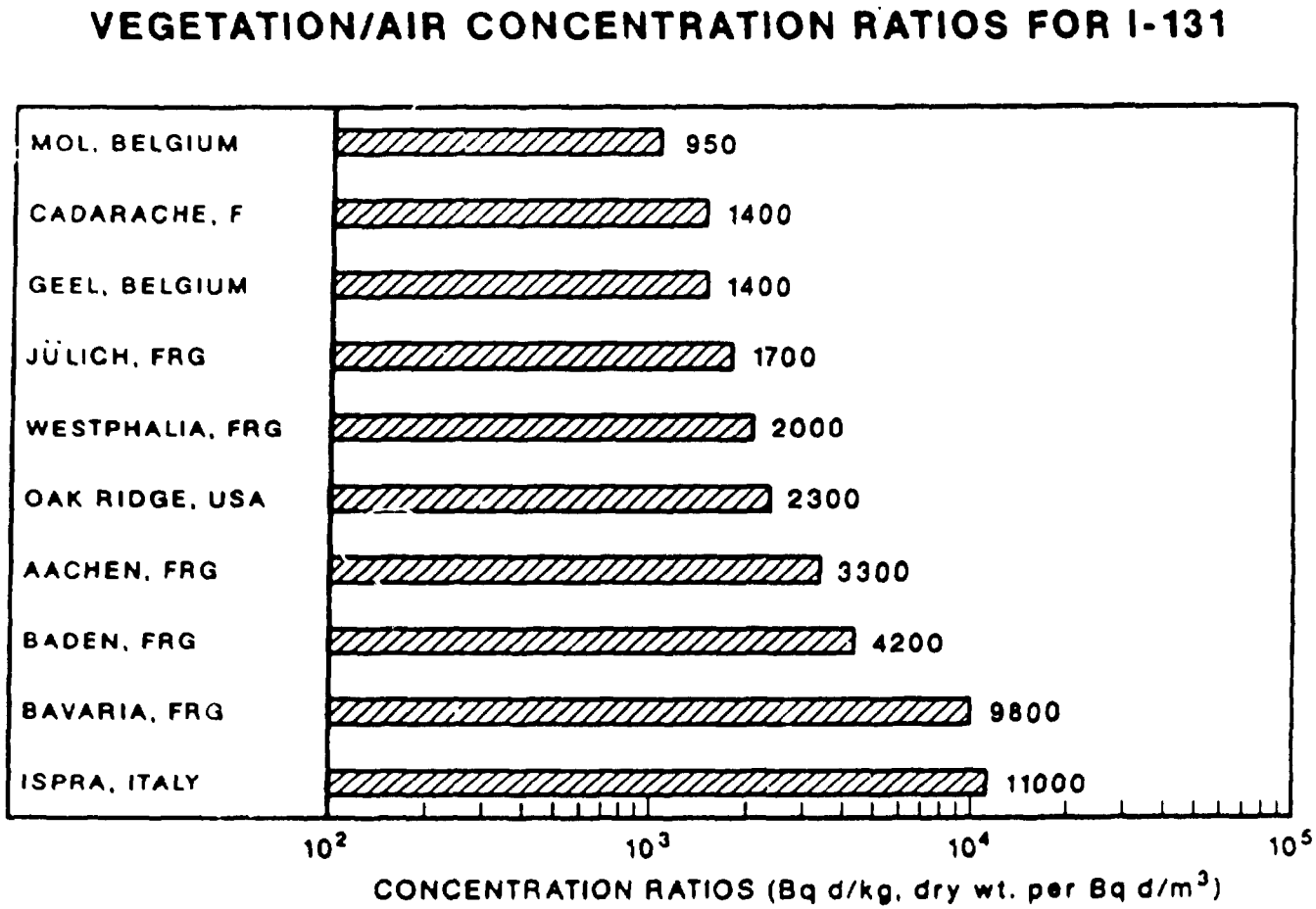

Fig. 2.3-4. Vegetation/air concentration ratios for ^{131}I obtained from Chernobyl fallout data [m^3/kg (dry)].

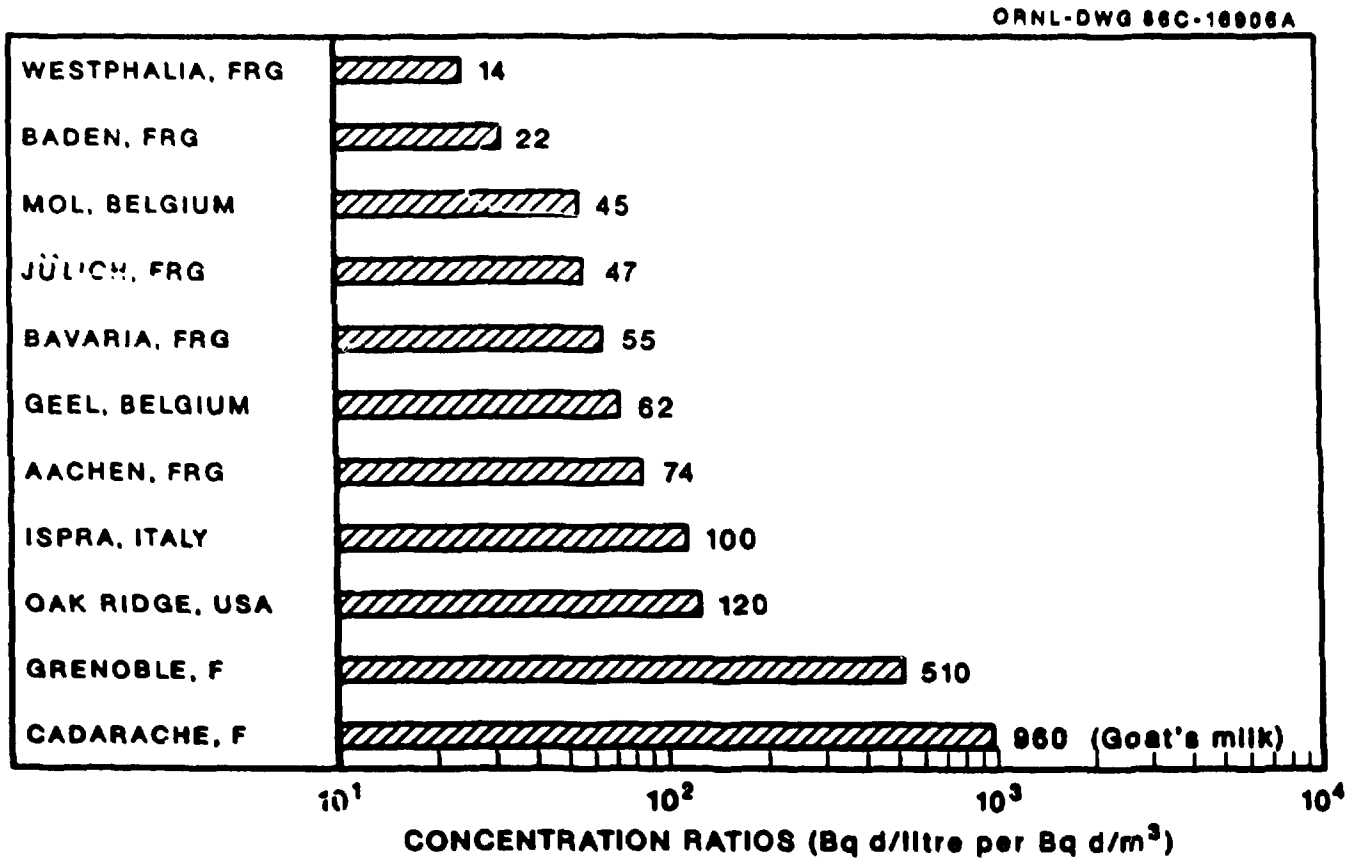

Fig. 2.3-5. Milk/air concentration ratios for ^{131}I obtained from Chernobyl fallout data (m^3/L).

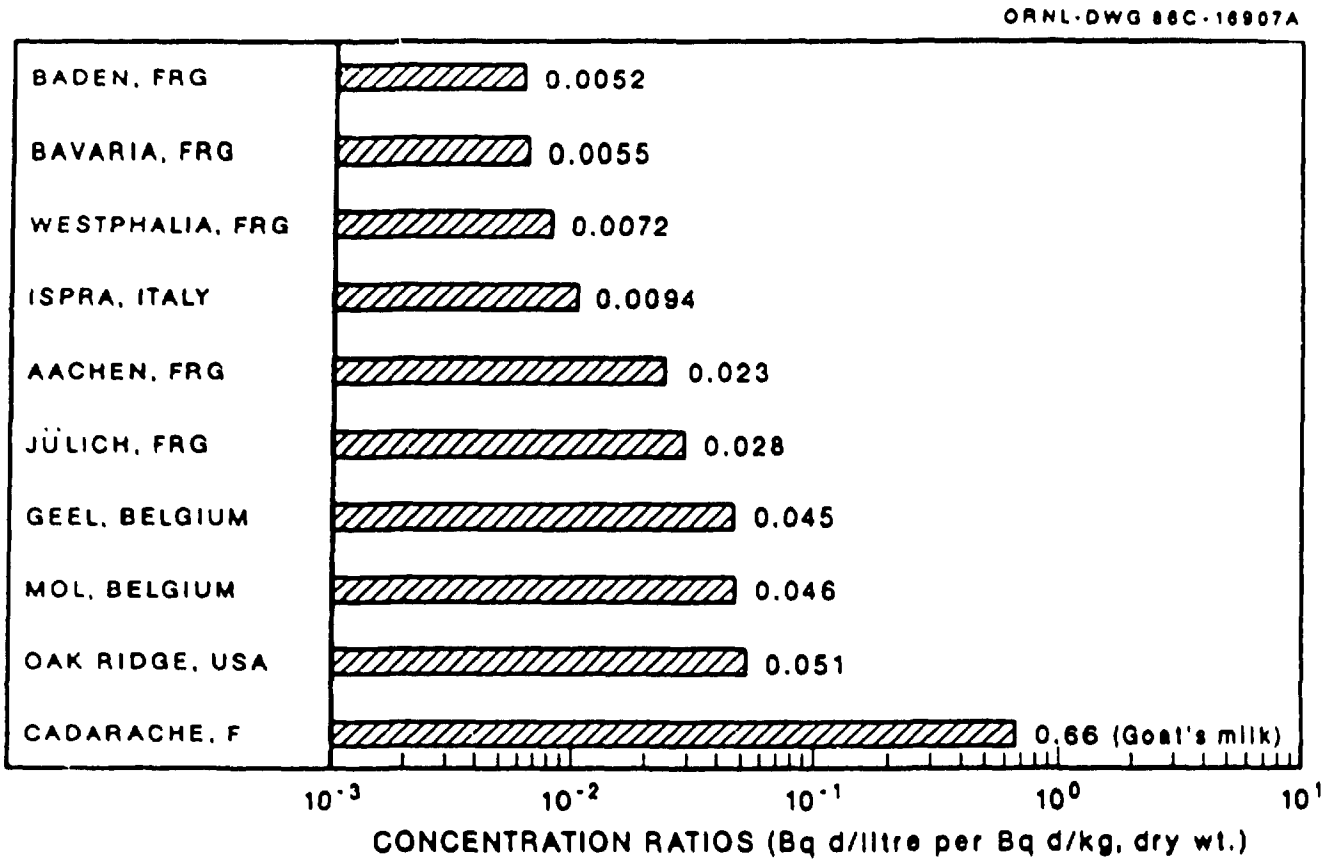

Fig. 2.3-6. Milk/vegetation concentration ratios for ^{131}I obtained from Chernobyl fallout data (kg (dry)/L).

forage, and milk is conceptually equal to equilibrium concentrations resulting from a continuous rate of release of the same amount of radioactivity (Barry 1979). Therefore, time-integrated concentrations derived from Chernobyl fallout data were compared with calculated time-integrated concentrations produced from models developed for the assessment of routine discharges.

The models included in this analysis are: IAEA Safety Series No. 57 (IAEA 1982), AIRDOS/EPA (Moore et al. 1979), NRC Regulatory Guides 1.109 and 1.111 (USNRC 1977a, 1977b), the CRRIS system (Miller et al. 1986, Baes et al. 1984), and the screening model recently introduced by Scientific Committee 64 of the National Council on Radiation Protection and Measurements (NCRP 1986). These models are frequently used in the United States and internationally to evaluate the potential environmental contamination of routine discharges of radionuclides to the atmosphere.

Differences among the model predictions are expected because these models were developed for different purposes. The IAEA and NCRP models were developed as screening tools for establishing compliance with dose limits to critical population subgroups. The CRRIS system and AIRDOS/EPA were developed to assess average exposures to members of the general public for assisting EPA in promulgating standards under the auspices of the Clean Air Act. The NRC Regulatory Guides were produced to assess doses to maximally exposed and average individuals within a 50-mile radius of commercial light water power reactors to determine compliance with mandated design objectives. Nevertheless, for conditions in which cows receive contaminated pasture as part of their diet, the results of these models should be similar to time-integrated concentrations produced by dynamic models because of similarities in the data sets from which all of these models were developed (Hoffman 1977, Hoffman et al. 1984).

2.3.1.5 Assumptions

When information was not available, assumptions were made to approximate environmental conditions prevailing at the time that pastures and grazing animals were exposed to Chernobyl fallout.

Specific information required by the models, but not available at the time of this analysis, included the amount of wet deposition that occurred, the amount of stored, uncontaminated feed given as a supplement to cows on pasture, and the composition of the chemical form of radioiodine in air. To permit a preliminary comparison of model predictions with data, the following assumptions were made:

1. a precipitation rate of 2.7 mm/d, corresponding to an average precipitation of 1 m/year in the southeastern United States and western Europe;
2. a 60% stored feed supplement to a diet of fresh pasture [a practice often used by major dairies (Shor and Fields 1979)];
3. a composition of ^{131}I in air of 22% particulates, 51% organic iodide, and 27% elemental iodine, averaged from data provided by the Federal Health Office in Neuherberg, Federal Republic of Germany (FRG) (Wirth 1986); and
4. a dry matter content of fresh pasture vegetation of 20% to convert concentrations reported on a fresh weight basis.

2.3.1.6 Model Predictions

The concentration ratios (CRs) predicted by the models are given in Table 2.3-2 for the relationships of milk to air, vegetation to air, and milk to vegetation. The differences among model predictions vary within each category of CR by a factor of 2 to 6. For $CR_{milk/air}$ and $CR_{milk/veg}$, the highest values are predicted by the NCRP screening model (2800 m^3/L and 0.14 kg/L, respectively). For $CR_{veg/air}$, the highest value is predicted by AIRDOS/EPA [42,000 m^3/kg (dry)]. The lowest values for $CR_{milk/air}$ and $CR_{milk/veg}$ are produced by the NRC Regulatory Guides (510 m^3/L, and 0.026 kg/L). The NRC Regulatory Guides and the NCRP screening model share the lowest value predicted for $CR_{veg/air}$ [20,000 m^3/kg (dry)].

Table 2.3-2. Model predictions of concentration ratios for ^{131}I transferred from the atmosphere to pasture vegetation and milk

Concentration ratio	Model predictions[a]				
	IAEA	AIRDOS/EPA	NRC	CRRIS	NCRP
$CR_{milk/air}$, m^3/L	9.6E+02	2.2E+03	5.1E+02	1.4E+03	2.8E+03
$CR_{veg/air}$, m^3/kg (dry)	2.1E+04	4.2E+04	2.0E+04	2.3E+04	2.0E+04
$CR_{milk/veg}$, kg (dry)/L)	4.5E-02	5.2E-02	2.6E-02	6.1E-02	1.4E-01

[a]Assumptions for all models but NCRP: a daily precipitation rate of 2.7 mm/day; a 60% stored feed supplement to a diet of fresh pasture; and a composition of ^{131}I in air of 22% particulates, 51% organic iodide, and 27% elemental iodine.

2.3.1.7 Comparison of Model Predictions with Data

The comparison of concentration ratios obtained from model predictions and measurements of Chernobyl fallout is presented as a ratio of predictions to observations (P/O). Values of P/O that approach unity indicate good agreement between model predictions and observations; values of P/O that are greater than unity indicate that the model overpredicts observations; values less than unity indicate that the model underpredicts (Hoffman and Gardner 1983).

In general, the model predictions of all categories of CR are substantially higher than the CRs obtained from Chernobyl fallout data. Exceptions to this generality are the NRC, AIRDOS/EPA, CRRIS, and IAEA predictions of $CR_{milk/veg}$ for about five of nine locations. Results of the comparison of predicted to observed ratios are presented in Table 2.3-3 and in Figs. 2.3-7 through 2.3-9.

In these comparisons, IAEA Safety Series No. 57 is used as a reference. For example, IAEA overpredicts $CR_{milk/air}$ by a factor of approximately 15 on the average, with overpredictions for individual locations varying by factors of 1.9 to 68 (Table 2.3-3 and Fig. 2.3-7). By comparison, the extent of overprediction of the other models varies on the average from about a factor of 8.1 (NRC) to 45 (NCRP) with P/O ratios for individual locations varying by factors of 1.0 to 200.

For $CR_{veg/air}$, the IAEA model overpredicts by a factor of 7.9 on the average, with overpredictions for single locations ranging from factors of 1.9 to about 22 (Table 2.3-3 and Fig. 2.3-8). Overpredictions produced by the other models vary on the average by a factor of 7.5 (NRC and NCRP) to 16 (AIRDOS/EPA) and by factors of 1.8 to 43 for single locations.

$CR_{milk/veg}$ is predicted reasonably well for five of nine locations by all models, with the exception of the NCRP screening model (which overpredicts all observations by factors of from 2.7 to 27) (Fig. 2.3-9). Excluding the NCRP results, P/O ratios for these five locations vary from 0.49 (NRC) to 2.7 (CRRIS) (Table 2.3-3). For the other four locations, all models overpredict observations by factors varying from 2.8 (NRC) to 12 (CRRIS).

Table 2.3-3. Predicted-to-observed ratios (P/O) for the concentration ratios (CR) of milk/air, vegetation/air, and milk/vegetation

Location	IAEA	NRC	CRRIS	AIRDOS/EPA	NCRP
			P/O for $CR_{milk/air}$		
Grenoble, France	1.9E+00	1.0E-00	2.8E+00	4.3E+00	5.5E+00
Oak Ridge, United States	8.4E+00	4.4E+00	1.2E+01	1.9E+01	2.4E+01
Ispra, Italy	9.2E+00	4.9E+00	1.4E+01	2.1E+01	2.7E+01
Aachen, Germany (FRG)	1.3E+01	6.9E+00	1.9E+01	3.0E+01	3.7E+01
Geel, Belgium	1.6E+01	8.2E+00	2.3E+01	3.6E+01	4.5E+01
Bavaria, FRG	1.8E+01	9.4E+00	2.6E+01	4.0E+01	5.1E+01
Jülich, FRG	2.1E+01	1.1E+01	3.0E+01	4.7E+01	6.0E+01
Mol, Belgium	2.1E+01	1.1E+01	3.1E+01	4.9E+01	6.2E+01
Baden, FRG	4.4E+01	2.3E+01	6.4E+01	1.0E+02	1.3E+02
Westphalia, FRG	6.8E+01	3.6E+01	9.9E+01	1.6E+02	2.0E+02
Geometric mean	1.5E+01	8.1E+00	2.2E+01	3.5E+01	4.5E+01
Geometric standard deviation	2.7E+00	2.7E+00	2.7E+00	2.7E+00	2.7E+00
			P/O for $CR_{veg/air}$		
Ispra, Italy	1.9E+00	1.8E+00	2.1E+00	3.8E+00	1.8E+00
Bavaria, FRG	2.2E+00	2.1E+00	2.4E+00	4.3E+00	2.1E+00
Baden, FRG	5.0E+00	4.8E+00	5.5E+00	1.0E+01	4.8E+00
Aachen, FRG	6.5E+00	6.2E+00	7.2E+00	1.3E+01	6.2E+00
Oak Ridge, United States	9.4E+00	9.0E+00	1.0E+01	1.9E+01	8.9E+00
Westphalia, FRG	1.1E+01	1.0E+01	1.2E+01	2.2E+01	1.0E+01
Jülich, FRG	1.3E+01	1.2E+01	1.4E+01	2.5E+01	1.2E+01
Cadarache, France	1.6E+01	1.5E+01	1.7E+01	3.1E+01	1.5E+01
Geel, Belgium	1.6E+01	1.5E+01	1.7E+01	3.1E+01	1.5E+01
Mol, Belgium	2.2E+01	2.1E+01	2.4E+01	4.3E+01	2.1E+01
Geometric mean	7.9E+00	7.5E+00	8.7E+00	1.6E+01	7.5E+00
Geometric standard deviation	2.3E+00	2.3E+00	2.3E+00	2.3E+00	2.3E+00
			P/O for $CR_{milk/veg}$		
Oak Ridge, United States	8.8E-01	4.9E-01	1.2E+00	1.0E+00	2.7E+00
Mol, Belgium	9.9E-01	5.5E-01	1.3E+00	1.1E+00	3.1E+00
Geel, Belgium	1.0E+00	5.6E-01	1.4E+00	1.2E+00	3.1E+00
Jülich, FRG	1.6E+00	9.1E-01	2.2E+00	1.9E+00	5.0E+00
Aachen, FRG	2.0E+00	1.1E+00	2.7E+00	2.3E+00	6.1E+00
Ispra, Italy	4.8E+00	2.7E+00	6.6E+00	5.6E+00	1.5E+01
Westphalia, FRG	6.3E+00	3.5E+00	8.5E+00	7.3E+00	1.9E+01
Bavaria, FRG	8.2E+00	4.6E+00	1.1E+01	9.4E+00	2.5E+01
Baden, FRG	8.7E+00	4.9E+00	1.2E+01	1.0E+01	2.7E+01
Geometric mean	2.6E+00	1.5E+00	3.6E+00	3.0E+00	8.2E+00
Geometric standard deviation	2.6E+00	2.6E+00	2.6E+00	2.6E+00	2.6E+00

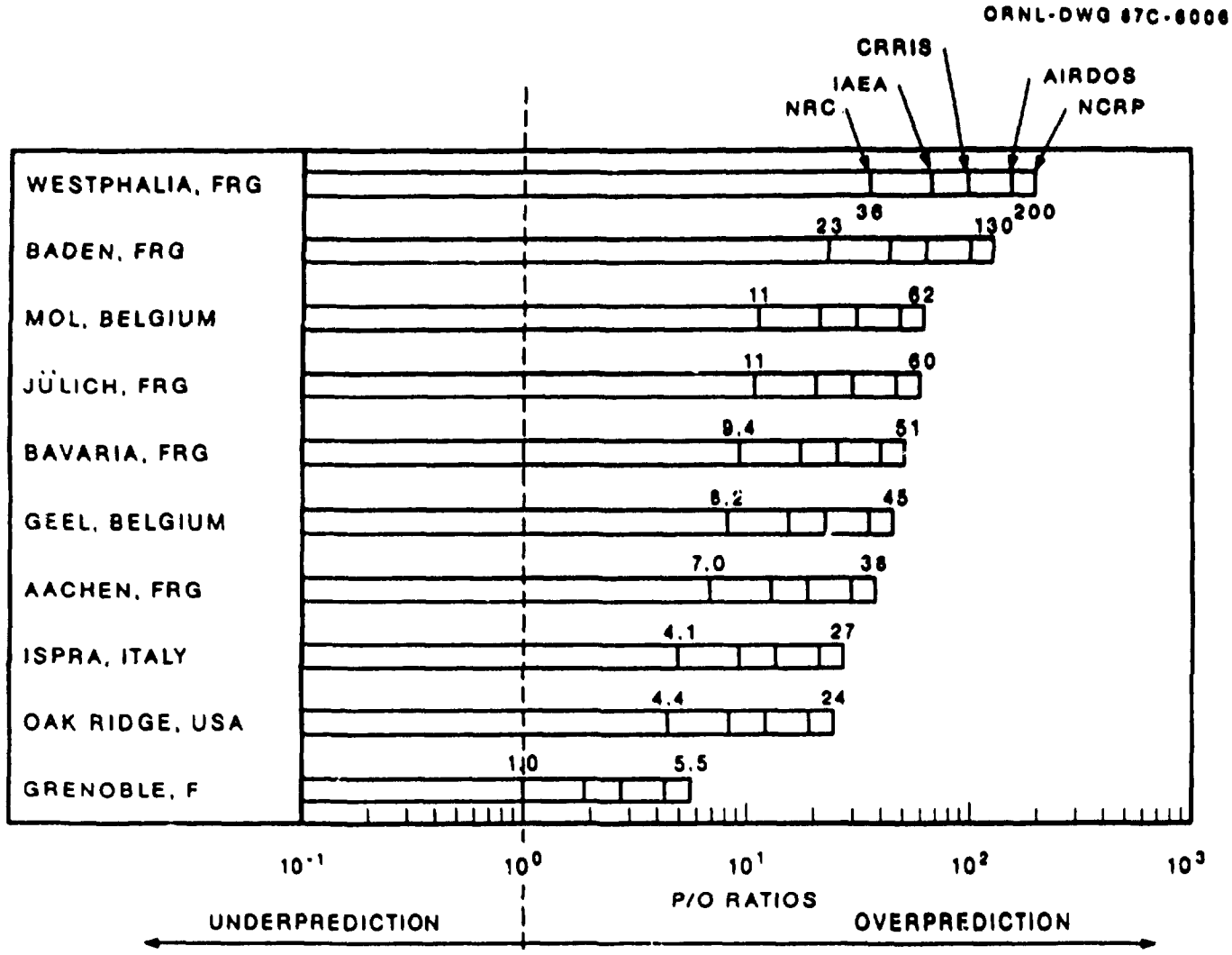

Fig. 2.3-7. A comparison of model predictions vs. observations (P/O) for the milk/air concentration ratio for ^{131}I.

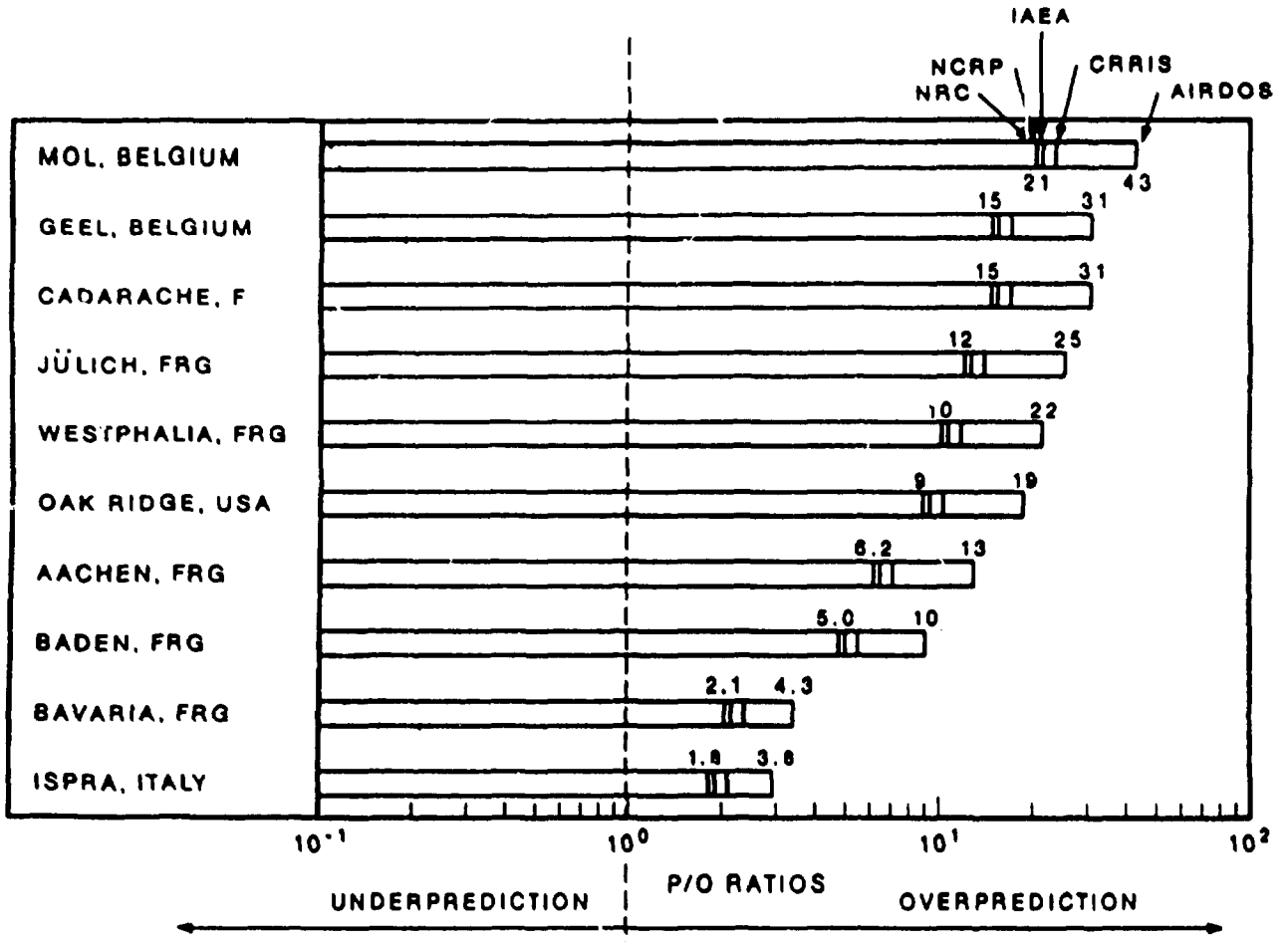

Fig. 2.3-8. A comparison of model predictions vs. observations (P/O) for the vegetation/air concentration ratio for ^{131}I.

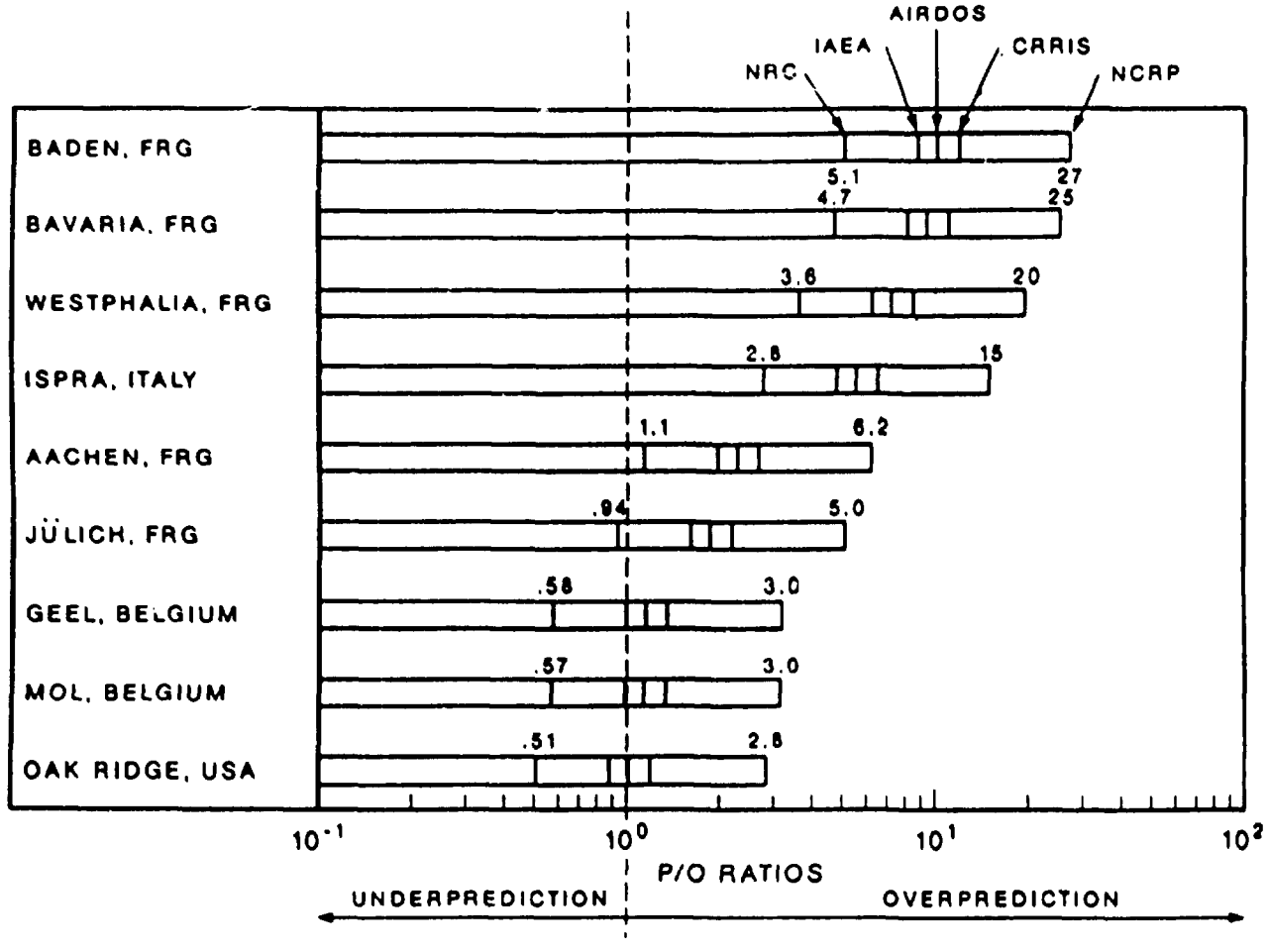

Fig. 2.3-9. A comparison of model predictions vs. observations (P/O) for the milk/vegetation concentration ratio for ^{131}I.

2.3.1.8 Goat's Milk

Cadarache, France, was the only location from which data were obtained for goat's milk (Figs. 2.3-5 and 2.3-6). Of the models selected, however, only the NRC Regulatory Guides explicitly consider goat's milk. The IAEA and NCRP models, on the other hand, are screening tools, and, as such, their predicted values are intended to be applicable to situations in which milk has been obtained from either cows or goats. It was assumed that the diet of the goat was composed entirely of fresh pasture. All three of these models moderately overpredict the ^{131}I transfer from air to goat's milk ($CR_{milk/air}$) by factors of from 1.5 to almost 3 (Table 2.3-4). This seemingly close prediction is the consequence of an overprediction of the air-to-vegetation transfer offsetting an underprediction of the transfer from vegetation to goat's milk ($CR_{milk/veg}$). The $CR_{milk/veg}$ value for goat's milk obtained from Chernobyl fallout data is comparable to the situation in which a goat receives 1.5 kg/d (dry) of contaminated forage and has a diet-to-milk transfer coefficient (F_m) of 0.45 d/L. This situation is consistent with values reported in the literature for goat's milk (Hoffman 1979).

2.3.1.9 Discussion

The models selected in this preliminary analysis for predicting the transfer of ^{131}I over the air-pasture-cow-milk pathway have been developed from an extensive base of data. The results of other models, including dynamic models developed for accident assessment, should not differ substantially from the model predictions summarized here because of basic similarities among the data sets (Hoffman et al. 1984). Despite this fact, the transfer of ^{131}I from air to milk derived from Chernobyl data is grossly overpredicted for all locations except Grenoble, France.

The reported air concentrations for Grenoble could possibly represent the particulate fraction of iodine in air rather than the total amount (particles and gaseous forms). The particulate fraction

Table 2.3-4. A comparison of model predictions and observations of concentration ratios for ^{131}I transferred from the atmosphere and pasture vegetation to goat's milk

	Observed values	Model predictions[a]		
	(Cadarache, France)	IAEA	NRC	NCRP
$CR_{milk/air}$ (m^3/L)	9.6E+02	2.4E+03	1.4E+03	2.8E+03
$CR_{milk/veg}$ [kg (dry)/L]	6.6E-01	1.1E-01	7.6E-02	1.4E-01
Predicted-to-observed ratios				
$P/O_{milk/air}$		2.5E+00	1.5E+00	2.9E+00
$P/O_{milk/veg}$		1.7E-01	1.2E-01	2.1E-01

[a]Assumptions for all models but NCRP: a daily precipitation rate of 2.7 mm/d; a 100% diet of fresh pasture; and a composition of ^{131}I in air of 22% particulates, 51% organic iodide, and 27% elemental iodine.

of iodine in air has been reported to vary from about 15 to 40% of the total, with the higher value being an upper limit estimate (Bondietti and Brantley 1986, Bonka 1986, Schell et al. 1986, STUK 1986). If the Grenoble air concentrations represented only the particulate fraction of ^{131}I, correcting for the total quantity of ^{131}I in air would reduce $CR_{milk/air}$ for Grenoble by a factor of 2.5 to about 7, and models would also grossly overpredict for this location.*

Two questions therefore arise: Why is the extent of misprediction so large, and why do all models tend to grossly overestimate observed relationships? To answer these questions explicitly, data on all parameters pertinent to the model calculations would be needed for each location. This information is not currently available, although in time much of these data are expected to be obtained through continued participation in international model validation studies. In the absence of such data, explanation of the sources of misprediction is limited to the speculative process of calibrating initial assumptions and parameter values.

Explanation of discrepancies between model predictions and Chernobyl data

One source of misprediction would be that the models included in this study were intentionally designed to overpredict actual concentrations and doses. However, among the models included in this study, only IAEA Safety Series No. 57 and the NCRP screening model admit a conservative bias. For example, under extreme circumstances, dose estimates made with the IAEA and NCRP models are intended to underpredict actual doses to critical population groups by no more than

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*Recent information received from N. Parmentier of the CEA, Paris, confirms that air concentrations reported for Grenoble should be increased by a factor of 4 to account for the total amount of ^{131}I in air. Thus the $CR_{milk/air}$ for Grenoble should be reduced by the same factor.

one order of magnitude. The other models contain no quantitative guidance regarding the uncertainty associated with their results.

Another source of error is the initial assumptions made about site-specific conditions of precipitation, the amount of fresh pasture consumed by dairy cows, and the physicochemical forms of ^{131}I in air.

However, any changes that would increase the assumed amount of precipitation, the consumption of fresh pasture, or the fraction of total iodine in air that is present as elemental vapor would result in even larger predicted ^{131}I concentrations in pasture vegetation and in milk. Thus, the relationships obtained from Chernobyl data would be even more severely overpredicted. Therefore, to calibrate model predictions to the relationships derived from observations, the values associated with initial assumptions about site-specific conditions must be decreased.

For most sites in Europe and at Oak Ridge, concentrations of ^{131}I in vegetation were influenced by wet deposition; the data on ^{131}I concentrations in milk were reportedly based on cows grazing outside, and the assumption about the fraction of elemental ^{131}I in air was based on the only measurement available to date. In addition, some of the models selected for testing are not sensitive to changes in these assumptions. For example, the NCRP screening model cannot be adjusted for differences in the chemical forms of iodine in air, local agricultural practices, or local amounts of precipitation. NRC Regulatory Guide 1.111 does not include the process of wet deposition within its air-to-ground transfer model. Nevertheless, changes in the assumption of wet deposition should have only a minor influence on the estimates of total deposition predicted by all other models. This is because the magnitude of values chosen for the deposition velocity of elemental iodine (2 to 3.5 cm/s) results in dry deposition of ^{131}I being a large fraction (25 to 60%) of the total predicted deposition. Calibration of model results to Chernobyl fallout data therefore requires the adjustment of more than one assumption and/or parameter value.

Adjusting values for wet and dry deposition and the atmospheric forms of ^{131}I

For the entire air-pasture-cow-milk pathway, the most severe overpredictions were evident for the portion of the pathway between air and pasture (Fig. 2.3-8). Bavaria, Germany, and Ispra, Italy, were the only two locations for which some model predictions (IAEA, NRC, and NCRP) came relatively close (i.e., within a factor of 2). By assuming no elemental iodine in air and half the initial precipitation rate (1.4 mm/d), the NRC and IAEA models come within a factor of 2 of $CR_{veg/air}$ of the four other German sites and Oak Ridge (Fig. 2.3-10). The other models overpredicted these locations by factors ranging from 1.6 to 9.6. No changes were made to the NCRP screening model because it cannot accept changes in assumptions about precipitation and iodine chemistry.

By assuming no wet deposition, 5% elemental iodine of the total ^{131}I in air, and a dry deposition velocity of 1 cm/s, the IAEA, CRRIS, and AIRDOS/EPA models come within a factor of 2 of the remaining locations (Mol and Geel, Belgium, and Cadarache, France), but NRC overpredicts these locations by factors ranging from 5.4 to 7.8 (Fig. 2.3-11). Cadarache, in fact, did not report any wet deposition (CEA 1986), but Mol and Geel received wet deposition some days after occurrence of the maximum ^{131}I concentrations in air (CEN/SCK 1986).

The assumption of little or no elemental ^{131}I in air is necessary to calibrate model predictions. Nevertheless, this assumption cannot be justified unless the measured values of the physicochemical forms of ^{131}I in air obtained from the Federal Health Office in Neuherberg, FRG, are either unrepresentative or biased. [Informal communication with Japanese investigators has provided preliminary information on measurements they have obtained indicating that the fraction of total iodine in air in the elemental form is on the order of 5% (Homma 1986).]

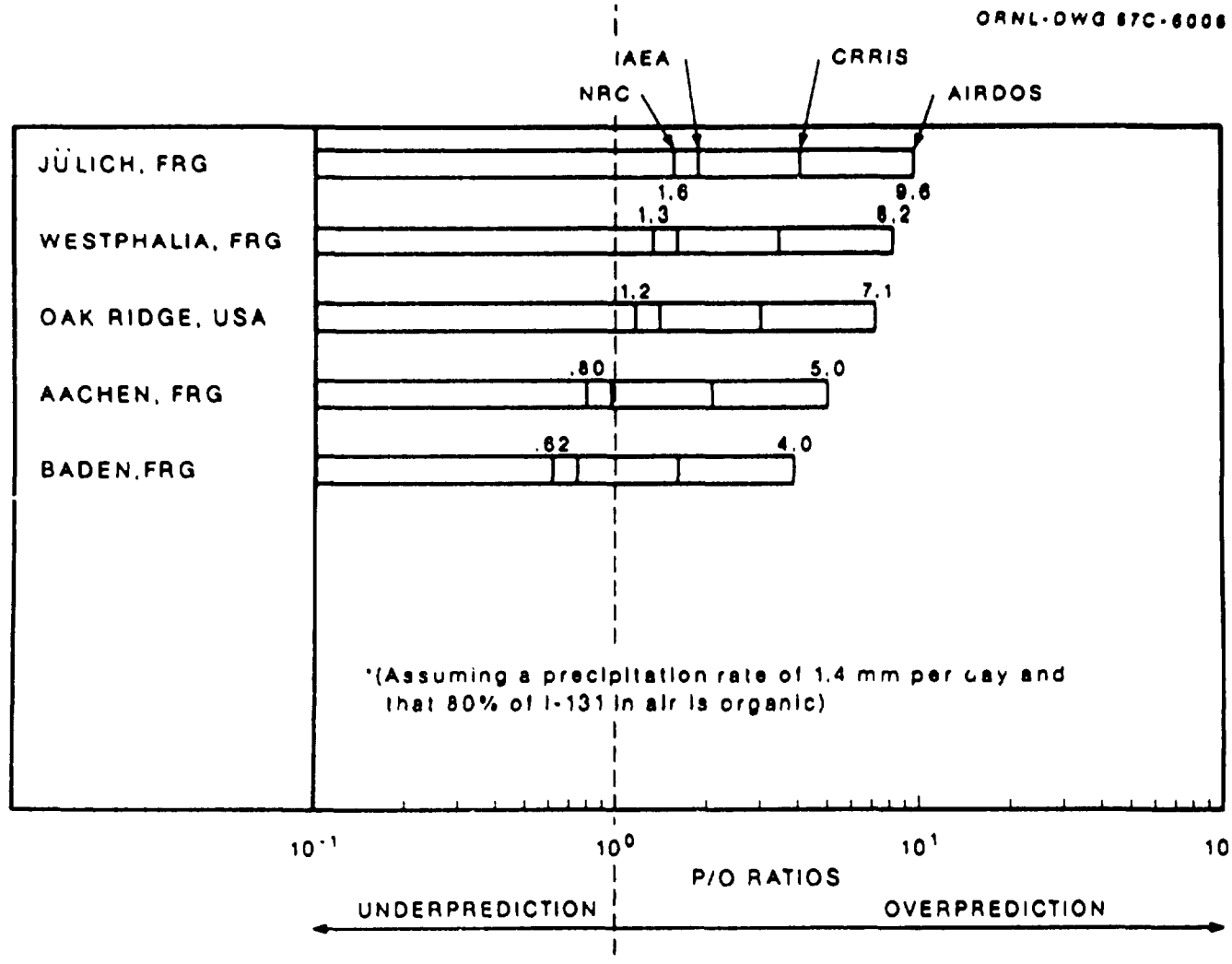


Fig. 2.3-10. A comparison of calibrated model predictions vs. observations (P/O) for the vegetation/air concentration ratio (assuming that precipitation rate is 1.4 mm/d and that 80% of ^{131}I in air is organic and 20% is particulate).

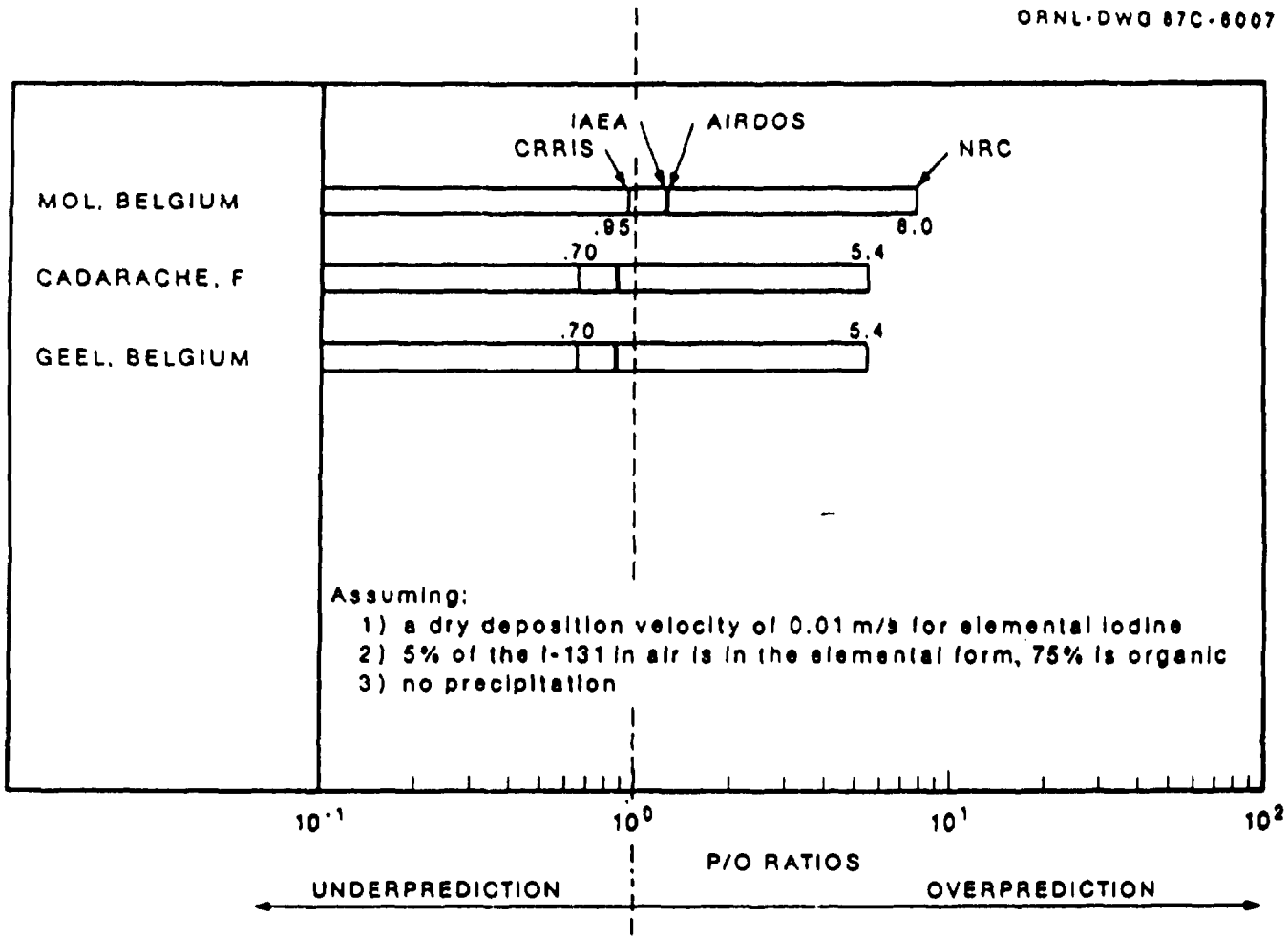


Fig. 2.3-11. A comparison of calibrated model predictions vs. observations (P/O) for the vegetation/air concentration ratio (assuming that the dry deposition velocity is 0.01 m/s for elemental iodine, that 5% of the ^{131}I in air is in the elemental form and 75% is organic, and that there is no precipitation).

Vegetation interception and retention of wet-deposited ^{131}I

The effect of wet deposition can be partially compensated for by reducing the fraction of wet deposited ^{131}I initially intercepted by vegetation. For example, Zeevaert (1986) reports a mass interception factor for pasture grasses during wet deposition in Belgium of 0.4 m^2/kg (dry), which is a factor 4 to 5 times less than assumed by those models that consider wet deposition. A similar value, 0.8 m^2/kg (dry), was obtained from Neuherberg, FRG (GSF 1986), which received a total of 80% wet deposition. These lower values of mass interception factors are probably the result of decreased retention of ^{131}I by vegetation surfaces and increased washoff during heavy precipitation. These obvious processes that occur during periods of wet deposition are not considered by any radiological assessment model known to the authors of this document.

The retention of ^{131}I on pasture vegetation assumed by the models is related to "weathering" half-lives of from 8 to 14 d. Estimates made from data obtained from Munich, Jülich, and Aachen, FRG, are 8.8, 8.9, and 7.5 d, respectively (GSF 1986, KFA 1986, Bonka 1986), and are representative of conditions of rapid spring growth. Differences between assumed and observed values, however, will be of minor importance because of the relatively short radiological half-life of ^{131}I (8.05 d).

^{131}I milk transfer coefficient

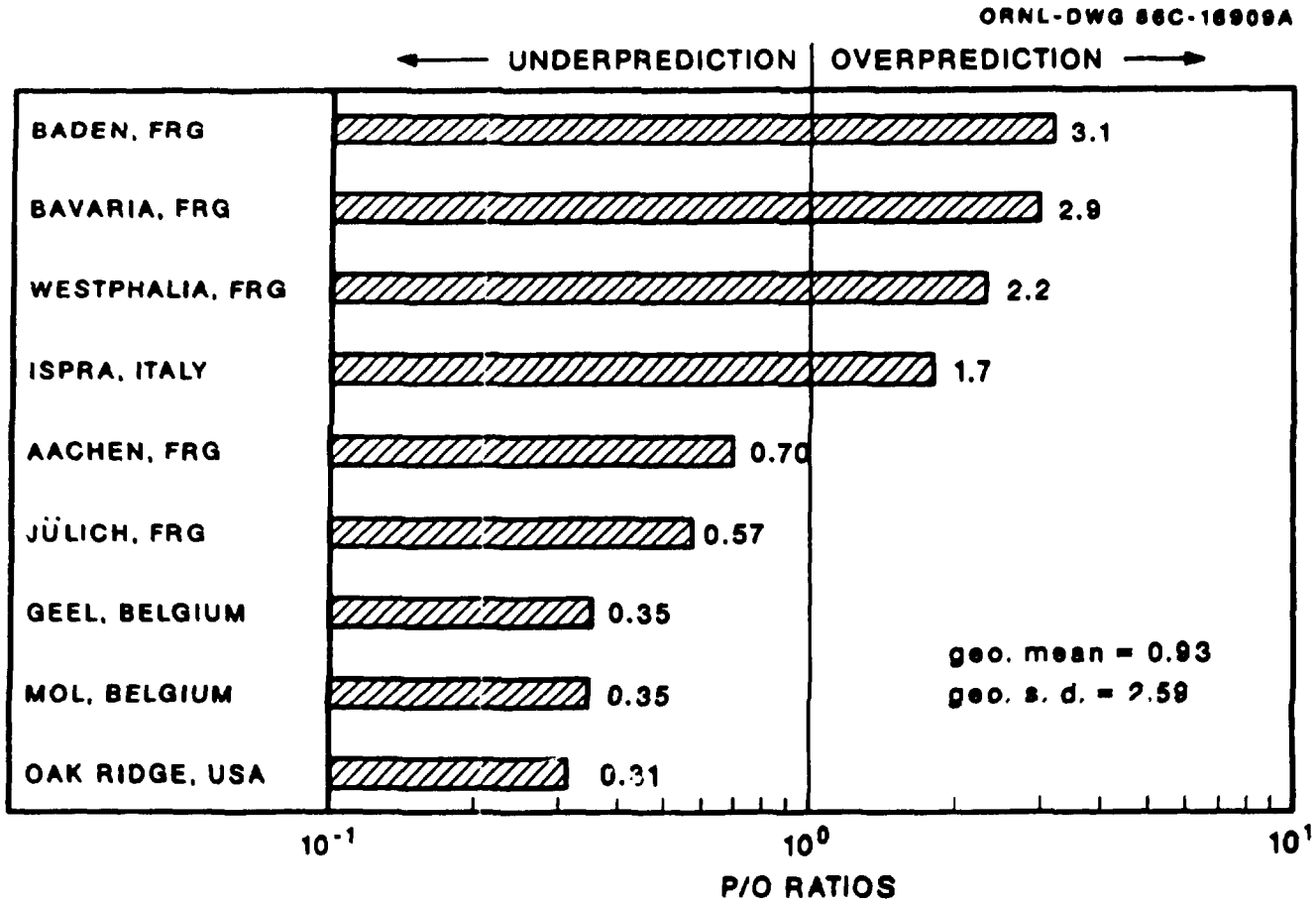
The literature abounds with experimental evidence (see reviews by Hoffman 1978, 1979; Ng et al. 1977; Ng and Hoffman 1983) substantiating a cow's diet-to-milk transfer coefficient F_m for ^{131}I of 0.01 d/L, which is the value most commonly used for ^{131}I in radiological assessment models. However, subsequent to Chernobyl fallout, values approaching 0.002 d/L have been reported by several European investigators (Bonka et al. 1986, Kühn et al. 1986). These low values of F_m for ^{131}I may represent metabolic changes occurring in the spring when cows are in transition from a diet of stored winter feed to

fresh pasture (Peterson 1983, Lengemann et al. 1957). Calculated values of $CR_{milk/veg}$ would be within a factor of approximately 3 for all locations if model assumptions were changed to an F_m of 0.002 d/L, a stored feed intake of 50%, and a total dry matter intake by the cow of 16 kg/d (Fig. 2.3-12). However, the $CR_{milk/veg}$ is still overpredicted for Baden, Bavaria, and Westphalia, FRG, and underpredicted for the five locations for which the models previously produced reasonably accurate estimates.

The overpredictions produced for the three German locations cannot be explained unless the diet of the animals was supplemented with about 75 to 80% stored feed. On the other hand, the underpredictions produced for the other locations might be explained by reducing the stored feed intake to 25% and increasing the F_m to 0.004 d/L. Determining which set of assumptions is most correct cannot be accomplished without additional site-specific information. As mentioned previously, detailed information on the specific management of cows at these German locations is not yet available. Note, however, that the ^{131}I time-integrated milk concentrations obtained for Bavaria (4.1 kBq d/L) is quite comparable to that (5.4 kBq d/L) obtained from the GSF report (1986) for a cow in the Munich area that was reportedly on pasture 100% of the time. We therefore assumed that the values reported for Bavaria were for cows that were grazing on pasture.

Implications of model calibration

By correcting model overpredictions to fit the observed relationships, the initially predicted values of $CR_{milk/air}$ are reduced by one to two orders of magnitude. A typical value of $CR_{milk/air}$ representative of locations receiving Chernobyl fallout would be about 55 m^3/L. Such a low value for the transfer of ^{131}I over the air-pasture-cow-milk pathway means that direct inhalation of ^{131}I in the air would be a major route of human exposure. This conclusion is based on assumptions that the (1) average consumption of fresh milk is 0.25 L/d per capita in the United States (Yang and Nelson 1984), (2) that the inhalation rate for average adults is



*(Assuming 16 kg/d dry matter intake, 50% stored feed, and an F_m OF .002 d/L)

Fig. 2.3-12. A comparison of calibrated model predictions vs. observations (P/O) for the milk/vegetation concentration ratio (assuming 15 kg/d dry matter intake, 50% stored feed, and an F_m of 0.002 d/L).

22 m^3/d (NCRP 1984), and that the retention of inhaled airborne iodine is approximately (85%) (Hoffman 1973a). Thus, on the average, the intake of Chernobyl ^{131}I into the human body via the direct inhalation of iodine in air is about twice as large as for the consumption of contaminated milk. These results contradict the common assumption that exposures to ^{131}I via the consumption of contaminated milk clearly dominate over exposures from inhalation.

For critical population subgroups, however, the milk pathway is still the most important pathway of exposure (Table 2.3-5). Milk is a major constituent of the diet of young children. Children are more sensitive to thyroid irradiation and breathe less air than do adults. In addition, the ^{131}I thyroid dose per unit intake is about one order of magnitude higher for young children than for adults (Hoffman 1973a, Dunning and Schwarz 1981, Killough and Eckerman 1986).

Cisterns as a potential source of exposure

A comparison of ^{131}I exposure pathways via inhalation and milk ingestion has not been analyzed for the case where either cows or humans obtain drinking water from cisterns. A small fraction of the population uses cisterns to collect rainwater for human and animal consumption, and the highest ^{131}I doses to individuals may have occurred as the result of the use of cisterns. However, this potential route of human exposure is not considered by current environmental radiological assessment models. Concentrations of radionuclides in rainwater (Bq/L) may approach or exceed the concentrations of particulate radioaerosols in ground-level air (Bq/m^3) by as much as three orders of magnitude (Slinn 1984). This relationship is approximated by rain/air ratios for particulate ^{131}I obtained from U.S. monitoring data on Chernobyl fallout (see Sect. 3).

Because humans consume more than 1 L/d (and cows up to 100 L/d) of water, the use of cisterns for drinking water results in potential human exposures to ^{131}I that exceed any of the exposure pathways listed in Table 2.3-5. The amount of dilution resulting from the

Table 2.3-5. A comparison of the radiological importance of different pathways of human exposure to ^{131}I given a unit time-integrated concentration of ^{131}I in air (Bq d/m^3)

Exposure pathway	Human exposure (Bq)	
	Average individual	Critical population subgroup
Inhalation	19[a]	4[b]
Ingestion of milk	14[c]	100[d]
Ingestion of leafy vegetables	6.5[e]	50[f]

[a]Based on an average inhalation rate for adults of 22 m^3/day and a retention of inhaled ^{131}I of 0.85.

[b]Based on an average inhalation rate for infants of 5 m^3/d and a retention of inhaled ^{131}I of 0.85.

[c]Based on a $CR_{milk/air}$ of 55 m^3/L and a consumption of 0.25 L/d of milk by adult members of the population.

[d]Based on a $CR_{milk/air}$ of 100 m^3/L and a consumption of 1.0 L/d of milk by small children.

[e]Based on a $CR_{veg/air}$ of 540 m^3/kg (fresh wt.), a 70% loss from washing and preparation, and an average consumption of 0.04 kg/d of leafy vegetables for all age groups.

[f]Based on a $CR_{veg/air}$ of 2000 m^3/kg (fresh wt.), a 50% loss from washing and preparation, and a consumption of 0.05 kg/d of leafy vegetables by adults.

presence of uncontaminated rainwater in cisterns before the arrival of Chernobyl fallout is unknown. Thus, additional investigation of the potential dose from the use of cisterns is warranted.

2.3.1.10 Conclusions

This analysis represents a preliminary attempt to test model predictions against independent data obtained on Chernobyl fallout of ^{131}I concentrations in air, pasture, and milk. Although literature dealing with the transfer of ^{131}I over the air-pasture-cow-milk pathway is extensive, model predictions grossly overestimated observed relationships between air and milk by one to two orders of magnitude. However, the concentration ratios for milk to air ($CR_{milk/air}$) for cow's milk are among the lowest values ever recorded for ^{131}I. Explicit explanation of the causes of misprediction cannot be made without additional site-specific information. Nevertheless, calibration of predictions indicates that overprediction was caused by overestimation of the amount of elemental ^{131}I in air and its associated dry deposition velocity, overestimation of the flux of wet deposited material and its retention by pasture vegetation, and overestimation of the cows' diet-to-milk transfer coefficient.

Corrected model predictions imply that direct inhalation of air may have been more important than the consumption of contaminated milk in contributing to the average ^{131}I exposure to members of the general population. This conclusion is contrary to the common assumption that population doses resulting from exposure to ^{131}I are largely dependent on the consumption of contaminated milk. Recommendations for changes in model parameter values for the models included in this study cannot be made at this time because it is not known if the same extent of overprediction would have occurred again if the accident had occurred during a different time of the year. However, improvements in the model structure of accident assessment models are recommended to permit consideration of the different physicochemical forms of ^{131}I in air, the transfer of ^{131}I to vegetation during periods of wet deposition, and seasonal influences on growth dilution by pasture vegetation and ^{131}I secretion into milk.

Additional results on model testing will become available in the coming year through the International Biospheric Model Validation Project (BIOMOVS 1986), in which the U.S. Department of Energy (DOE) has just recently become a participant. This project involves the collection of Chernobyl data from scientific research establishments for the specific purpose of testing model predictions. At present, detailed data sets from ten countries are being assembled to test model predictions of the transfer of Chernobyl ^{131}I from air to cow's milk. Results from the BIOMOVS project will be very valuable in either substantiating or refuting the conclusions made in this paper.

2.3.2 The Use of Chernobyl Data to Test Model Predictions of the Transfer of ^{137}Cs in Terrestrial Foods

Cesium-137 is another major fission product that has been released to the atmosphere from nuclear weapons tests and nuclear fuel cycle facilities. Its transport through the environment and entry into food chains have been extensively investigated since nuclear devices were first tested.

About 13% of the inventory of the reactor core or about 35 PBq of ^{137}Cs were released from Chernobyl Unit 4 from April 26 through May 6, 1986 (USSR report to the IAEA 1986). Data collection to date on measurements of ^{137}Cs is worldwide. Concentrations so far reported on environmental samples have been used in a preliminary attempt to test model predictions of the transfer of ^{137}Cs over the air-pasture-cow-milk pathway. The potential is discussed for further testing of model predictions of the transfer of ^{137}Cs over this and other food chain pathways as additional results become available.

2.3.2.1 Methods

Sources of data

The data sources for measurements of ^{137}Cs include some of the same sources previously cited for testing model predictions of the

transfer of ^{131}I over the air-pasture-cow-milk pathway (CEN/SCK 1986, GRS 1986) and others (Blaauboer 1986, Dreicer et al. 1986, Klusek et al. 1986, Koehler 1986, Larson et al. 1986). The data resources are part of the ORNL environmental data base on Chernobyl.

Models to be tested

For the immediate objective of exploring the potential of post-Chernobyl ^{137}Cs measurements for testing models that predict ^{137}Cs transfer over the air-pasture-cow-milk pathway, the IAEA Safety Series No 57 model (IAEA 1982) has been selected as the reference model. This model was developed for predicting equilibrium concentrations in vegetation and milk as the result of a continuous release of a radionuclide. Remember that, for an accidental release, the infinite time integral of the concentration in air, pasture vegetation, and milk is conceptually equal to the equilibrium concentration in these media resulting from a continuous rate of release of the same amount of activity (Barry 1979). Therefore, as in the case of ^{131}I, time-integrated ^{137}Cs concentrations from Chernobyl fallout data were compared with time-integrated concentrations calculated by the IAEA Safety Series No. 57 model.

As noted earlier in this report, the IAEA model and other equilibrium models selected for testing predictions of the transfer of ^{131}I over the air-pasture-cow-milk pathway are comparable models that were developed for the assessment of routine releases. These models are structurally very similar, and the required parameter values have been derived from the same data sets. Time-integrated concentrations calculated by these models can therefore be expected to be comparable. The time-integrated concentrations yielded by these models should also be similar to those produced by dynamic models because the dynamic models are largely based on the same data sets (Hoffman et al 1984).

Data analysis

As described in Sect. 2.3.1 for ^{131}I, time-integrated concentrations were calculated for ^{137}Cs in surface air, pasture vegetation, and cow's milk. Because ^{137}Cs, unlike ^{131}I, is long-lived (T_r = 30 years), the concentrations of ^{137}Cs assembled for this report have not decreased to zero or to very low levels. The ^{137}Cs concentrations, particularly those in grass and milk, are therefore incomplete. Indeed, ^{137}Cs concentrations from Chernobyl will continue to be reported in future months and years.

Accordingly, the time-integrated concentrations estimated for this report are truncated integrals determined for selected periods of observations rather than time integrals to infinity as were estimated for ^{131}I. The time-integrated concentrations for this report were estimated for the same period of observation according to the model pathway being tested (e.g., air and grass, air and milk, and grass and milk).

Concentration ratios were then determined from the time-integrated concentrations to normalize the time-integrated concentrations in grass and milk. The concentration ratios take the form and units of $CR_{grass/air}$ (m^3/kg), $CR_{milk/air}$ (m^3/L), and $CR_{milk/grass}$ (kg/L).

Model predictions

Specific information pertaining to environmental conditions and agricultural practices is required as a starting point for making predictions by environmental transport models. This kind of information is not now available for the locations and periods of observation selected for the analysis. Therefore, to allow a preliminary comparison of model predictions with data, some of the same assumptions made for ^{131}I were used for ^{137}Cs:

1. a precipitation rate of 2.7 mm/d, which corresponds to an average precipitation of 1 m/year;

2. a 60% supplement of stored feed to the basal diet of fresh pasture vegetation; and
3. a dry-matter content of 20% for fresh pasture vegetation to convert concentrations reported on a fresh weight basis.

Unlike ^{131}I, atmospheric ^{137}Cs can be regarded as 100% particulate.

Predictions by the IAEA model have taken into account only the transfer of ^{137}Cs to vegetation via deposition on plant surfaces. Transfer of ^{137}Cs to vegetation via root uptake from soil will not be detected until later. The predictions also take into account an integration period, which has been set at 21 d.

2.3.2.2 Results

Time-integrated concentrations

The time-integrated concentrations of ^{137}Cs in surface air (Table 2.3-6) over New York and New Jersey are two to three orders of magnitude less than those for the European locations. The time integral for some of the locations (New York City; Baden-Wurtemberg, FRG) are fairly complete (i.e., the concentrations integrated over the period of observation appear to be very nearly equal to the concentrations integrated to infinity).

The time-integrated concentrations of ^{137}Cs in dry grass (Table 2.3-7) also vary by two to three orders of magnitude. In general, the time-integrated concentrations in grass are incomplete; because the concentrations in grass may be substantial at the end of the observation period, the time-integrated concentrations can be expected to increase as the period of observation increases. Because the concentrations in grass tended to vary irregularly over the observation period, it was generally not possible to extrapolate beyond the period of observation. Time-integrated concentrations in vegetation may also be reported on an areal basis (Table 2.3-8).

The time-integrated concentrations of ^{137}Cs in cow's milk from New York and New Jersey are two to three orders of magnitude less than

Table 2.3-6. Time-integrated concentrations of ^{137}Cs in surface air

Location	Observation period	Integrated air concentration (Bq d/m^3)	References
Chester, N.J., United States (US)	5.5 - 3.6	0.051	Larson et al. (1986)
New York, N.Y., US	6.5 - 12.6	0.05[illegible]	Larson et al. (1986)
Nordrhein-Westfalen, Federal Republic of Germany (FRG)	1.5 - 7.5	6.1	GRS (1986)
Baden-Wurtemberg, FRG	29.4 - 15.5	9.3	GRS (1986)
Munich, FRG	30.4 - 8.5	12	Koehler (1986)
Bavaria, FRG	29.4 - 9.5	38	GRS (1986)
FRG	29.4 - 15.5	71	GRS (1986)

Table 2.3-7. Time-integrated concentrations of ^{137}Cs in grass

Location	Observation period	Integrated grass concentration[a] [kBq d/kg (dry)]	References
Chester, N.J., United States	5.5 - 6.6	0.56	Dreicer et al. (1986)
Groningen, Netherlands	8.5 - 20.5	5.2	Blaauboer (1986)
Mol, Belgium	4.5 - 20.5	13	CEN/SCK (1986)
Hamburg, Federal Republic of Germany (FRG)	3.5 - 26.5	14	GRS (1986)
Niedersachsen, FRG	5.5 - 17.5	17	GRS (1986)
Nordrhein-Westfalen, FRG	6.5 - 16.5	25	GRS (1986)
Rheinland-Pfalz, FRG	3.5 - 14.5	25	GRS (1986)
Baden-Wurtemberg, FRG	5.5 - 15.5, 26.5 - 2.6	57	GRS (1986)
Federal Republic of Germany	30.4 - 23.5	140	GRS (1986)
Bavaria, FRG	1.5 - 20.5	210	GRS (1986)

[a]Values reported in fresh weight have been converted to dry weight assuming 20% dry matter for pasture vegetation.

Table 2.3-8. Time-integrated concentrations of ^{137}Cs in grass

Location	Observation period	Integrated grass concentration (kBq d/m^2)	References
Chester, N.J., United States	5.5 - 6.6	0.092	Dreicer et al. (1986)
Mol, Belgium	13.5 - 26.5	1.8	CEN/SCK (1986)
Geel, Belgium	13.5 - 30.5	2.2	CEN/SCK (1986)
Netherlands	3.5 - 29.5	2.5	Blaauboer (1986)

those for the European locations (Table 2.3-9). In Europe, the time-integrated concentrations vary by more than a factor of 10. Like the time-integrated concentrations in grass, the time-integrated concentrations in milk can be expected to increase as the period of observation increases.

Concentration ratios

Normalization of the time-integrated ^{137}Cs concentrations in grass (Table 2.3-7) by those in air (Table 2.3-6) yields grass-to-air concentration ratios, $CR_{grass/air}$ (Table 2.3-10). The two- and three-order-of-magnitude differences between the maximum and minimum time-integrated concentrations have been reduced to a difference of about a factor of six between the maximum and minimum values of $CR_{grass/air}$. It is interesting that Chester, New Jersey, is associated with the highest value of the grass-to-air ratio.

Normalization of the time-integrated ^{137}Cs concentrations in milk (Table 2.3-9) by those in air (Table 2.3-6) yields milk-to-air concentration ratios, $CR_{milk/air}$ (Table 2.3-11). Again, two- and three-order-of-magnitude differences between the maximum and minimum of the time-integrated concentrations have been reduced to a difference of a factor of 16 between the maximum and minimum values of $CR_{milk/air}$. It is interesting that Chester, New Jersey, is associated with the highest milk-to-air ratio.

Normalization of the time-integrated ^{137}Cs concentrations in milk (Table 2 3-9) by those in grass (Table 2.3-7) yields milk-to-grass concentration ratios, $CR_{milk/grass}$ (Table 2.3-12). The milk-to-grass ratios are more numerous than the grass-to-air and milk-to-air ratios. The large differences within the time-integrated concentrations in grass and milk have been reduced so that the maximum and minimum $CR_{milk/veg}$ values differ by only a factor of 10. It is interesting that Chester, New Jersey, is associated with a mid-range milk-to-grass ratio; both higher and lower values were determined for various European locations.

Table 2.3-9. Time-integrated concentrations of ^{137}Cs in cow's milk

Location	Observation period	Integrated milk concentration (kBq d/L)	References
New York, N.Y., United States (US)	6.5 - 12.6	0.0074	Klusek et al. (1986)
Chester, N.J., US	5.5 - 6.6	0.0097	Klusek et al. (1986)
Nordrhein-Westfalen, Federal Republic of Germany (FRG)	6.5 - 16.5	0.15	GRS (1986)
Rheinland-Pfalz, FRG	3.5 - 14.5	0.19	GRS (1986)
Groningen, Netherlands	8.5 - 20.5	0.30	Blaauboer (1986)
Mol, Belgium	13.5 - 26.5	0.34	CEN/SCK (1986)
Netherlands	3.5 - 29.5	0.36	Blaauboer (1986)
Geel, Belgium	13.5 - 30.5	0.37	CEN/SCK (1986)
Hamburg, FRG	3.5 - 26.5	0.45	GRS (1986)
Mol, Belgium	4.5 - 20.5	0.52	CEN/SCK (1986)
Baden-Wurtemberg, FRG	5.5 - 15.5, 26.5 - 2.6	0.63	GRS (1986)
Niedersachsen, FRG	5.5 - 17.5	0.84	GRS (1986)
Munich, FRG	30.4 - 8.5	1.1	Koehler (1986)
FRG	30.4 - 23.5	1.2	GRS (1986)
Bavaria, FRG	1.5 - 20.5	1.9	GRS (1986)

Table 2.3-10. Grass-to-air concentration ratios for ^{137}Cs

Location	Observation period	$CR_{grass/air}$ [m^3/kg (dry)]
Federal Republic of Germany (FRG)	29.4 - 2.6	1,700
Rheinland-Pfalz, FRG	1.5 - 14.5	2,600
Nordrhein-Westfalen, FRG	1.5 - 16.5	3,200
Bavaria, FRG	29.4 - 9.5	3,340
Baden-Wurtemberg, FRG	29.4 - 15.5	5,300
Chester, N.J., United States (US)	5.5 - 3.6	11,000
Geometric means		3,740
Geometric s.d.		1.90

Table 2.3-11. Milk-to-air concentration ratios for ^{137}Cs

Location	Observation period	$CR_{milk/air}$ (m^3/L)
Federal Republic of Germany (FRG)	30.4 - 15.5	10.0
Nordrhein-Westfalen, FRG	2.5 - 7.5	14.0
Bavaria, FRG	30.4 - 9.5	18.0
Baden-Wurtemberg, FRG	29.4 - 15.5	33.0
Munich, FRG	30.4 - 8.5	95.0
New York, N.Y., United States (US)	6.5 - 12.6	130.0
Chester, N.J., US	5.5 - 3.6	160.0
Geometric means		40.0
Geometric s.d.		3.12

Table 2.3-12. Milk-to-grass concentration ratios for ^{137}Cs

Location	Observation period	$CR_{milk/grass}$ [kg (dry)/L]
Nordrhein-Westfalen, Federal Republic of Germany (FRG)	6.5 - 16.5	0.0059
Rheinland-Pfalz, FRG	3.5 - 14.5	0.0076
Bavaria, FRG	1.5 - 20.5	0.0092
FRG	30.4 - 23.5	0.010
Baden-Wurtemberg, FRG	5.5 - 15.5, 26.5 - 2.6	0.011
Chester, N.J., United States (US)	5.5 - 6.6	0.017
Hamburg, FRG	3.5 - 26.5	0.032
Mol, Belgium	4.5 - 20.5	0.039
Niedersachsen, FRG	5.5 - 17.5	0.050
Groningen, Netherlands	8.5 - 20.5	0.058
Geometric means		0.0176
Geometric s.d.		2.32

Model predictions

The concentration ratios $CR_{grass/air}$, $CR_{milk/air}$, and $CR_{milk/grass}$ predicted by the IAEA model are summarized in Table 2.3-13. The predictions are based on the example parameter values listed in IAEA Safety Series No. 57 (IAEA 1982). Remember that the IAEA model was designed not to underpredict actual doses to critical population groups by more than an order of magnitude and thus is intentionally conservatively biased. The concentration ratios calculated for this analysis are based for the most part on concentrations integrated over periods of 2 to 4 weeks, so concentration ratios predicted by the model at 3 weeks (21 d) have been chosen as the values predicted by the model.

Comparison of predictions and observations

The comparison of ^{137}Cs concentration ratios predicted by the IAEA model with those estimated from measurement of Chernobyl fallout is presented in Table 2.3-14. The comparison is presented in the form of ratios of predictions to observations (P/O).

For the most part, values of P/O exceed unity by a factor ranging from about 5 to 50, which means that the model greatly overpredicts observations. P/O values for the milk-to-grass concentration ratios for a few of the locations vary from 0.9 to 1.6, which indicates that predictions and observations are in very good agreement. These differences (and similarities) between the predicted and observed CR values are discussed below.

2.3.2.3 Discussion

This section examines the differences between predicted and observed concentration ratios and discusses the appropriateness of model parameter values in an attempt to explain these differences.

Table 2.3-13. Concentration ratios predicted by the IAEA Safety Series No. 57 model for the transfer of ^{137}Cs over the air-pasture-cow-milk pathway

Pathway	Concentration ratio	Predicted value[a]
Air to grass	$CR_{grass/air}$	45,000 m^3/kg (dry)
Air to milk	$CR_{milk/air}$	2,300 m^3/L
Grass to milk	$CR_{milk/grass}$	0.051 kg (dry)/L

[a]The concentration ratios are based on concentrations predicted at t = 21 d.

Table 2.3-14. Predicted to observed ratios for the grass-to-air, milk-to-air, and milk-to-grass concentration ratios

Location	Predicted to observed ratio		
	$CR_{grass/air}$	$CR_{milk/air}$	$CR_{milk/grass}$
Chester, N.J., United States (US)	4.1	14	3.0
Federal Republic of Germany (FRG)	26	230	5.1
Rheinland-Pfalz, FRG	17		6.7
Nordrhein-Westfalen, FRG	14	160	8.6
Bavaria, FRG	13	130	5.5
Baden-Wurtemberg, FRG	8.5	70	4.6
Munich, FRG		24	
New York, N.Y., US		18	
Hamburg, FRG			1.6
Mol, Belgium			1.3
Niedersachsen, FRG			1.0
Groningen, Netherlands			0.88
Geometric means			11.9
Geometric s.d.			1.89
Geometric means			57.3
Geometric s.d.			3.12
Geometric means			2.89
Geometric s.d.			2.32

Differences between predicted and observed $CR_{grass/air}$

It should not be surprising that CR values predicted by the IAEA model exceed the observed CR values because the model was intentionally designed with a conservative bias. However, it is surprising that $CR_{grass/air}$ was overestimated to such an extent, with P/O values ranging from 4 to 26 (Table 2.3-14). In the current absence of site-specific parameter values, one can only speculate about how model parameters may be adjusted appropriately to calibrate the model so that predicted and observed concentration ratios are more agreeable.

By assuming no wet deposition, the values of $CR_{grass/air}$ would be lowered by a factor of 10. This reduction would not be justified, however, because wet deposition did occur at all the sites for which $CR_{grass/air}$ values are listed in Table 2.3-14 (Juzdan et al. 1986, BUNR 1986). Other parameters that could influence the predicted concentration of ^{137}Cs in vegetation are the weathering half life and the mass interception fraction. The weathering half-life is set at 14 d in the model calculations, but an enhanced precipitation rate is associated with enhanced weathering of radioactivity from plant surfaces. A weathering rate of about 8 d was measured for deposition on grass in Munich, FRG, which was attributed to both washoff by wind and rain and growth dilution (GSF 1986).

The mass interception fraction would also be expected to be reduced because of the influence of rain. The mass interception fraction is set at 2.0 m^2/kg (dry) in the model calculations. Indeed, values of about 0.4 m^2/kg (dry) were measured under strictly rainy conditions in Mol, Belgium (Zeevaert 1986). Values of about 1 m^2/kg (dry) can be estimated for Munich, FRG (GSF 1986). The combined effect of reduction in the weathering half-life and mass interception fraction under the influence of rainfall could then reduce the predicted grass-to-air concentration ratio by a factor of 2, 3, or even more. A variable degree of uncertainty has been introduced into the estimates of concentration ratio because of the incompleteness of the time integrals and the varying periods of observation.

Differences between predicted and observed $CR_{milk/grass}$

Predicted values of the milk-to-grass concentration ratio for ^{137}Cs exceeded the observed values by factors of 3 to 9 for six locations; predicted and observed concentration ratios were comparable within a factor of 2 at four locations.

Parameters that determine $CR_{milk/grass}$ include the rate of consumption of dry food by the cow, the fraction of daily feed that is fresh forage, and the milk transfer coefficient F_m. The dry feed consumption rate is set at 16 kg/d, and the fraction of dry feed that is represented by fresh forage is set at 0.4. Upward or downward adjustment of these parameters to conform to the practice at a specific site will increase or decrease the predicted values of $CR_{milk/grass}$.

The milk transfer coefficient F_m for ^{137}Cs has been at 8.0×10^{-3} d/L. Much lower F_m values of 2×10^{-3} to 3×10^{-3} d/L have been reported for ^{137}Cs in Chernobyl fallout in the FRG (Bonka 1982, Händl and Pfau 1986). Relatively low, but normal, F_m values of 2×10^{-3} to 3×10^{-3} d/L have been exhibited for ^{137}Cs in worldwide fallout and in diets rich in hay or silage that are high in potassium; higher F_m values ($\sim 1.0 \times 10^{-2}$ d/L) were observed for ^{137}Cs in a predominantly grain diet that was low in potassium (Ng et al. 1977). Accordingly, appropriate reductions in the reference F_m value of 8.0×10^{-3} d/L would lead to a reduction in the predicted milk-to-grass concentration ratio.

Differences between predicted and observed $CR_{milk/air}$

The milk-to-air concentration ratio may be regarded as a combination of the grass-to-air concentration ratio, $CR_{grass/air}$, and the milk-to-grass concentration ratio, $CR_{milk/grass}$. The P/O ratio for $CR_{milk/air}$ would be expected to be equal to the product of the P/O ratios for $CR_{milk/grass}$ and $CR_{grass/air}$. The above relationship is displayed exactly by the ^{131}I data in Table 2.3-3 but only approximately by the ^{137}Cs data in Table 2.3-14.

Because ^{137}Cs is long-lived, time-integrated concentrations of ^{137}Cs in environmental media integrated to infinity are not readily

forthcoming. The ^{137}Cs data are not so abundant as those for ^{131}I, and it is generally not possible to obtain complete data sets so that grass-to-air, milk-to-air, and milk-to-grass concentration ratios could be estimated over the same time interval. Besides the problems of overlapping time intervals and time intervals varying in duration, missing data points often had to be estimated. The data used in this analysis have been accepted at face value. Because they have not been verified, it is likely that some of the reported concentrations are in error.

It was generally not possible to extrapolate the available concentration data for ^{137}Cs because they would vary irregularly with time and would not display a clear pattern from which to estimate a rate constant for the anticipated decrease in concentration. This was the case for the data source for many of the locations in the Federal Republic of Germany, which was a data bank that was under development (GRS 1986). The incompleteness of the milk-concentration data for Rheinland-Pfalz, Nordrhein-Westfalen, Bavaria, Baden-Wurtemburg, and the Federal Republic of Germany, and the resulting underestimates of the time-integrated concentrations of ^{137}Cs concentrations in milk are at least a partial explanation of the high predicted-to-observed ratios of $CR_{milk/air}$ and $CR_{milk/grass}$ at these locations.

Implication of model testing for ^{137}Cs

The preceding examples that use Chernobyl fallout data for testing model predictions of ^{137}Cs transport are, of course, preliminary and have been presented for illustrative purposes only. As additional data become available on ^{137}Cs in environmental media and milk and other foodstuffs, they should lead to more complete characterizations of concentration vs time relationships and improved time-integrated concentrations for use in testing models.

Milk from goats and sheep and meat from various species of livestock and game are important contributors to the diet. It may be possible from data being reported on ^{137}Cs concentrations in various

animal products and environmental media to test and refine models that predict the transfer of ^{137}Cs to these products. It should also be possible from carefully planned series of measurements of ^{137}Cs and a knowledge of site-specific conditions to test dynamic models that predict the transfer of ^{137}Cs to foods. Testing could be performed both from the standpoint of the time history of the concentration and time-integrated concentrations.

2.3.2.4 Conclusions

Data on concentrations of ^{137}Cs from Chernobyl fallout in surface air, pasture grass and cow's milk were used in a preliminary attempt to test model predictions of the transfer of ^{137}Cs over the air-pasture-cow-milk pathway. The reference model selected for the analysis tended to overpredict the transfer of ^{137}Cs from air to pasture vegetation and from pasture vegetation to cow's milk. The extent of the overprediction is approximately one to two orders of magnitude for several locations in Europe and the United States.

Appropriate adjustments in parameter values of the model would lower the predicted concentrations and improve the agreement between predictions and observations. These include a decrease in the deposition velocity for periods of little or no rainfall and a decrease in the mass interception fraction and weathering half life for periods of wet deposition. The above changes would lower the predicte concentrations in pasture grass. A lowering of the milk transfer coefficient from the model default value would lower the predicted concentrations in milk.

It should be possible to improve and extend the analysis and better characterize the overall air-to-milk transfer of ^{137}Cs as additional data become available. It should also be possible to test and refine models that predict the transfer of ^{137}Cs from air and vegetation to other animal products such as milk from goats and sheep and meat from various species of livestock and game. The relative

importance of internal and external exposures following deposition of ^{137}Cs should be reevaluated as new information emerges regarding the transfer of ^{137}Cs to foodstuffs.

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2.4 ANALYSIS OF CHERNOBYL RADIONUCLIDES IN AQUATIC ECOSYSTEMS

2.4.1 Introduction

Available data from the Chernobyl accident indicate that the concentration of ^{137}Cs in freshwater fish reached higher levels than had been anticipated from atmospheric releases of radionuclides. These results suggest that the ingestion of radionuclides via aquatic food chain pathways may be more important than previously predicted and therefore should be included in radiological assessment models.

For atmospheric releases the process of dilution is generally accepted as being sufficient to reduce radionuclide concentrations in aquatic foods to levels where they will be less important than terrestrial food from the standpoint of exposure to humans. Radionuclides such as ^{137}Cs that are deposited on the surface of streams and lakes are rapidly diluted by the volume of water, whereas such radionuclides that fall on terrestrial environments are assumed to be retained on the landscape or removed over prolonged periods of time by weathering. Many of the radionuclides that enter aquatic ecosystems accumulate rapidly in sediments and eventually attain an equilibrium between sediments and the overlying water column, with most of the radionuclide being present in the sediment. These processes contribute further to the dilution of radionuclides in the water column.

Other reasons exist for assuming that terrestrial foods are more important than aquatic foods for human exposure via food-chain pathways. The process of bioaccumulation of radionuclides in aquatic food chains usually operates over a longer time scale than it does in terrestrial food chains, making it highly unlikely that short-lived radionuclides such as ^{131}I would be of radiological significance. In addition, with the exception of a few critical population subgroups, larger quantities of terrestrial derived foods are consumed by the average individual.

However, exceptions may occur where the ingestion of aquatic foods could contribute significantly to the exposure of humans consuming foods contaminated by fallout radionuclides. Relatively high concentrations of radionuclides may occur in aquatic biota in lakes that have a high surface-to-volume ratio and a slow rate of exchange. In addition, environmental conditions such as rainfall and snow melt could enhance the transport of radionuclides in runoff from terrestrial to the aquatic environments, resulting in relatively high concentrations of radionuclides in aquatic biota.

Preliminary results from Chernobly fallout radionuclides indicate that relatively high concentrations of ^{137}Cs were observed in freshwater fish in Finland, Sweden, England, and Bavaria. In this analysis, Chernobyl data will be used to compare the concentrations of ^{137}Cs in aquatic and terrestrial foods and to examine possible reasons for the relatively high concentrations in freshwater fish.

2.4.2 Approach

Early environmental monitoring of Chernobyl fallout stressed sampling of air, surface contamination, and terrestrial food products such as milk, beef, mutton, reindeer, fruits, berries, leafy vegetables, etc. Although a small amount of data was collected from marine and freshwater ecosystems (water, sediments, and biota), these data have received little attention. The available aquatic data are not comprehensive but can be used to determine whether they support generally accepted hypotheses on radionuclide transport and food chain pathways:

1. comparison of the concentration of Chernobyl fallout radionuclides in aquatic and terrestrial foods,
2. transport of ^{137}Cs from terrestrial ecosystems into aquatic ecosystems,
3. dilution of atmospheric releases of radionuclides in aquatic ecosystems, and
4. concentration factors for aquatic organisms.

Examples in this report rely on a limited number of reports and personal communications. The following literature provided most of the data cited in this section: a Ministry of Agriculture Fisheries and Food report on radioactivity in surface and coastal waters of the British Isles (Camplin et al. 1986), a publication by Peterson et al. (1986) showing levels of radioactivity in the aquatic environment in Sweden, and a Finnish report (STUK 1986) and an unpublished German report (Koehler 1986) that compare concentrations of radioactivity in terrestrial and aquatic foods.

2.4.3 Comparison of ^{137}Cs in Terrestrial and Aquatic Food Products

Cesium-137 accumulates in aquatic food chains and is considered the major long-term dose contributor through food chain pathways. Concentrations of ^{137}Cs in food products from these reports are compared in Table 2.4-1. A range of concentrations, when available, is given for each food; otherwise, maximum concentrations are listed. Most of the measurements included data extending through the first part of August 1986. Of considerable interest is the fact that freshwater fish had the highest concentration of ^{137}Cs of all food products in each country listed, except for Bavaria, where mushrooms were slightly higher.

The ingestion of radionuclides is dependent on the consumption rate, and in most cases it is relatively low for freshwater fish. Table 2.4-2 shows the consumption rates for the average Finnish population and the intake of ^{137}Cs for selected food products taken from the Finnish report (STUK 1986). The highest average intake of ^{137}Cs was from milk and milk products. However, if the highest concentrations observed in foods are used, the freshwater fish pathway has the potential for having the highest ingestion rate of ^{137}Cs, although the consumption rate is only 10 g/d for freshwater fish compared to 900 g/d for milk and milk products.

Another example that emphasizes the significance of the freshwater food chain pathway for the ingestion of radionuclides from Chernobyl fallout is shown in Table 2.4-3. The calculated doses were taken from a report by Camplin et al. (1986) and shows that, for a critical pathway analysis, the ingestion of ^{137}Cs is higher for freshwater fish than for other exposure pathways. Because this is a critical pathway analysis, the consumption rates (Table 2.4-3) are greater than they would be for the average population and therefore would probably overestimate the ingestion rates. In this analysis, time-integrated concentrations were used when available for such organisms as mollusc and seaweed. For other pathways (marine fish, crustaceans, and freshwater fish) a conservative assumption was made that the peak concentration observed was representative of the annual average.

The doses (committed dose equivalent) calculated by Camplin et al. (1986) for the continuous consumption for one year of different aquatic foods containing all measured Chernobyl fallout radionuclides are listed in Table 2.4-3. It was possible for Camplin et al. to calculate doses attributable to only the Chernobyl radionuclides because previous background information was available on radionuclide releases from nuclear facilities in the area. For the pathways considered in these examples, consumption of freshwater fish would be the most significant pathway for the ingestion of Chernobyl radionuclides.

2.4.4 Transport of Chernobyl Radionuclides from Terrestrial to Aquatic Ecosystems

In addition to direct deposition, radioactivity is transported by runoff from watersheds to surface waters. According to the Swedish report by Peterson et al. (1986), the timing and quantities of various radionuclides entering streams and lakes can be monitored by following the concentration of radionuclides in sludge from treatment plants. Given enough sampling points, the average transport and behavior of a particular radionuclide can be observed. In the sewage sludge from 47 plants in central Sweden, ^{137}Cs reached a maximum level of 16 kBq/kg 23 d after it was detected over Sweden. Iodine-131 had a

Table 2.4-1. Concentration of ^{137}Cs in foods produced in countries receiving Chernobyl fallout radionuclides (Bq/kg)

Food	Finland	Sweden	Bavaria	UK
Milk & milk products	20	8-80	300	360
Grains	10	>10	17-194	
Leafy vegetables	10		5-12	
Fruits and berries	30		47-300	
Mushrooms	20-6,700		30-2,200	
Beef	100	10-500	20-147	
Elk		20-1,600		
Venison		150-1,200		
Reindeer	500-8,000			
Aquatic birds				
Duck	10-6,900			
Canada goose		3,800		
Mallard duck		1,300		
Marine fish				
Baltic herring	30	23		
Atlantic codfish		2		170
Other fish	50			
Freshwater fish				
Brown trout		19,000		660
Rainbow trout		6,300		82
Perch	30-16,000	14,000		450
Pike	30-1,300	4,700		42
Crucian carp		1,900		
Other fish	50-9,300		13-1,100	
Shellfish		2,300		
Signal crayfish		2,280		
Winkles				800
Mussels				150
Shrimp			48	

Sources: Camplin et al. (1986), Fulker et al. (1986), Koehler (1986), Moberg (1986), Peterson et al. (1986), and STUK (1986).

Table 2.4-2. Estimated ingestion by the average population of ^{137}Cs from foods produced in the fallout area of Finland

Food	Consumption rate (g/d)	Intake of ^{137}Cs (Bq/d)
Milk and milk products	900	18
Grains	200	2
Fruits and berries	200	6
Beef	60	6
Marine fish	30	0.6
Freshwater fish	10	3-30

Source: STUK (1986).

Table 2.4-3. Individual doses from the continuous consumption for one year of aquatic foods exposed to Chernobyl fallout radionuclides in areas of highest observed concentrations in England

Pathway	Consumption ($g\ d^{-1}$)	Effective dose in one year[a] (mSv)
Marine fish	300	0.054
Freshwater fish	150	1.1
Crustaceans	50	0.0087
Mollusc	50	0.084
Seaweed	0	0.068

[a]Dose quantities are committed dose equivalents based on concentration data for Whitehaven cod, Ennerdale brown trout, Annon shrimp, Southerness winkles, and Seascale porphyria.

Source: Camplin et al. (1986).

similar behavior pattern and reached a peak concentration of 17 kBq/kg 24 d after the accident (Peterson et al. 1986).

The fact that both radionuclides had similar patterns indicates that ^{137}Cs was not tied up in surface soils as is generally assumed (Beasley and Jennings 1984) but was transported in runoff to surface waters. Environmental conditions existing in Sweden before the accident (i.e., soils tended to be saturated with snow melt and surface flow was common) apparently enhanced the transport of ^{137}Cs. Cesium-137 fallout was rapidly transported to surface streams before it could penetrate and be tied up in the soil. In addition, soils in these areas are generally thin layers overlying rocky substrata, which would tend to increase runoff.

2.4.5 Dilution of Atmospheric Releases of Chernobyl Fallout Radionuclides in Aquatic Ecosystems

2.4.5.1 Fresh Water

Atmospheric releases of radionuclides falling on surface waters are rapidly diluted in the volume of the body of water; however, available data from the Chernobyl accident indicate that ^{137}Cs was rapidly transported from the terrestrial environment to surface waters. This rapid transport to freshwater ecosystems would result in increased concentrations of radionuclides in the water and sediment and could account for the relatively high concentrations observed in aquatic organisms in these systems. Although radiological transport and dosimetry models do not consider the transport of radionuclides from the terrestrial watersheds to the aquatic ecosystems as being significant, data from the Chernobyl accident should provide an opportunity to test this parameter under different geochemical and climatic conditions.

2.4.5.2 Marine Systems

As was predicted, most data from the Chernobyl accident indicated that atmospherically deposited radionuclides were rapidly diluted in

the marine system because of the large volume of water. This hypothesis is supported by the low concentrations of radionuclides measured in seawater and marine organisms (Camplin et al. 1986, Aarkrog 1986, Peterson et al. 1986). However, in the littoral zones on the Solway coast of England, species of molluscs reached total concentrations of radionuclides on the order of 10,000 Bq/kg (Camplin et al. 1986). This was partly due to the propensity of molluscs to concentrate certain elements, but it also reflected water concentrations that were relatively high at the beginning of May but quickly declined. This was a very short-lived phenomenon involving radionuclides of short half-life (^{103}Ru, ^{129m}Te, and ^{131}I) in addition to ^{106}Ru, ^{134}Cs, and ^{137}Cs. Limited evidence indicates that the high concentrations on the coastline were correlated with precipitation and washout of radionuclides from the terrestrial system to the shallow littoral zone.

2.4.6 Concentration Factors

Aquatic models currently in use for dose assessment simulate the transport of radionuclides in aquatic environments and transfer to man. In these models, the assimilation of a radionuclide in aquatic biota is calculated by using a single empirical relationship to represent the transfer of the radionuclide from water to organism (Bq/kg wet wt organism divided by Bq/kg water).

The most significant radionuclide from the standpoint of long-term dose commitments from the Chernobyl fallout is ^{137}Cs. The concentration factor for ^{137}Cs in freshwater fish ranges from 100 in eutrophic systems to more than 14,000 in oligotrophic systems (Vanderploeg et al. 1975). According to Vanderploeg et al. (1975), the concentration factor for ^{137}Cs in fish is related to the potassium and suspended solid concentrations in water. Recommended concentration factors for dose assessment range from 400 to 2000 (Thompson et al. 1972, IAEA 1982) for freshwater fish.

The Chernobyl accident affords an opportunity to determine concentration factors for ^{137}Cs and several other radionuclides in

fish and other aquatic organisms from many different aquatic environments. For example, a concentration factor of 1200 can be calculated for brown trout in England, compared to 700 calculated for the same species in Sweden. Data available from Sweden (Camplin et al. 1986) indicate that the concentration of ^{137}Cs in predator fish species was still rising and had not leveled off after the first 130 d. Concentration factors should be based on the time-integrated concentration of the radionuclides in fish and water. Although data to determine time-integrated concentrations of radionuclides are not currently available, apparently they are available for a number of aquatic ecosystems.

2.4.7 Conclusions

The examples contained in this section indicate that the aquatic food chain pathway is of more importance from the standpoint of ingestion of Chernobyl fallout ^{137}Cs than had been previously assumed. Other radionuclides probably follow a similar pattern to that of cesium; however, further analysis of data is needed to identify the behavior of other nuclides.

In Finland, Sweden, and England at the beginning of August 1986, the concentration of ^{137}Cs from Chernobyl fallout was higher in freshwater fish than in any other food.

The relatively high concentrations of radioactivity observed in fish and other aquatic organisms appear to be the result of the rapid transport by runoff of ^{137}Cs and other radionuclides from the terrestrial environments into the aquatic ecosystems. For short-term atmospheric releases, contrary to the general assumption, ^{137}Cs does not necessarily remain tied up in the soils of terrestrial ecosystems for many half-lives but, depending on environmental conditions, may be rapidly transported by runoff to surface waters. Therefore, the concentration of radionuclides in surface waters may be enhanced.

2.4.8 Recommendations

Apparently, a considerable amount of information has been collected on Chernobyl fallout radionuclides in aquatic ecosystems; however, it has received little attention and is not readily available. The long-term collection and publication of data on aquatic ecosystems should be encouraged. Emphasis must be placed on collecting data on parameters that will permit the testing of models and generally accepted hypotheses, which for the most part have been developed for long-term releases of radionuclides in environments with different geochemical and climatic conditions. The following types of data would be particularly useful:

1. concentrations of radionuclides on terrestrial environments and in surface waters in areas that received the highest deposition of Chernobyl fallout radionuclides, and
2. time-integrated concentrations of radionuclides in water, sediment, and aquatic organisms.

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2.5 DOSIMETRY MODELS

Estimation of organ doses from inhaled and ingested radionuclides continues to be one of the more difficult problems encountered in assessing the radiological consequence of radioactivity releases to the environment because of the limited information regarding the behavior in the body of numerous radionuclides potentially released into the environment. For the most part, models to describe the movement of radionuclides within the body (biokinetic models) are empirical representations of the observed behavior in humans (generally workers) or an extrapolation of observations on laboratory animals. For a few radionuclides, the models reflect an in-depth synthesis of available information. However, limited opportunities occur by which one can obtain information in humans to validate these models.

Laboratories throughout the world have measured in various environmental media the radionuclides released during the Chernobyl nuclear power plant accident. The presence of gamma-emitting radionuclides within the body as a consequence of inhalation and ingestion of these radionuclides has also been confirmed. For the most part, the measurements were part of surveillance efforts to assess the extent and magnitude of potential exposures and thus are not generally based on an established sampling program. This is particularly true of *in vivo* measurements where the participation of individuals in the measurement program is generally a matter of convenience rather than selection through random sampling. Despite these difficulties, *in vivo* measurements of ^{131}I and ^{137}Cs can be useful in validating models of their behavior.

2.5.1 Comparison of Measured and Calculated ^{131}I Burdens

When taken into the human body, iodine concentrates in the thyroid gland, and measurements of ^{131}I in the gland provide the most reliable means of estimating dose. The mathematical description of the biokinetics of ^{131}I in the body used in radiation protection is that of Riggs as most recently implemented by the ICRP (1979). Thirty

percent of the iodine entering blood is assumed to translocate to the thyroid, where it is excreted as organic iodine, at a rate corresponding to a halftime of 120 d. The gland's excretion is assumed to be uniformly distributed among all body tissues and retained there with a biological half-life of 12 d. One-tenth of the organic iodine is assumed to go directly to fecal excretion, and the rest returns to blood as inorganic iodine. For ^{131}I with a halftime of only 8.04 d, the model is equivalent to assuming 30% of the ^{131}I entering blood deposits in the thyroid, where it is retained with an effective half time of 7.5 d.

To illustrate application of monitoring data to validation of the biokinetic model for iodine, we compare, in Table 2.5-1, observed and calculated burdens of ^{131}I in the thyroid. The observed thyroid burdens and ^{131}I levels in milk were taken from Hill et al. (1986). We assume milk is consumed at a rate of 0.3 L/d and that a delay of two days exists between the measured level in milk at the dairy and the consumption of milk contaminated at that level by an individual. We assumed that 22% (see Sect. 2.3.1.5) of the airborne ^{131}I in the passing cloud was particulate (integrated air concentration of particulate ^{131}I was estimated 1.6×10^{5} Bq•s•m^{-3} as estimated by Fry et al. 1986) and that inhalation of the cloud, as on May 2, resulted in the deposition of 180 Bq of ^{131}I into the blood of an adult. As seen in the Table 2.5-1, the data are consistent with the ICRP model for retention of ^{131}I in the thyroid; calculated values fall generally within a factor of three of the measured values. It appears, however, that the calculated values tend to overestimate the ^{131}I levels in the thyroid. Analysis of additional measurements are needed to confirm this observation. Uncertainties in the integrated air concentration of ^{131}I arising from questions regarding speciation, as well as the relationship between iodine levels in indoor and outdoor air, may be limiting factors in the analyses.

With the assumed input into the body, the model results contained in Table 2.5-1 indicate that inhalation of iodine during the passage of the plume was the major contributor to the thyroid content of ^{131}I. Only after May 22, some three weeks after the cloud passage, did ingestion become the major contributor to the thyroid burden of ^{131}I.

Table 2.5-1. Comparison of measured and calculated thyroid burdens

Date	Thyroid burden[1] (Bq) Measured	Calculated	Percent by inhalation	Calculated-to-measured ratio	
May 10	8	27.5	92.6	3.4	
	15	27.5	92.6	1.8	2.03
	17	27.5	92.6	1.6	
	21	27.5	92.6	1.3	
May 11	8	27.7	83.8	3.5	
	17	27.7	83.8	1.6	2.0
	27	27.7	83.8	1.0	
May 14	20	27.0	65.0	1.3	1.3
May 15	30	25.9	61.6	0.9	0.9
	30	25.9	61.6	0.9	
May 16	10	24.5	59.2	2.4	2.2
	13	24.5	59.2	1.9	
May 17	23	23.1	57.3	1.0	1.0
May 19	7	20.3	54.2	2.9	
	13	20.3	54.2	1.6	1.7
	33	20.3	54.2	0.6	
May 20	23	19.3	52.6	0.8	0.8
May 21	6	17.8	51.1	3.0	
	8	17.8	51.1	2.2	2.0
	20	17.8	51.1	0.9	
May 22	15	16.7	49.8	1.1	0.8
	26	16.7	49.8	0.6	

[1]The 22 measurements tabulated herein were performed in eight subjects ranging in age from 26 to 60 (Hill et al. 1986).

2.5.2 Radiocesium ^{137}Cs

Available information indicates that cesium is distributed rather uniformly throughout the body and that its retention can be represented by a two-exponential expression, one-tenth being retained with a half-life of 2 d and the remainder with a half-life of 110d (ICRP 1979). A considerable body of information on the behavior of ^{137}Cs in the environment and within the body has been assembled as a result of its presence in the environment as a component of the fallout from weapons testing (see, for example, Richmond and Furchner 1967 and UNSCEAR 1982). Using this information, UNSCEAR (1982) derived empirical functions to represent the transfer of ^{137}Cs from fallout into man's diet and his body. Because of the short biological half-life of cesium in the body, levels in the body tend to follow rather closely the dietary levels. UNSCEAR defines a factor representing the dose commitment per unit deposition, namely 5.5×10^{-8} Sv/(Bq•m^{-2}). Observed body burdens of fallout ^{137}Cs indicate that this factor ranges from 3.6 to 14.9 x 10^{-8} Sv/(Bq•m^{-2}). The experience from weapons fallout indicates that the major route of irradiation for the population is external irradiation from deposited ^{137}Cs for which UNSCEAR suggests the dose commitment is 12×10^{-8} Sv/(Bq•m^{-2}); about twice that for ^{137}Cs in the diet.

Although *in vivo* measurements of ^{137}Cs in man following the Chernobyl accident have been performed worldwide, only limited data have appeared in the literature. Unlike the radioiodines, where attention can be restricted to only two major environmental pathways (inhalation during cloud passage and the ingestion of milk), for radiocesium many items in the diet are of concern in estimating intake. The lack of complete data sets hampers attempts to compare measured and estimated cesium levels in the body.

The August 1986 IAEA Post-Accident Review Meeting on the Chernobyl Accident indicated some difficulties in assessing the dose contribution for the ingestion pathway. The Soviets used a model, reportedly based on the UNSCEAR model, to calculate the collective dose to a population of 75 million individuals. The model was adjusted, in some manner, to reflect local conditions regarding transfer of cesium to vegetation.

In applying the model, the Soviets estimated a collective dose to the above population of 2.1 million Sv through ingestion and 0.29 million Sv through external irradiation. The reported relationship between these two components of the collective dose is inverted from what one would expect based on weapons fallout observations, and thus the relationship was of concern to the experts assembled at the meeting. In further discussions, the Soviets indicated that rather extensive in vivo measurements suggested that 97% of the individuals measured had levels less than that predicted by the model (0.048 MBq); the measured values were suggested to be log-normally distributed with a median of 0.0048 MBq and a geometric standard deviation of 2.7. A geometric standard deviation of this magnitude for ^{137}Cs suggests that variations among the dietary levels of the individuals must be contributing substantially to the observed variations. An in-depth analysis of the measured ^{137}Cs measurements is needed to determine the utility of current models in providing meaningful information immediately following the introduction of radiocesium into the environment.

2.5.3 Conclusions

Information from programs for monitoring the levels of radioactive contamination in the environment is useful in validating components of the dose assessment models. Although sampling of environmental media is the primary means of detecting the presence of radioactive contamination in the environment, it is, of course, the potential for incorporation into tissues of the body that one seeks to assess. Thus, detection and interpretation of levels of radioactivity in the body are of primary importance to decision-makers in determining potential countermeasures to limit population exposure.

The illustrative example of ^{131}I thyroid burdens was found to be consistent with the ICRP metabolic model. Note the significance of inhalation with regard to thyroid burden: levels observed within a week after the contamination event are more related to inhalation than to the ingestion of milk.

As levels of ^{131}I decreased with time, monitoring efforts were directed toward ^{137}Cs. An individual's dietary intake of ^{137}Cs is dependent on his preference for particular food items and the levels in these foods. During the first year following an acute release, the ^{137}Cs levels in food strongly reflect differences in the rate that Cs is moving in these pathways to man. At this time, insufficient data have been reported to test the reasonableness of current models to predict the ^{137}Cs levels in the body. Caution should be exercised in using the empirical functions of UNSCEAR in assessing the significance of single, isolated depositions of ^{137}Cs. Additional mathematical simulation models are needed to provide insight into the significance of accidental releases by placing increased importance on the prediction of contamination levels in environmental media and in man rather than on the current objective of simply estimating collective dose.

It is generally assumed that countermeasures would be effective in reducing radiation exposures to individuals following an accidental release. This assumes knowledge of the significant pathways of exposure and assumes that individuals have not, on their own, taken actions to reduce their exposure. The in vivo measurements data compiled on the Chernobyl accident should be analyzed in an attempt to understand the magnitude and significance of exposure as well as actions taken by individuals.

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3. DESCRIPTION AND ANALYSIS OF THE EPA DATA BASE ON CHERNOBYL FALLOUT IN THE UNITED STATES

3.1 PROCUREMENT OF DATA

Because such large-scale contamination resulted from the Chernobyl reactor accident, various organizations around the world developed computerized data bases to manage an overwhelming amount of information on environmental measurements. One of the repositories for such data was the U.S. Environmental Protection Agency (EPA) office in Montgomery, Alabama. This EPA office was primarily responsible for compiling data from the Environmental Radiation Ambient Monitoring System (ERAMS). The raw and sampling procedures are described in EPA's Environmental Radiation Data Report No. 46 (USEPA 1986). In addition, information received by EPA on environmental radioactivity measurements in other countries was also included in a separate "world" data base, which was maintained until mid-June 1986. The U.S. data were compiled until mid-July 1986. Access to these data was made possible through a telecommunications link between Oak Ridge National Laboratory (ORNL) and the EPA Montgomery office. This section of our report describes the maximum reported concentrations of ^{131}I in air, rain, and milk and the total deposition of ^{131}I in rain for the United States. The report also provides documentation of the procedures used to estimate time-integrated concentrations of ^{131}I in air and milk. Time-integrated concentrations are required for the eventual testing of predictive models and for the assessment of radiological risk to the U.S. population.

3.2 PROCESSING OF DATA

The U.S. data base received from EPA comprises nearly 7000 entries. Each entry contains the following information: location of the sample, the medium sampled (air, milk, or rain), the date of collection, the radionuclide detected, the value measured, the error of the measurement, and the units of the measurement (see Table 3.1.) The

Table 3.1. A sample of the ORNL-edited EPA data base for the United States

State and city	Medium	Date	Nuclide	Measure	Flag/error	Units
Colorado, Denver	Air	5/6/86	^{134}Cs	0.0002	±66%	pCi/m^3
Colorado, Denver	Air	5/6/86	Field estimate	10	LT[a]	pCi/m^3
Colorado, Denver	Air	5/6/86	Gross beta	0.37		pCi/m^3
Colorado, Denver	Air	5/6/86	^{131}I	0.0057	±10%	pCi/m^3
Ohio, Toledo	Air	5/23/86	Field estimate	10	LT[a]	pCi/m^3
Ohio, Toledo	Air	5/23/86	Gross beta	0.53		pCi/m^3
Ohio, Toledo	Air	5/23/86	^{131}I	0.081	±15%	pCi/m^3
Idaho, Boise	Milk	5/14/86	^{131}I	0	ND[b]	pCi/L
Idaho, Boise	Milk	5/20/86	^{131}I	97.6	±12%	pCi/L
Idaho, Boise	Milk	5/23/86	^{137}Cs	26.0	±49%	pCi/L
Idaho, Boise	Milk	5/23/86	^{131}I	101.8	±13%	pCi/L
Washington, Seattle	Milk	6/4/86	^{131}I	16.7	±55%	pCi/L
Washington, Seattle	Milk	6/10/86	^{137}Cs	34.4	±28%	pCi/L
Washington, Seattle	Milk	6/16/86	^{140}Ba	16.5	±72%	pCi/L
Washington, Seattle	Milk	6/16/86	^{137}Cs	3' 2	±31%	pCi/L
Kansas, Topeka	Rain	5/10/86	^{131}I	0	ND[b]	pCi/L
Kansas, Topeka	Rain	5/10/86	^{131}I	0	ND[b]	pCi/m^2
Kansas, Topeka	Rain	5/13/86	^{131}I	44	±43%	pCi/L
Kansas, Topeka	Rain	5/13/86	^{131}I	1140	±43%	pCi/m^2
Kansas, Topeka	Rain	5/15/86	^{131}I	57	±28%	pCi/L
Kansas, Topeka	Rain	5/15/86	^{131}I	290	±28%	pCi/m^2
South Dakota, Pierre	Rain	5/23/86	^{131}I	75	±31%	pCi/L
South Dakota, Pierre	Rain	5/23/86	^{131}I	2960	±31%	pCi/m^2
South Dakota, Pierre	Rain	5/26/86	^{131}I	33.9	±46%	pCi/L
South Dakota, Pierre	Rain	5/26/86	^{131}I	271	±46%	pCi/m^2

[a] LT = less than.

[b] ND = no detection.

units used in ORNL's U.S. data base are picocuries (pCi). The resulting tables and figures, however, will be presented in this report in standard metric (SI) units. Because of its size, the data base was partitioned into files specific to air, milk, or rain.

At ORNL, the EPA U.S. data base was enhanced with the following additional data:

1. Oak Ridge, Tennessee, data from the unpublished data sets of I. L. Larson, M. L. Frank, and J. S. Eldridge, which include particulate air, milk, and rain concentrations;
2. Hanford, Washington, data from an EPA mailing, which include particulate air and milk concentrations;
3. Data from the Environmental Measurements Laboratory in New York on the concentrations of ^{131}I in air and milk (Klusek et al. 1986; Leifer et al. 1986); and
4. EPA's world data base, which contains 16 Canadian stations reporting rain concentrations, 4 stations reporting milk concentrations, and 9 stations reporting air concentrations.

3.3 ANALYZING DATA

Close examination of the data indicated that five Chernobyl radionuclides, ^{140}Ba, ^{134}Cs, ^{137}Cs, ^{131}I, and ^{103}Ru were included. However, ^{140}Ba, ^{134}Cs, and ^{103}Ru had so few entries that they were not examined in any detail. Because of the abundance of data reported for ^{131}I and the interest in examining this isotope, only data reported for ^{131}I will be presented in this report. The environmental media analyzed were air, rain, and milk.

3.3.1 Procedures for Estimating Time-Integrated Concentrations of ^{131}I in Milk

To facilitate the use of these data for testing model predictions and for the eventual assessment of radiological exposures to the U.S. population, estimated time-integrated concentrations (ETICs) for ^{131}I in milk were obtained in the following manner:

1. All concentrations below the detection limit or with a "no detection" (ND) indicator were excluded from calculations. This reduced the data base from 728 to 188 observations representing 64 sites.
2. The maximum concentration (MC) occurring prior to June 1, 1986, was determined for each site. Maximum concentrations occurring after that date were not considered because of poor counting statistics and the long time since the arrival of the peak ^{131}I concentration in air.
3. The 64 stations were then analyzed relative to the following criteria:
   a. At least four dates must have reported concentrations above the detection limit.
   b. The first detectable concentration in milk must have been reported before May 20, 1986. This date was selected to ensure that the early arrival of Chernobyl fallout in milk was not missed by an individual monitoring station. This reduced the number of observations to 76 for 13 stations in which time-integrated concentration in milk could be calculated.
4. For each of the remaining 13 sites, the reported concentrations are scattered as discrete samples over time (see example, Table 3.2). To estimate a time-integrated concentration for a site, the total reporting time period for each site was divided into daily intervals. The concentration for each daily interval was then determined by interpolation between reported values. An example of this interpolation is demonstrated in Table 3.3.
5. The last reported concentration was multiplied by the effective mean residence time of ^{131}I on pasture vegetation, which is equal to 5.9 d (Miller and Hoffman 1983). This value is an estimate of the infinite time-integrated milk concentration occurring after the last sampling date. It is assumed that the rate of decrease in the concentration in milk is directly related to the rate of decrease of ^{131}I on vegetation. The effect of this extrapolation is presented as the last entry in Table 3.3.

Table 3.2. Reported concentrations of ^{131}I in milk from Boise, Idaho

Date	Concentration (pCi/L)
May 16, 1986	58.0
May 20, 1986	97.6
May 23, 1986	102
May 27, 1986	97.0
May 30, 1986	72.1
June 2, 1986	92.5
June 5, 1986	39.4

Table 3.3. Interpolated concentrations of ^{131}I in milk from Boise, Idaho

Date	Concentration (pCi/L)	Date	Concentration (pCi/L)
May 16, 1986	58.0[a]	May 27, 1986	97.0[a]
May 17, 1986	67.9	May 28, 1986	88.7
May 18, 1986	77.8	May 29, 1986	80.4
May 19, 1986	87.7	May 30, 1986	72.1[a]
May 20, 1986	97.6[a]	May 31, 1986	78.9
May 21, 1986	99.0	June 1, 1986	85.7
May 22, 1986	100	June 2, 1986	92.5[a]
May 23, 1986	102[a]	June 3, 1986	74.8
May 24, 1986	101	June 4, 1986	57.1
May 25, 1986	99.4	June 5, 1986	39.3
May 26, 1986	98.2		(232)[b]

[a]Reported value.

[b]This value is the product of the last reported value times the effective mean residence time of ^{131}I on vegetation (5.9d). This is an estimate of the infinite time-integrated concentration in milk from the last reported date of a detectable concentration.

6. The estimated time-integrated concentration (ETIC) for each of the 13 sites is equal to the sum of all the concentrations reported, interpolated, and extrapolated (Table 3.4.).

To estimate time-integrated concentrations for the remaining sites for which milk concentrations were reported, the following procedures were used:

7. A ratio of time-integrated concentration to maximum concentration was calculated for each of the 12 sites (Table 3.4).
8. A factor for estimating time-integrated concentrations from maximum reported values was then determined by calculating the average of these ratios. The resulting statistics were
   a. number of stations, 12;
   b. multiplication factor, 15.52 d;
   c. standard deviation, 4.38 d;
   d. standard error of mean, 1.26 d;
   e. 95% confidence limit, 13.04 and 18.0 d; and
   f. coefficient of variation, 28.23%.
9. The factor, 15.52 d, was then multiplied by the maximum concentration (Table 3.5) reported for each of the remaining 51 stations. Thus, an estimated time-integrated concentration (ETIC) was obtained for 64 reporting stations. All stations used in mapping the ETIC of ^{131}I in milk are listed in Table 3.6.

3.3.2 Procedures for Estimating the Time-Integrated Concentration of ^{131}I in Air

The following itemized list shows the steps used in calculating the ETIC of ^{131}I in air:

1. All concentrations below the detection limit were excluded from further calculations. This reduced the data base from 5015 to 266 observations representing 64 sites.
2. All sites with a first-detectable concentration reported after May 20, 1986, were also eliminated, thus reducing the number of sites to 46. This date was selected to ensure that the actual maximum air concentration occurring at a given station was not missed.

Table 3.4. Estimated time-integrated concentrations (ETIC), maximum concentrations (MC), and ETIC/MC ratios for ^{131}I in milk

State and city	Days	ETIC (Bq d/L)	MC (Bq/L)	ETIC/MC (d)
California, Sacramento	29	35.2	1.52	23.2
California, San Francisco	15	23.2	1.42	16.3
Idaho, Boise[a]	21	72.0	3.77	19.1
Maine, Portland	12	17.8	1.97	9.04
Montana, Helena	18	44.5	3.37	13.2
Nevada, Las Vegas	22	24.5	1.45	16.9
New York, New York	26	19.2	1.48	12.9
Oregon, Portland	15	23.9	1.69	14.2
Tennessee, Oak Ridge	33	14.1	1.59	8.87
Utah, Salt Lake City	30	50.9	2.38	21.4
Washington, Seattle	23	35.9	2.18	16.5
Washington, Spokane	28	73.1	5.03	14.5

[a]See Table 3.3 for details of interpolated concentrations.

Table 3.5. Maximum concentration of ^{131}I in milk

State and city	Maximum concentration (Bq/L)	State and city	Maximum concentration (Bq/L)
Alberta, Edmonton (Canada)	No detection (ND)	Nebraska, Lincoln	ND
Alaska, Anchorage	ND	Nebraska, Omaha	1.61[a]
Alaska, Palmer	ND	New Hampshire, Concord	0.810[a]
Alabama, Ashford	0.851[a]	New Hampshire, Manchester	0.855[a]
Alabama, Montgomery	0.559[a]	New Jersey, Trenton	0.888[a]
Arkansas, Little Rock	0.710[a]	New Mexico, Albuquerque	0.677[a]
Arizona, Phoenix	1.18[a]	Nevada, Las Vegas	1.45
California, Los Angeles	1.08[a]	New York, Buffalo	0.747[a]
California, Oakland	ND	New York, New York	1.48
California, Sacramento	1.52	New York, Syracuse	0.703[a]
California, San Francisco	1.42	Ohio, Cincinnati	1.19[a]
Colorado, Denver	0.662[a]	Ohio, Cleveland	0.777[a]
Connecticut, Hartford	1.96[a]	Ohio, Dayton	ND
DC, Washington	1.69[a]	Oklahoma, Oklahoma City	1.78
Delaware, Wilmington	ND	Ontario, Ottawa (Canada)	0.05[a]
Florida, Tampa	ND	Ontario, Toronto (Canada)	0.399[a]
Georgia, Atlanta	1.16[a]	Oregon, Portland	1.69
Hawaii, Hilo	1.89[a]	Pennsylvania, Harrisburg	ND
Hawaii, Honolulu	ND	Pennsylvania, Philadelphia	0.799[a]
Idaho, Boise	3.77	Pennsylvania, Pittsburgh	0.932[a]
Idaho, Idaho Falls	ND	Panama Canal, Cristobal	1.09[a]
Iowa, Des Moines	1.35[a]	Puerto Rico, San Juan	ND
Iowa, Iowa City	ND	Rhode Island, Providence	ND
Illinois, Chicago	0.873[a]	South Carolina, Charleston	0.966[a]
Indiana, Indianapolis	ND	South Dakota, Rapid City	2.58[a]
Kansas, Wichita	1.41[a]	Tennessee, Chattanooga	ND
Kentucky, Louisville	ND	Tennessee, Knoxville	0.792[a]
Louisiana, New Orleans	0.966[a]	Tennessee, Memphis	1.01[a]
Massachusetts, Boston	0.814[a]	Tennessee, Oak Ridge	1.59
Maryland, Baltimore	1.45[a]	Texas, Arlington	0.725[a]
Maine, Portland	1.97	Texas, Austin	ND
Michigan, Detroit	1.52[a]	Texas, Fort Worth	ND
Michigan, Grand Rapids	1.94[a]	Utah, Salt Lake City	2.38
Minnesota, Minneapolis	1.39[a]	Virginia, Norfolk	0.844[a]
Minnesota, Saint Paul	0.869[a]	Vermont, Burlington	0.966[a]
Missouri, Kansas City	0.925[a]	Vermont, Montpelier	ND
Missouri, Saint Louis	0.644[a]	Washington, Hanford	5.16[a]
Mississippi, Jackson	1.18[a]	Washington, Seattle	2.18
Montana, Helena	3.37	Washington, Spokane	5.03
New Brunswick, Fredericton (Canada)	0.599[a]	Wisconsin, Milwaukee	0.932[a]
North Carolina, Charlotte	0.788[a]	West Virginia, Charleston	0.810[a]
North Dakota, Minot	2.46[a]	Wyoming, Laramie	1.04[a]

[a]Values to be adjusted by the average ETIC/MC ratio, to estimate a time-integrated concentration in Table 3.6.

Table 3.6. Estimated time-integrated concentrations (ETIC) of ^{131}I in milk

State and city	ETIC (Bq•d•L^{-1})	State and city	ETIC (Bq•d•L^{-1})
Alberta, Edmonton (Canada)	No detection (ND)	Nebraska, Lincoln	ND
Alaska, Anchorage	ND	Nebraska, Omaha	12.9[b]
Alaska, Palmer	ND	New Hampshire, Concord	12.6[a]
Alabama, Ashford	13.2[a]	New Hampshire, Manchester	13.2[a]
Alabama, Montgomery	8.67[a]	New Jersey, Trenton	13.7[a]
Arkansas, Little Rock	11.0[a]	New Mexico, Albuquerque	10.5[a]
Arizona, Phoenix	18.3[a]	Nevada, Las Vegas	24.5
California, Los Angeles	16.8[a]	New York, Buffalo	11.5[a]
California, Oakland	ND	New York, New York	19.2
California, Sacramento	35.2	New York, Syracuse	10.9[a]
California, San Francisco	23.2	Ohio, Cincinnati	18.5[a]
Colorado, Denver	10.3[a]	Ohio, Cleveland	12.1[a]
Connecticut, Hartford	13.3[b]	Ohio, Dayton	ND
DC, Washington	26.2[a]	Oklahoma, Oklahoma City	15.5[b]
Delaware, Wilmington	ND	Ontario, Ottawa (Canada)	7.75[a]
Florida, Tampa	ND	Ontario, Toronto (Canada)	6.21[a]
Georgia, Atlanta	18.0[a]	Oregon, Portland	23.9
Hawaii, Hilo	29.4[a]	Pennsylvania, Harrisburg	ND
Hawaii, Honolulu	ND	Pennsylvania, Philadelphia	12.3[a]
Idaho, Boise	72.0	Pennsylvania, Pittsburgh	14.5[a]
Idaho, Idaho Falls	ND	Panama Canal, Cristobal	16.9[a]
Iowa, Des Moines	20.9[a]	Puerto Rico, San Juan	ND
Iowa, Iowa City	ND	Rhode Island, Providence	ND
Illinois, Chicago	13.5[a]	South Carolina, Charleston	14.9[a]
Indiana, Indianapolis	ND	South Dakota, Rapid City	39.9[a]
Kansas, Wichita	17.5[b]	Tennessee, Chattanooga	ND
Kentucky, Louisville	ND	Tennessee, Knoxville	12.2[a]
Louisiana, New Orleans	14.9[a]	Tennessee, Memphis	15.5[a]
Massachusetts, Boston	12.6[a]	Tennessee, Oak Ridge	14.1
Maryland, Baltimore	22.6[a]	Texas, Arlington	11.2[a]
Maine, Portland	17.8	Texas, Austin	ND
Michigan, Detroit	23.5[a]	Texas, Fort Worth	ND
Michigan, Grand Rapids	10.4[b]	Utah, Salt Lake City	50.9
Minnesota, Minneapolis	17.9[b]	Virginia, Norfolk	11.3[b]
Minnesota, Saint Paul	13.5[a]	Vermont, Burlington	14.9[a]
Missouri, Kansas City	14.4[a]	Vermont, Montpelier	ND
Missouri, Saint Louis	9.98[a]	Washington, Hanford	80.1[a]
Mississippi, Jackson	18.3[a]	Washington, Seattle	35.9
Montana, Helena	44.5	Washington, Spokane	73.1
New Brunswick, Fredericton (Canada)	9.31[a]	Wisconsin, Milwaukee	14.5[a]
North Carolina, Charlotte	12.2[a]	West Virginia, Charleston	12.6[a]
North Dakota, Minot	38.2[a]	Wyoming, Laramie	16.1[a]

[a]ETIC = maximum concentration multiplication factor (15.52 d).

[b]June maximum value was climated and substituted for the maximum value that occurred in May.

The main portion of the airborne radioactive material from Chernobyl should have passed all sites in the U.S. prior to this date.

3. The MC was determined for each site.
4. Sites reporting a frequency of detectable concentrations of less than 5 were not included in the initial calculations of time-integrated concentrations. This reduced the observations to 169 for 20 stations.
5. For each of the remaining 20 sites, the reported concentrations were scattered as discrete values over the time of the reporting period. One example is shown in Table 3.7.

Table 3.7. Reported concentrations of ^{131}I in particulate air for Helena, Montana

Date	Concentration (pCi/m^3)
May 10, 1986	0.650
May 12, 1986	0.410
May 21, 1986	0.061
June 2, 1986	0.009
June 3, 1986	0.006
June 4, 1986	0.008

6. The air samplers were operated continuously, but samples were collected only at intervals; air concentrations were reported as averages. The average value of a reporting period was then assigned to all previous days for which no value was reported. An example of this method is demonstrated in Table 3.8.
7. The time-integrated concentrations for each of the 20 sites (Table 3.9) are equal to the sum of all the concentrations (reported and interpolated).

Table 3.8. Interpolated concentrations of ^{131}I in particulate air for Helena, Montana

Date	Concentration (pCi/m^3)	Date	Concentration (pCi/m^3)
May 10, 1986	0.650[a]	May 23, 1986	0.009
May 11, 1986	0.410	May 24, 1986	0.009
May 12, 1986	0.410[a]	May 25, 1986	0.009
May 13, 1986	0.061	May 26, 1986	0.009
May 14, 1986	0.061	May 27, 1986	0.009
May 15, 1986	0.061	May 28, 1986	0.009
May 16, 1986	0.061	May 29, 1986	0.009
May 17, 1986	0.061	May 30, 1986	0.009
May 18, 1986	0.061	May 31, 1986	0.009
May 19, 1986	0.061	June 1, 1986	0.009
May 20, 1986	0.061	June 2, 1986	0.009[a]
May 21, 1986	0.061[a]	June 3, 1986	0.006[a]
May 22, 1986	0.009	June 4, 1986	0.008[a]

[a]Reported value.

Table 3.9. Estimated time-integrated concentrations (ETIC), maximum concentrations (MC), and ETIC/MC ratios for ^{131}I in particulate air

State and city	Days	ETIC ($Bq \cdot d \cdot m^{-3}$)	MC ($Bq \cdot m^{-3}$)	ETIC/MC (d)
Alabama, Montgomery	25	0.028	0.009	3.11
Arizona, Phoenix	29	0.327	0.059	5.54
California, Los Angeles	10	0.038	0.010	3.80
Colorado, Denver	17	0.109	0.010	10.9
Idaho, Boise	11	0.203	0.059	3.44
Idaho, Idaho Falls	21	0.183	0.028	6.54
Montana, Helena[a]	26	0.079	0.024	3.29
Nebraska, Lincoln	16	0.068	0.012	5.67
Nevada, Las Vegas	15	0.162	0.031	5.22
New York, New York	34	0.074	0.010	7.44
Ohio, Columbus	24	0.077	0.009	8.56
Ohio, Toledo	22	0.031	0.003	10.3
South Dakota, Pierre	21	0.093	0.018	5.17
Tennessee, Oak Ridge	28	0.022	0.002	11.0
Texas, El Paso	27	0.097	0.016	6.06
Washington, Spokane	11	0.160	0.044	3.64
Wyoming, Cheyenne	24	0.158	0.016	9.88
Manitoba, Winnipeg (Canada)	8	0.075	0.020	3.75
Ontario, Ottawa (Canada)	9	0.174	0.062	2.81
Ontario, Toronto (Canada)	7	0.104	0.050	2.108

[a]See Table 3.8 for detail of interpolated concentrations.

8. A ratio of ETIC to MC, found in Table 3.9, was calculated for each site.
9. A factor for estimating time-integrated concentrations from maximum reported values was then determined by calculating the average of these ratios. The resulting statistics on this factor (compiled for the ETIC/MC ratios in Table 3.9) are
   a. number of sites, 20;
   b. multiplication factor, 5.83 d;
   c. standard deviation, 2.75 d;
   d. standard error of mean, 0.614 d;
   e. 95% confidence limits, 4.62 and 7.04 d; and
   f. coefficient of variation, 47.07%.
10. The factor 5.83 d is then multiplied by the maximum concentration (Table 3.10) of each of the remaining stations, and an ETIC is determined for 46 reporting stations. All stations used in mapping the ETIC of ^{131}I in particulate air are listed in Table 3.11.

3.4 PROCEDURES DEVELOPED FOR GRAPHIC PRESENTATION OF THE DATA

To study the regional and national patterns in the data, it was necessary to apply spatial distribution algorithms that would interpolate from the sparse number of monitoring stations to create a continuous surface over the United States. This surface then had to be presented as a map, with the concentration variations readily apparent. The total process included the following steps:

1. Access the processed EPA U.S. data base.
2. Match the name, address, and zip code of each monitoring station with a file of latitude and longitude centroids.
3. Adapt and test interpolation algorithms, using sparse station locations to estimate concentration values for a gridded array of latitude-longitude cells covering the United States.
4. Calculate many contour levels over the surface to represent the spatial variation semicontinuously.

Table 3.10. Maximum concentrations of ^{131}I in particulate air

State and city	Maximum concentration (Bq/m^3)	State and city	Maximum concentration (Bq/m^3)
Alaska, Anchorage	0.007[a]	Nevada, Las Vegas	0.031
Alabama, Ashford	0.009[a]	New York, Albany[b]	0.0004
Alabama, Montgomery	0.009	New York, New York	0.010
Arizona, Phoenix	0.059	New York, Niagara Falls[b]	0.001
British Columbia, Vancouver (Canada)	0.100[a]	New York, Syracuse[b]	0.0004
California, Berkeley	0.031[a]	Ohio, Columbus	0.009
California, Los Angeles	0.010	Ohio, Painesville[b]	0.001
Colorado, Denver	0.010	Ohio, Toledo	0.003
Connecticut, Hartford[b]	0.0004	Ontario, Ottawa (Canada)	0.062
Delaware, Wilmington[b]	0.001	Ontario, Thunder Bay (Canada)	0.002[a]
Florida, Jacksonville	0.020[a]	Ontario, Toronto (Canada)	0.050
Iowa, Iowa City	0.007[a]	Ontario, Windsor (Canada)	0.005[a]
Idaho, Boise	0.059	Oregon, Portland	0.004[a]
Idaho, Idaho Falls	0.028	Pennsylvania, Pittsburgh[b]	0.0002
Indiana, Indianapolis	0.006[a]	Pennsylvania, Three Mile Island	
0.007[a]			
Kansas, Topeka	0.009[a]	Quebec, Quebec (Canada)	0.003[a]
Louisiana, New Orleans[b]	0.0001	Rhode Island, Providence	0.007[a]
Massachusetts, Lawrence[b]	0.001	South Carolina, Barnwell[b]	0.0003
Manitoba, Winnipeg (Canada)	0.020	South Carolina, Columbia	0.002[a]
Maine, Augusta[b]	0.001	South Dakota, Pierre	0.018
Michigan, Lansing[b]	0.003	Tennessee, Knoxville[b]	0.002
Minnesota, Minneapolis	0.007[a]	Tennessee, Nashville	0.002[a]
Missouri, Jefferson City[b]	0.003	Tennessee, Oak Ridge	0.002
Mississippi, Jackson[b]	0.001	Texas, El Paso	0.016
Montana, Helena	0.024	Utah, Salt Lake City	0.041[a]
New Brunswick, Fredericton (Canada)	0.020[a]	Virginia, Lynchburg[b]	0.001
North Carolina, Charlotte	0.006[a]	Virginia, Virginia Beach	0.0004
North Carolina, Goldsboro	0.006[a]	Washington, Hanford	0.050[a]
North Dakota, Bismarck	0.016[a]	Washington, Olympia	0.014[a]
Nebraska, Lincoln	0.012	Washington, Spokane	0.044
Newfoundland, St. John's (Canada)	0.050[a]	Wisconsin, Madison	0.004[a]
New Jersey, Trenton[b]	0.001	Wyoming, Cheyenne	0.016

[a]Values selected for estimating time-integrated concentrations in Table 3.11.

[b]Station eliminated from analysis because the date of the first detectable concentration was after May 20, 1986.

Table 3.11. Estimated time-integrated concentrations (ETIC) of ^{131}I in particulate air

State and city	ETIC ($Bq \cdot d \cdot m^{-3}$)	State and city	ETIC ($Bq \cdot d \cdot m^{-3}$)
Alaska, Anchorage	0.039[a]	Nevada, Las Vegas	0.162
Alabama, Ashford	0.054[a]	New York, New York	0.074
Alabama, Montgomery	0.028	Ohio, Columbus	0.077
Arizona, Phoenix	0.327	Ohio, Toledo	0.031
British Columbia, Vancouver (Canada)	0.583[a]	Ontario, Ottawa (Canada)	0.174
California, Berkeley	0.181[a]	Ontario, Thunder Bay (Canada)	0.001[a]
California, Los Angeles	0.038	Ontario, Toronto (Canada)	0.104
Colorado, Denver	0.109	Ontario, Windsor (Canada)	0.029[a]
Florida, Jacksonville	0.119[a]	Oregon, Portland	0.026[a]
Iowa, Iowa City	0.039[a]	Pennsylvania, Three Mile Island	0.039[a]
Idaho, Boise	0.203	Quebec, Quebec (Canada)	0.018[a]
Idaho, Idaho Falls	0.183	Rhode Island, Providence	0.043[a]
Indiana, Indianapolis	0.032[a]	South Carolina, Columbia	0.009[a]
Kansas, Topeka	0.054[a]	South Dakota, Pierre	0.093
Manitoba, Winnipeg (Canada)	0.075	Tennessee, Nashville	0.013[a]
Minnesota, Minneapolis	0.043[a]	Tennessee, Oak Ridge	0.022
Montana, Helena	0.079	Texas, El Paso	0.097
New Brunswick, Fredericton (Canada)	0.117[a]	Utah, Salt Lake City	0.237[a]
North Carolina, Charlotte	0.035[a]	Washington, Hanford	0.294[a]
North Carolina, Goldsboro	0.032[a]	Washington, Olympia	0.079[a]
North Dakota, Bismarck	0.093[a]	Washington, Spokane	0.160
Nebraska, Lincoln	0.068	Wisconsin, Madison	0.022[a]
Newfoundland, St. John's (Canada)	0.292[a]	Wyoming, Cheyenne	0.158

[a]ETIC = maximum concentration x multiplication factor (5.83 d).

5. Map the successive contour areas with gradually changing intent to show the spatial patterns, and note the specific estimated values at a given contour level.
6. Present the same data as a three-dimensional perspective surface with the changing color shades on top. The height of the surface corresponds to the concentration estimates.

Concentrations of ^{131}I were selected for graphing because this radionuclide was the one associated with the most monitoring stations, and the best distribution of concentrations was chosen for graphing. One of the most difficult parts of the mapping effort was to set up an interpolation algorithm that would create a consistent and reasonable concentration surface using input data points that were very sparse in some parts of the United States. The resulting surface is represented as a gridded array whose z axis (height) corresponds to the calculated values and whose x and y axes specify the geographic location of the cells (or mesh) on the earth's surface. Along the edges of a study region, extrapolation may be necessary because there may be no data points further out to aid in the estimation process.

The current interpolation approach estimates the concentration distribution by fitting a quadratic surface at each grid cell to all the input data points. The monitoring stations are weighted as a function of their distance from the grid cell in question so that the nearby control points have the greatest influence on the fit. Because this is a global technique, the surface is continuous and smooth, with the regional trends being portrayed fairly well. A two-step procedure is used first to calculate a moving polynomial function representing the surface and then to evaluate this function at the mesh points to estimate concentration values. The method of "least squares" is used to fit the polynomial function of the two independent variables (x and y) to the input values (z). This fit is performed at each mesh point, based on all input data points within the United States.

After the gridded surface is interpolated, contours are calculated for a set of specified concentration levels. The result for a given contour is represented as a polygonal boundary traveling across the landscape tracing out a constant concentration. The process is fully

automated, so it is easy to redefine concentration levels and to compute new contours. Final contour maps were produced from the quadratic fit. These maps were computed with many contour levels and plotted with a continuously varying color shade without the boundaries themselves being mapped. The following is a summary of the two-dimensional contour plots:

1. ETIC of ^{131}I in particulate air (Fig. 3.1),
2. ETIC of ^{131}I in milk (Fig. 3.2), and
3. total rain deposition of ^{131}I (Fig. 3.3).

Three-dimensional contour plots are included as follows:

1. ETIC of ^{131}I in particulate air (Fig. 3.4),
2. ETIC of ^{131}I in milk (Fig. 3.5), and
3. total rain deposition of ^{131}I (Fig. 3.6).

3.5 RESULTS

3.5.1 Particulate ^{131}I Concentration in Air

The ranges of MC of particulate ^{131}I in air (Table 3.10) show a high at Phoenix, Arizona, and Boise, Idaho, of 0.059 Bq/m^3, and a low of 0.0001 Bq/m^3 in New Orleans, Louisiana. The highest ETIC (Table 3.11) of 0.294 $Bq \cdot d \cdot m^{-3}$ was located at Hanford, Washington, and the lowest was 0.009 $Bq \cdot d \cdot m^{-3}$ at Columbia, South Carolina. The contour map of the ETICs (Fig. 3.1) displays a region of high values that stretches from Canada to Mexico through the western portion of the country. Except for the southern part of Florida, the ETIC steadily decreases to the east.

3.5.2 ^{131}I Concentrations in Milk

The ranges of maximum concentrations of ^{131}I in milk (Table 3.5) show a high of 5.16 Bq/L in Hanford, Washington, and a low of 0.559 in Montgomery, Alabama. The maximum value of the ETIC (Table 3.6) in milk of 80.1 $Bq \cdot d \cdot L^{-1}$ also appears at the Hanford station, with the minimum of 8.67 $Bq \cdot d \cdot L^{-1}$ again in Montgomery. Examination of the contour map

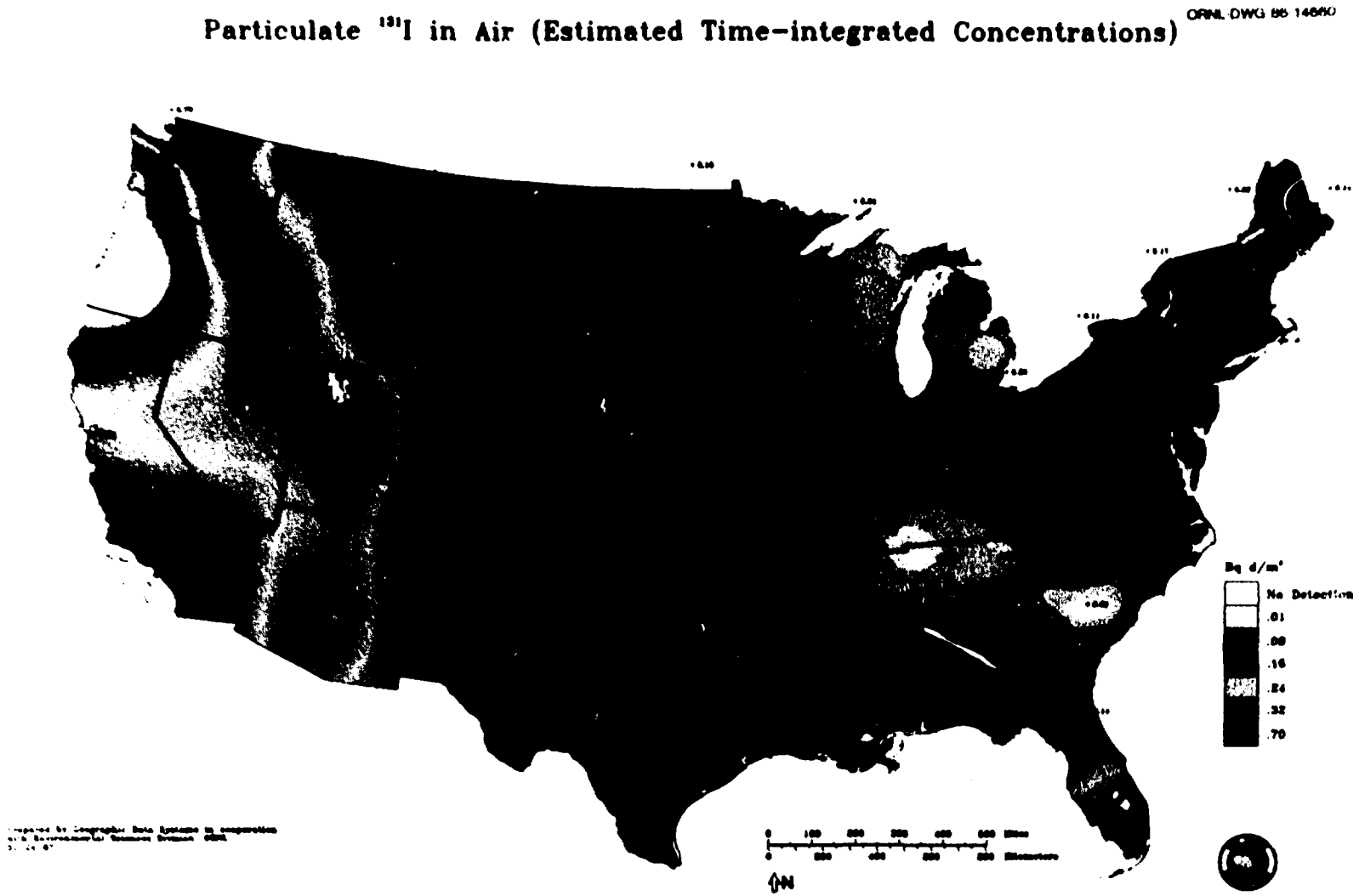

Fig. 3.1. Two-dimensional contcur plot of the estimated time-integrated concentrations of ^{131}I in particulate air.

^{131}I in Milk (Estimated Time--integrated Concentrations)

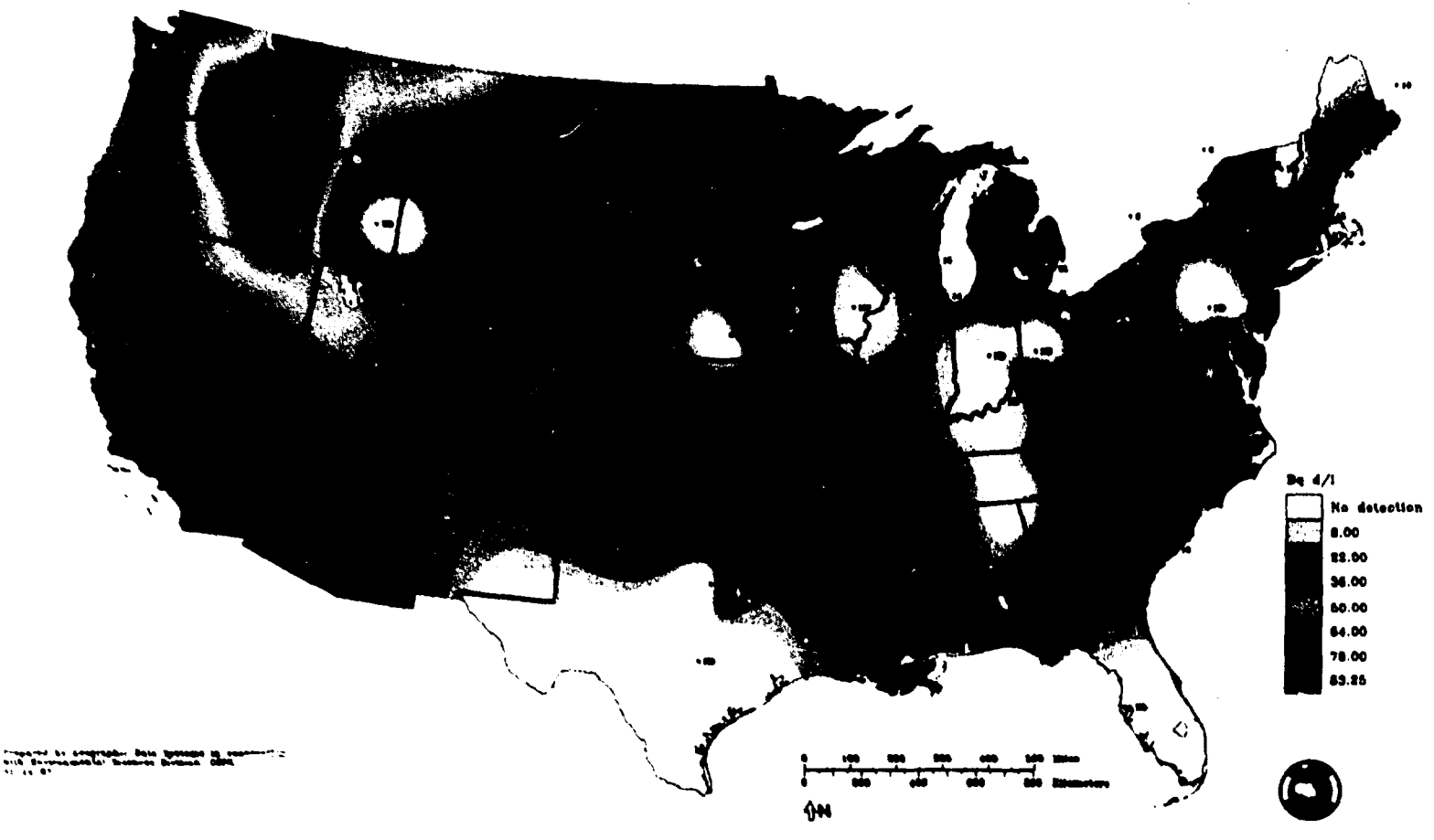

Fig. 3.2. Two-dimensional contour plot of the estimated time-integrated concentrations of ^{131}I in milk.

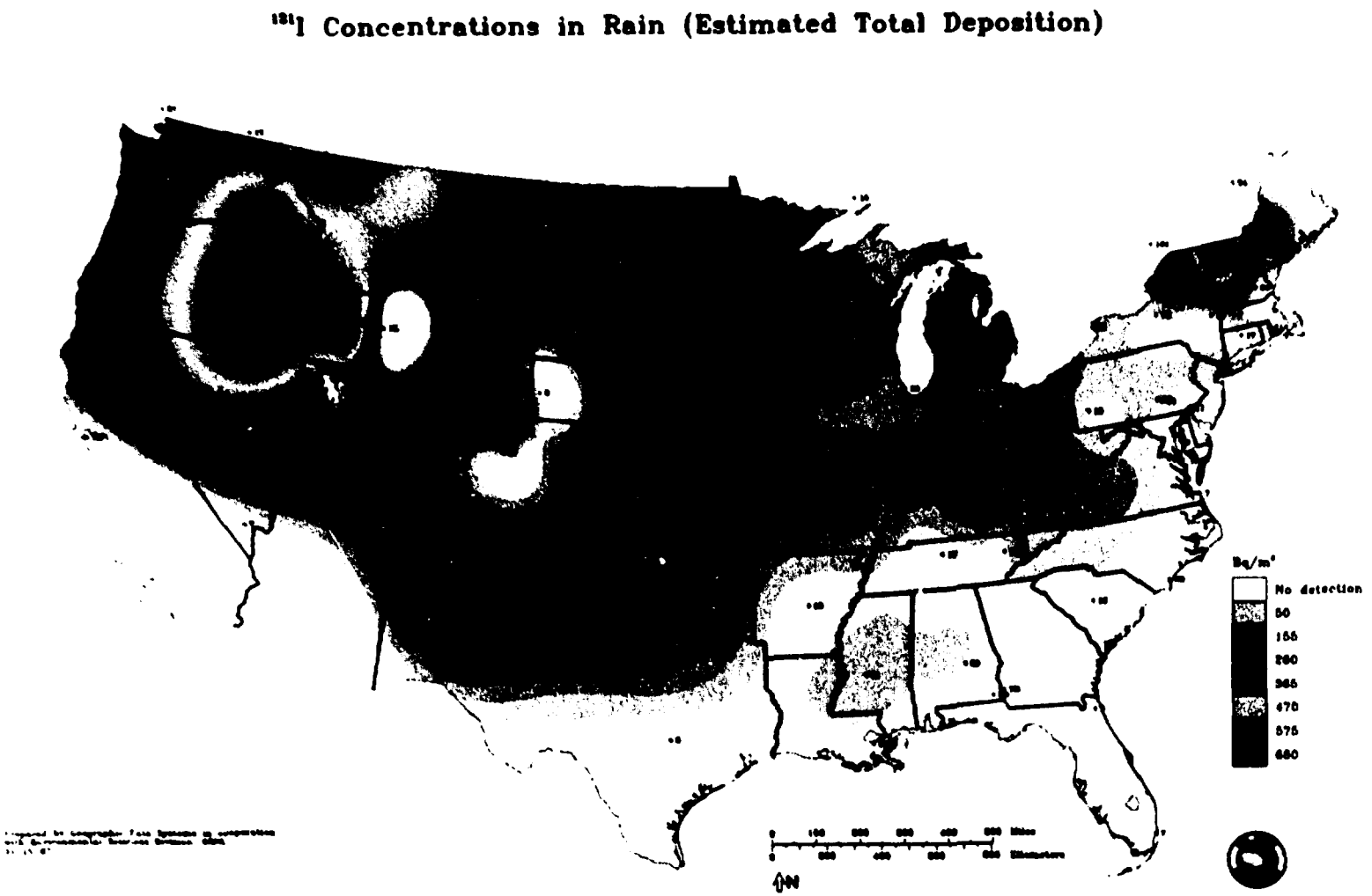

Fig. 3.3. Two-dimensional contour plot of total rain deposition of ^{131}I.

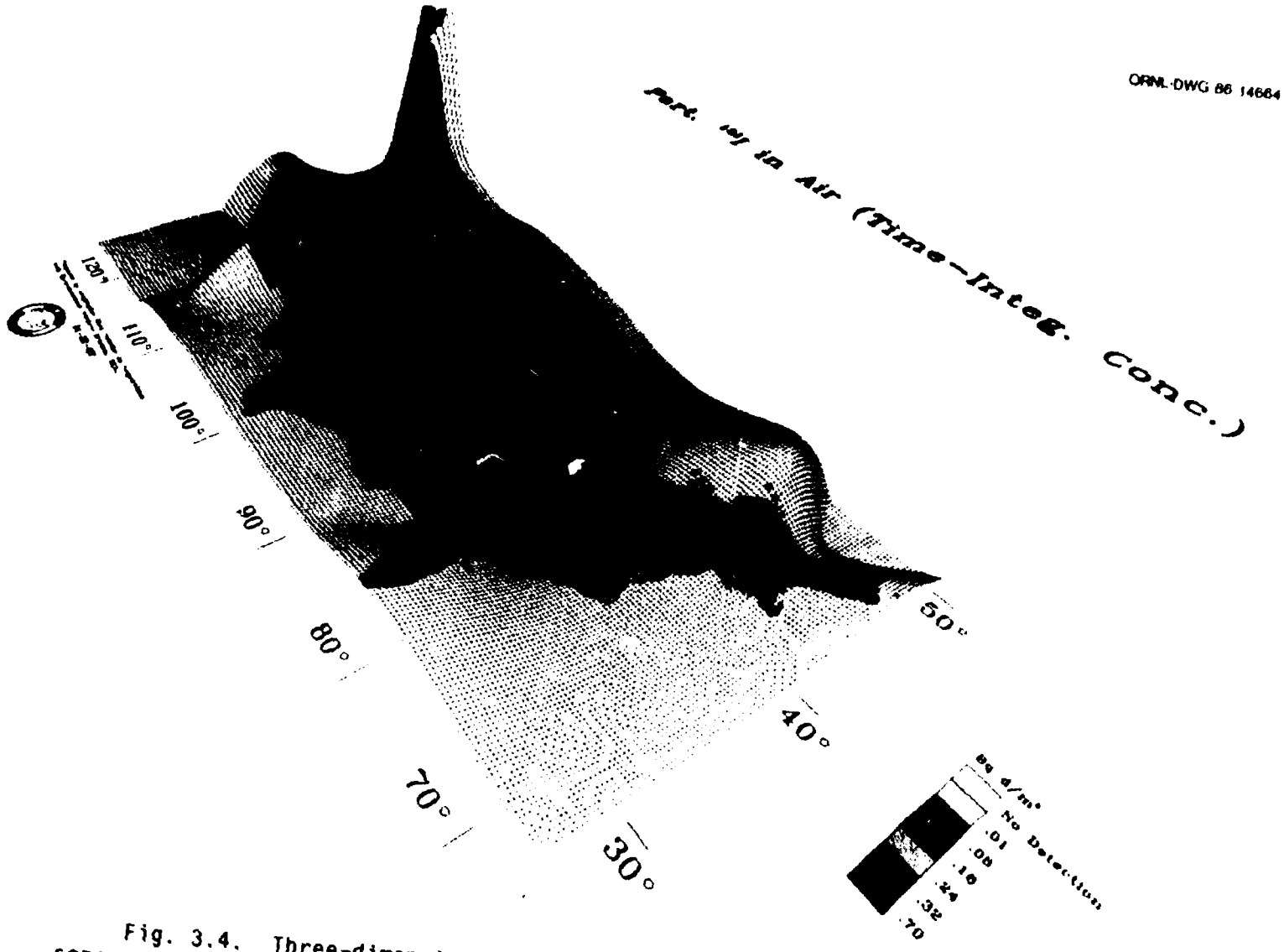

Fig. 3.4. Three-dimensional contour plot of the estimated time-integrated concentrations of ^{131}I in particulate air.

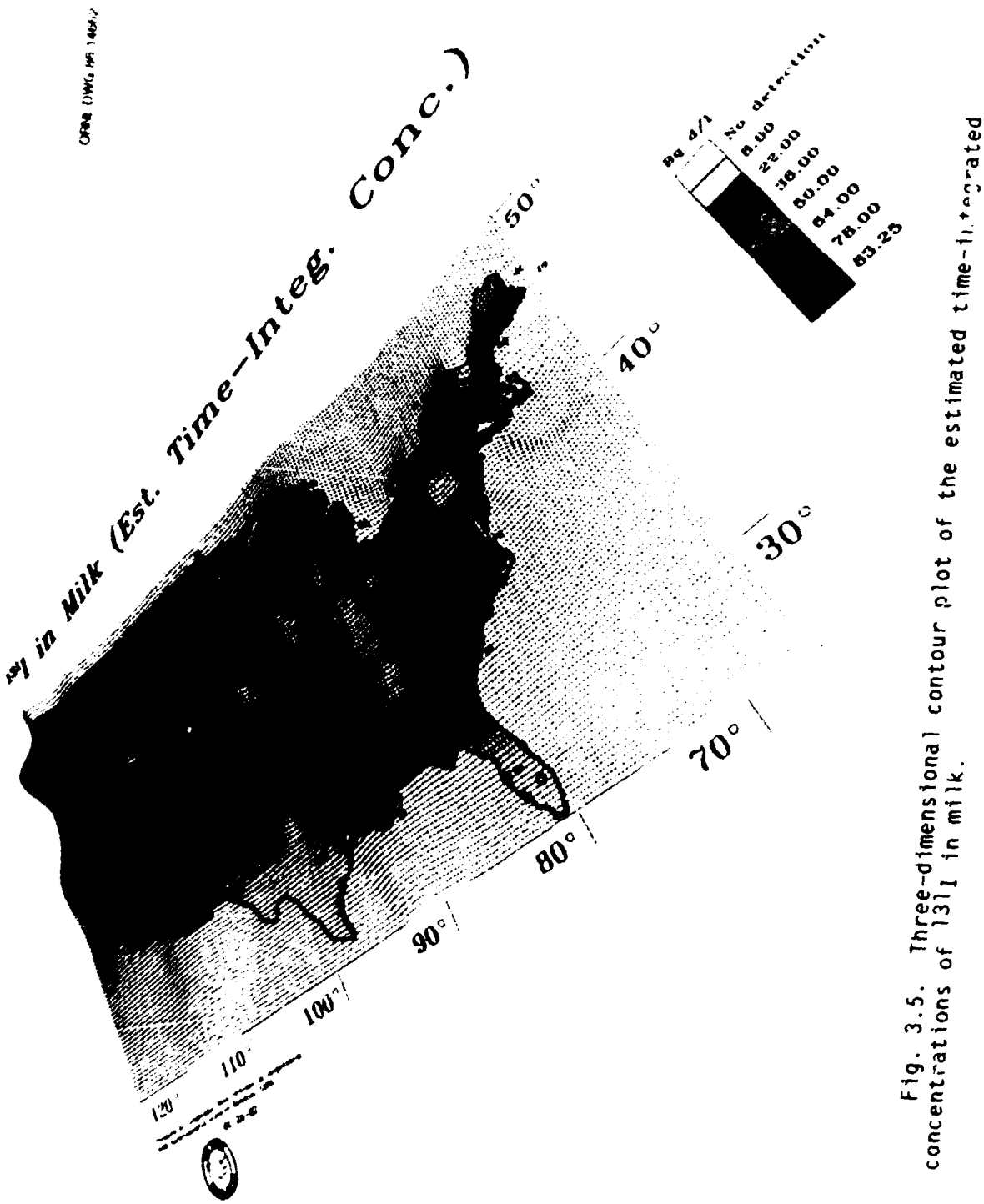

Fig. 3.5. Three-dimensional contour plot of the estimated time-integrated concentrations of ^{131}I in milk.

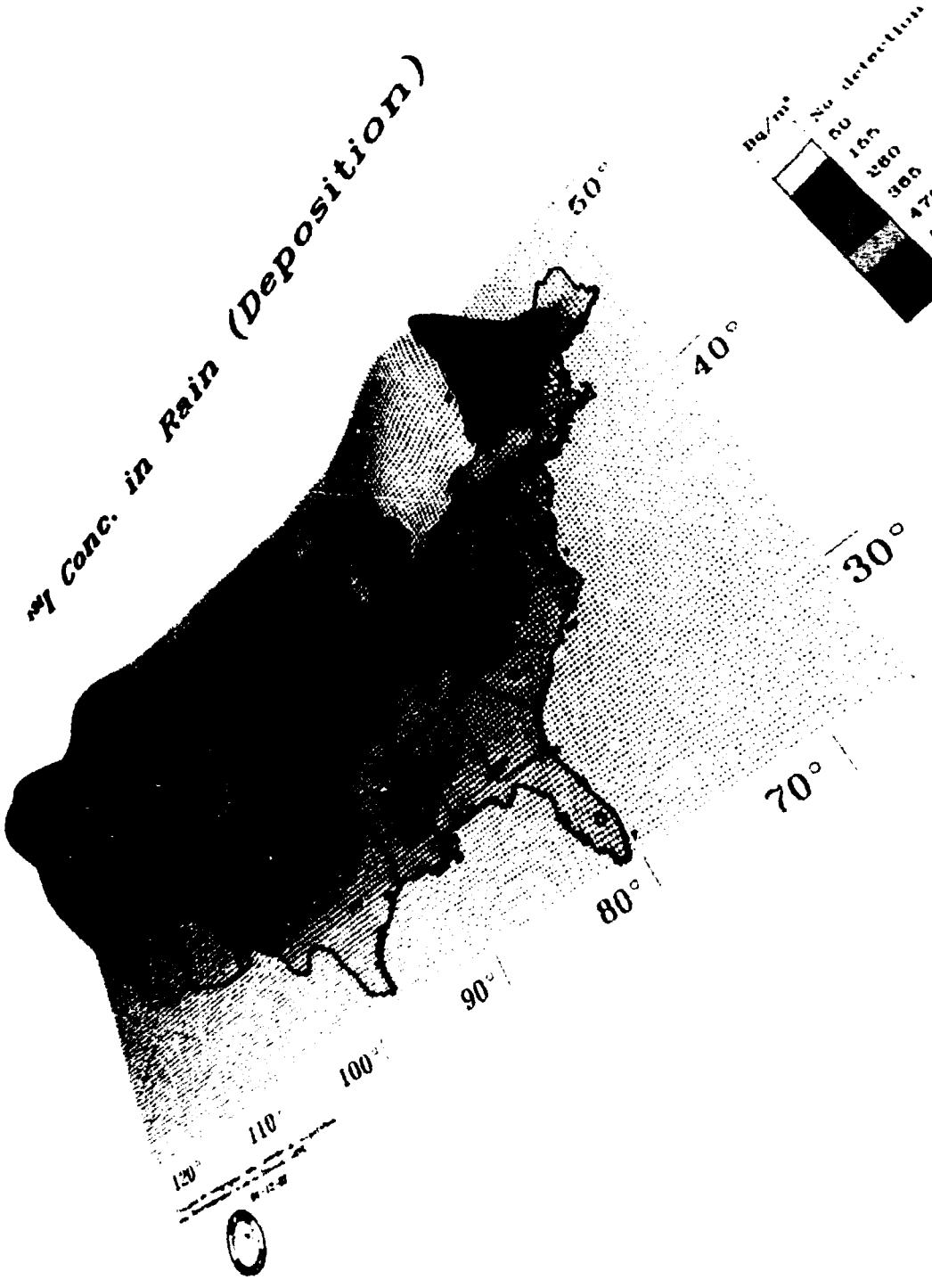

Fig. 3.6. Three dimensional contour plot of total rain deposition of ^{131}I.

for the ETICs in milk (Fig. 3.2) reveals a region of high values in the upper northwest third of the country. Except for a few pockets, the ETIC decreases toward the eastern and southern United States.

3.5.3 Total Deposition of ^{131}I in Rain

A maximum total deposition of ^{131}I (Table 3.12) in rain of 680 Bq/m^2 was recorded at Boise, Idaho, and a minimum of 1.33 Bq/m^2 was recorded at Yaphank, New York. The highest concentration of ^{131}I in rain deposition was located in the northwest part of the country and gradually decreased toward the southeast (Fig. 3.3). The maximum concentration of ^{131}I in rain is shown in Table 3.13.

3.5.4 Milk/Air Concentration Ratio for ^{131}I

To develop a data set amenable to potential model validation and the eventual estimation of radiological impact, ETICs in milk (measured as $Bq \cdot d \cdot L^{-1}$) were divided by ETICs derived for the total amount of ^{131}I in air ($Bq \cdot d \cdot m^{-3}$). The result was a concentration ratio of milk/air ($CR_{milk/air}$) with units of m^3/L. Within the EPA data, only 19 stations recorded both detectable milk and air ETICs, and 3 of these stations were located in Canada (Table 3.14). Note that for these ratios, the particulate air concentrations at all stations except New York were multiplied by 4.55 to estimate the total ^{131}I concentration (gaseous and particulate) in air. This factor was recommended by Snell et al. (1986) and is associated with a standard deviation of 1.5. The New York particulate ^{131}I air concentrations were multiplied by a factor of 2.33 to obtain the total amount of ^{131}I in air.

The $CR_{milk/air}$ (given in m^3/L) are summarized by the following statistics (as compiled from concentration ratios in Table 3.14):

1. number of ratios, 19;
2. geometric mean of ratios, 52.5 m^3/L;
3. geometric standard deviation, 2.51;

Table 3.12. Total deposition of ^{131}I in rain

State and city	Total deposition (Bq/m^2)	State and city	Total deposition (Bq/m^2)
Alaska, Anchorage	3.26	Nebraska, Scottsbluff	6.07
Alaska, Bethel-Mechin	No detection (ND)	Newfoundland, St. John's (Canada)	0.333
Alaska, Bethel-Morgan	ND	New Hampshire, Concord	29.8
Alaska, Bethel-Troat	ND	New Mexico, Santa Fe	111.
Alaska, Chefornak	ND	Nova Scotia, Halifax (Canada)	1.63
Alaska, Dutch Harbor	ND	NT, Hay River (Canada)	1.63
Alaska, Fairbanks	ND	Nevada, Las Vegas	6.66
Alaska, Juneau	29.9	New York, Albany	14.2
Alaska, Ketchikan	ND	New York, New York	28.1
Alaska, Kodiak	ND	New York, Niagara Falls	27.7
Alaska, Kotzebue	ND	New York, Syracuse	17.8
Alaska, Kwigillingok	ND	New York, Yaphank	1.33
Alaska, North Pole	ND	Ohio, Columbus	127.
Alabama, Dothan	ND	Ohio, Painesville	66.2
Alabama, Montgomery	19.6	Ohio, Toledo	35.9
Arkansas, Little Rock	10.4	Oklahoma, Oklahoma City	92.9
British Columbia, Greenwood (Canada)	.629	Ontario, Ottawa (Canada)	5.96
British Columbia, Vancouver (Canada)	2.48	Ontario, Thunder Bay (Canada)	0.481
California, Berkeley	ND	Oregon, Portland	265.
Colorado, Denver	26.6	Pennsylvania, Harrisburg	26.3
Connecticut, Hartford	17.4	Pennsylvania, Middletown	14.0
Delaware, Wilmington	10.7	Pennsylvania, Pittsburgh	18.5
Florida, Jacksonville	3.18	Quebec, Quebec (Canada)	1.26
Florida, Miami	6.66	Rhode Island, Providence	11.4
Georgia, Blakely	ND	South Carolina, Columbia	15.9
Hawaii, Honolulu	18.5	South Dakota, Pierre	120.
Iowa, Iowa City	35.6	Saskatchewan, Regina (Canada)	4.21
Idaho, Boise	680.	Saskatchewan, Saskatoon (Canada)	6.29
Idaho, Idaho Falls	520.	Tennessee, Knoxville	31.1
Illinois, Chicago	26.4	Tennessee, Nashville	12.2
Indiana, Indianapolis	46.1	Tennessee, Oak Ridge	13.5
Kansas, Topeka	97.1	Texas, Austin	5.53
Kentucky, Frankfort	68.5	Utah, Salt Lake City	217.
Louisiana, New Orleans	13.7	Virginia, Lynchburg	39.9
Maine, Augusta	62.4	Virginia, Virginia Beach	6.66
Michigan, Lansing	78.4	Vermont, Montpelier	589.
Minnesota, Minneapolis	138.	Washington, Olympia	269.
Missouri, Jefferson City	58.3	Washington, Spokane	437.
Mississippi, Jackson	31.1	Wisconsin, Madison	46.5
Montana, Helena	405.	West Virginia, Charleston	57.6
North Carolina, Wilmington	ND	Wyoming, Cheyenne	287
North Dakota, Bismarck	123.	Wyoming, Jackson	ND
Nebraska, Lincoln	75.6		

Table 3.13. Maximum concentration of ^{131}I in rain

State and city	Maximum concentration (Bq/L)	State and city	Maximum concentration (Bq/L)
Alberta, Calgary (Canada)	1.07	Montana, Helena	24.1
Alberta, Edmonton (Canada)	1.17	North Dakota, Bismarck	29.2
Alaska, Anchorage	13.7	Nebraska, Lincoln	30.7
Alaska, Bethel-Mechin	5.92	Nebraska, Scottsbluff	3.03
Alaska, Bethel-Morgan	6.29	Newfoundland, St. John's (Canada)	0.185
Alaska, Bethel-Troat	2.29	New Hampshire, Concord	5.92
Alaska, Chefornak	14.1	New Mexico, Santa Fe	13.3
Alaska, Dillingham	11.1	Nova Scotia, Halifax (Canada)	0.481
Alaska, Dutch Harbor	17.4	NT, Hay River (Canada)	0.222
Alaska, Fairbanks	50.3	NT, Inuvik (Canada)	0.592
Alaska, Ketchikan	12.6	New York, Albany	3.48
Alaska, Kodiak	7.40	New York, New York	7.40
Alaska, Kotzebue	No detection (ND)	New York, Niagara falls	1.89
Alaska, Kwigillingok	12.6	New York, Syracuse	2.59
Alaska, North Pole	22.2	New York, Yaphank	1.11
Alaska, Prudhoe Bay	1.67	Ohio, Columbus	9.62
Alabama, Dothan	10.7	Ohio, Painesville	1.41
Alabama, Montgomery	2.70	Ohio, Toledo	3.15
Arkansas, Little Rock	1.44	Oklahoma, Oklahoma City	1.99
British Columbia, Greenwood (Canada)	0.185	Ontario, Ottawa (Canada)	2.22
British Columbia, Prince Ge (Canada)	0.370	Ontario, Thunder Bay (Canada)	0.444
British Columbia, Revelstok (Canada)	0.667	Oregon, Portland	17.0
British Columbia, Vancouver (Canada)	2.52	Pennsylvania, Harrisburg	1.22
California, Berkeley	ND	Pennsylvania, Middletown	1.41
Colorado, Denver	ND	Pennsylvania, Pittsburgh	2.11
Connecticut, Hartford	1.96	Quebec, Quebec (Canada)	0.148
Delaware, Wilmington	0.892	Rhode Island, Providence	0.840
Florida, Jacksonville	1.11	South Carolina, Columbia	1.07
Florida, Miami	1.67	South Dakota, Pierre	2.78
Georgia, Blakely	ND	Saskatchewan, Regina (Canada)	0.111
Hawaii, Honolulu	4.44	Saskatchewan, Saskatoon (Canada)	0.222
Iowa, Iowa City	3.66	Tennessee, Knoxville	1.44
Idaho, Boise	62.2	Tennessee, Nashville	4.07
Idaho, Idaho Falls	97.3	Tennessee, Oak Ridge	5.07
Illinois, Chicago	1.74	Texas, Austin	0.829
Indiana, Indianapolis	1.85	Utah, Salt Lake City	86.2
Kansas, Topeka	2.11	Virginia, Lynchburg	4.44
Kentucky, Frankfort	3.26	Virginia, Virginia Beach	6.66
Louisiana, New Orleans	3.29	Vermont, Montpelier	61.4
Manitoba, Winnipeg (Canada)	0.148	Washington, Olympia	45.1
Maine, Augusta	3.29	Washington, Spokane	245.
Michigan, Lansing	7.40	Wisconsin, Madison	1.33
Minnesota, Minneapolis	12.2	West Virginia, Charleston	1.63
Missouri, Jefferson City	7.77	Wyoming, Cheyenne	120.
Mississippi, Jackson	1.29	Wyoming, Jackson	3.70

Table 3.14 Concentration ratio of milk/air (m^3/L) for ^{131}I

State and city	Concentration	State and city	Concentration
Alabama, Ashford	53.7	New York, New York	111
Alabama, Montgomery	68.1	Oregon, Portland	202
Arizona, Phoenix	12.3	Tennessee, Oak Ridge	141
California, Los Angeles	97.2	Utah, Salt Lake City	47.2
Colorado, Denver	20.8	Washington, Hanford	59.9
Idaho, Boise	77.9	Washington, Spokane	100
Minnesota, Minneapolis	91.5	New Brunswick, Fredericton (Canada)	17.5
Montana, Helena	124	Ontario, Ottawa (Canada)	9.79
North Carolina, Charlotte	76.6	Ontario, Toronto (Canada)	13.1
Nevada, Las Vegas	33.2		

4. 95% confidence limit, 8.33 and 331 m^3/L; and
5. coefficient of variation, 110%.

3.5.5 $CR_{rain/particulate\ air}$

The washout ratio of ^{131}I ($CR_{rain/particulate\ air}$) is determined for a given day and station as ^{131}I concentration in rain (Bq/L) divided by the particulate ^{131}I concentration in air (Bq/m^3). The results are a concentration ratio of rain to air with units of m^3/L. Only six dates were available with both rain and air concentrations above the limits of detection. Table 3.15 itemizes these ratios.

Table 3.15. Washout ratio of rain/air (m^3/L)

State and city	Date	Concentration
Idaho, Boise	May 10, 1986	1,139
Idaho, Boise	May 11, 1986	1,050
Montana, Helena	May 12, 1986	1,585
Montana, Helena	May 21, 1986	6,393
Ohio, Toledo	May 22, 1986	1,037
Ontario, Ottawa (Canada)	May 8, 1986	10,000

The method of collecting air samples actually results in an average concentration over a period of time, thus the small number of ratios. If an interval of time were examined around each reporting date of air concentrations, the number of ratios would increase dramatically. This additional analysis remains for a future research effort.

3.6 APPLICATIONS FOR MODEL VALIDATION

The EPA data base has the potential for use in model validation for predictions made on a regional or larger scale. The use of any single location for testing model predictions demands caution because of the possibility that the concentrations measured in air may not be entirely representative of the larger region for which milk samples were averaged. The lack of data on agricultural practices in use at the time the milk was collected is another factor that compromises the applicability of this data base for model testing.

Despite these limitations, the following observations can be made relative to the results obtained to date:

1. The estimated $CR_{milk/air}$ for ^{131}I (geometric mean of 52.5 m^3/L) is almost identical to values obtained for locations in Europe. The values of $CR_{milk/air}$ obtained from Chernobyl data are among the lowest ever reported for ^{131}I. All models reviewed to date by this committee overpredicted this relationship by one to two orders of magnitude (see Sect. 2.3.1).
2. The ARAC system at Lawrence Livermore National Laboratory predicted that maximum milk concentrations for the United States should not exceed approximately 300 Bq/L (9000 pCi/L), with a most probable MC of 30 Bq/L (900 pCi/L) (Dickerson and Sullivan 1986). These predictions exceed measured maximum concentrations in the United States by one to two orders of magnitude. Thus the predictions provided by the ARAC system provided a sufficient margin of safety for decisions.
3. The measured washout ratios ($CR_{rain/air}$) for particulate ^{131}I are generally about 10^3 m^3/L. These values are consistent with reported washout ratios for airborne particulates for other regionally dispersed particulate matter reported in the literature (Slinn 1984).
4. The EPA data could also be used to test model predictions of the regional pattern of deposition of Chernobyl fallout and to assess the terrestrial air-pasture-cow-milk transfer of radiocesium. Additional analysis of the patterns and processes governing wet deposition is also warranted.

3.7 REFERENCES

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4. CONCLUSIONS

An enormous amount of data has been collected as a result of the Chernobyl accident. These data have great potential for testing model predictions. However, the quality of data currently available for model testing is less than ideal. For example, most of the available data has been obtained from emergency response efforts rather than from carefully planned experimental designs established for the explicit purpose of model testing. In many cases, information about conditions of climate, meteorology, agricultural practices, and human dietary and living habits prevailing at the time of the measurements were not available. These limitations are expected to decrease in the near future as more research-quality analyses become available through scientific meetings and open literature publications.

Some aspects of radiological assessment models are more amenable to testing than others. Because of the abundance of data on ^{131}I and ^{137}Cs, the testing of terrestrial food chain models present the best opportunities. Testing of aquatic food chain models will probably be restricted to the long-term behavior of ^{137}Cs because initial attention was directed toward analyses of radioactivity in terrestrial food stuffs. Testing of models that predict dose from the inhalation and ingestion of radionuclides will depend on the extent to which information is available on the life and dietary habits of specific individuals at the time of exposure. Opportunities for testing model predictions of plume transport from the Chernobyl reactor are limited because of the lack of a well-characterized source term, the absence of nearby field meteorological data, and the fact that published predictions have been calibrated with environmental monitoring data.

Probably the most feasible testing of atmospheric transport models will compare their predictions of air concentrations, ground depositions, and exposures near the Chernobyl reactor against data provided by the USSR. Unfortunately, there are current obvious limitations associated with data received from the USSR to date.

Considering limitations associated with available data, model predictions of the source term appear to have been remarkably consistent. On the other hand, methodologies must be improved to derive source terms based on environmental measurements. A key lesson learned from the Chernobyl accident is that, regardless of where an accident may happen, data characterizing the source term may not be available because of inaccessibility of the area or a lack of information communicated by local authorities. Environmental measurements may be the only reliable system to estimate emissions of radionuclides. Based on this analysis, it is apparent that we currently lack a clear and consistent rationale for what environmental measurements should be taken, how these measurements should be used to derive source terms, and which format should be used to report source term data. Data acquired to date indicate that current radiological assessment models overpredict the transfer of ^{131}I and ^{137}Cs from air to pasture vegetation and from pasture vegetation to cow's milk. The extent of this overprediction is approximately one to two orders of magnitude for several locations in Europe and the United States. The model overpredictions were apparently influenced by the combined effect of (1) overestimating the amount of wet and dry deposited ^{131}I and wet deposited ^{137}Cs that were initially intercepted by and weathered from pasture vegetation and (2) the amount of these radionuclides transferred from the cow's diet into milk. Contrary to generally accepted assumptions, these results imply that the direct inhalation by humans of airborne ^{131}I may have been an important source of thyroid exposure for adult members of the population.

Analysis of data on ^{137}Cs in aquatic systems indicates that this radionuclide is transported more rapidly via runoff from the terrestrial system than is generally anticipated. Most assessment models consider the transfer of radionuclides from terrestrial to aquatic systems to be of minor importance with respect to potential radiological exposure.

In addition to the transfer of ^{137}Cs from watersheds into lakes and subsequent bioaccumulation into aquatic food chains, other

important exposure pathways not normally considered by radiological assessment models have been identified. Among the most prominent are the accumulation of particulate radionuclides in rain and subsequent use of cisterns to provide water for humans or livestock, the ingestion of goat's or sheep's milk, and the use of contaminated spring forage as a source of winter feed for livestock.

An illustrative analysis using the metabolic model for iodine of the ICRP shows reasonable agreement between predicted and observed ^{131}I levels in the adult thyroid. However, further information and data are needed to test the ICRP model for the metabolism of cesium. Predictions did tend to overestimate the ^{131}I levels in the thyroid. Although a number of factors could contribute to this overestimate, the relationship between ^{131}I levels in indoor and outdoor air should warrant further consideration because of the suggested increased significance of the inhalation route of intake.

This report presents the first published analysis of Environmental Protection Agency (EPA) monitoring data on Chernobyl fallout in the United States. Concentrations of ^{131}I in milk, rain, and air indicate that the highest exposures of individuals most probably occurred in the states of Idaho, Oregon, and Washington. Ratios of time-integrated ^{131}I concentrations in milk and air are comparable to those in Europe and are overpredicted by the radiological assessment models selected in this report. The maximum concentrations in milk are also well within upper limits forecast by the ARAC system at Lawrence Livermore National Laboratory.

Examples in this report of the use of Chernobyl fallout data for model testing are preliminary and have been presented for illustrative purposes only. Additional data will be required to confirm the results of these analyses and to explain the discrepancies between model predictions and observations. Such data may become available through international model testing programs such as BIOMOVS (International Biospheric Model Validation Study); the recent cooperative research agreement between the U. S. Department of Energy, Office of Health and

Environmental Research, and the Commission of the European Communities on the validation of terrestrial and aquatic food chain models; and renewed initiatives by the USSR in collaborative research related to the Chernobyl accident.

5. RECOMMENDATIONS

5.1 RECOMMENDATIONS FOR FURTHER MODEL TESTING

The following recommendations concern further testing of model predictions using Chernobyl data.

- Specific information on site specific conditions of climate and agricultural practices prevailing at the time data should be documented to reduce the number of assumptions required in making model predictions.

- Atmospheric concentrations of noble gases should be compared with concentrations of particulate radionuclides in the atmosphere to test predictions of plume transport and depletions resulting from deposition processes.

- When testing atmospheric models, predictions of plume trajectories, radionuclide concentrations in air, and depositions on the ground should be separated from estimates of dose. Failure to make this distinction will confuse processes affecting food chain transfers and human dosimetry with processes affecting atmospheric transport and deposition.

- Additional tests should be made of model predictions of plume depletion caused by wet and dry scavenging processes and wet and dry deposition in forests, urban areas, and open pastures. These results could apply to the assessment of other hazardous aerosols as well as radionuclides.

- Better and more complete data should be obtained to improve and extend the analysis of model predictions and observations of the transfer of ^{137}Cs through the air-forage-cow-milk pathway. Furthermore, terrestrial and aquatic food chain models should be tested for other radionuclides and other food types than those considered in this report.

- Model parameter values that are not typically varied according to site-specific conditions should be tested. These parameters include washout ratios, deposition velocities, vegetation mass interception factors and retention constants, and milk, meat, and soil-to-vegetation transfer coefficients.

- Tests should be made of model predictions of deposition on watersheds, runoff, and water and sediment concentrations in lakes and streams.

- Assumed steady-state concentration factors for terrestrial marine and freshwater environments should be tested. Data for testing should be based on ratios of time-integrated concentrations of radionuclides in receptor-to-donor compartments of the environment.

- Tests should be made of the role of soils and sediments as sinks as compared to their potential to act as secondary sources for the release of long-lived radionuclides.

- Dynamic accident assessment models should be tested against both time integrals and time histories of data.

- Probabilistic models should be tested to determine the extent to which the distributions of results can be explained by measured natural stochastic variability.

- Mandatory (e.g., KI administration) or voluntary (e.g., pathway avoidance) efforts implemented to reduce population exposures to radionuclides should be evaluated to determine the effectiveness of these actions.

- Support should be provided for scientists to become involved in cooperative international activities to validate models using Chernobyl data.

5.2 RECOMMENDATIONS FOR FURTHER USE OF THE EPA DATA BASE

The following recommendations concern future analysis using the EPA data base at Oak Ridge National Laboratory.

- Further analysis of washout ratios for particulate radionuclides is warranted and can be readily accomplished with only a small additional investment of time.

- Additional analyses should be conducted on ^{137}Cs in air, rain, and milk. Analysis of ^{137}Cs in milk will determine whether reported EPA values for milk concentrations are significantly different from concentrations resulting from historic atmospheric weapons testing.

- The radiological impact of Chernobyl fallout on the U.S. population should be evaluated. This effort would require merging radionuclide concentration data with data on population distributions and national milk production statistics for May and June of 1986.

- Techniques developed for analyzing data and contour mapping should be applied to other computer data bases on the global distribution of Chernobyl fallout.

5.3 RECOMMENDATIONS FOR ACCIDENT ASSESSMENT

The following recommendations concern improving future assessments of the consequences of major reactor accidents.

- The Chernobyl accident has demonstrated the need to assess accidents on a global and regional scale. Previous models have focused only on radiological impacts near a reactor (within 80 km).

- The Chernobyl accident has demonstrated that measurements of contamination in the environment must be used in conjunction with environmental transport models to estimate source terms. Use of these models is likely to depend on measurements performed far down wind of the reactor.

- The Chernobyl accident has also demonstrated the inadequacy of major monitoring networks to determine the total quantity of ^{131}I in air. Future emergency detection equipment should be designed to determine the particulate as well as the gaseous constituents of ^{131}I in air.

- Methodologies must be improved to derive source term estimates from environmental measurement to facilitate later dose reconstruction.

- Radiological assessment models should be refined at the process level to accurately reflect the dynamic behavior of radionuclides in the environment after an accidental release. Models should be amenable to altering initial conditions and should be capable of accepting different source terms (i.e., release rates, air concentrations or ground depositions of radionuclides). A clear distinction must be drawn between models for comparative risk assessment and models for assessing the consequences of actual releases.

- The identification of important exposure pathways that are not considered by current models necessitates reevaluation of which members of the population should be considered the "critical group." This reevaluation will have implications for environmental research as well as research on human dosimetry and should facilitate future development of protective action guides.

6. APPENDIX

6.1 REFERENCE MATERIALS OBTAINED AT ORNL RELATING TO THE CHERNOBYL ACCIDENT

This appendix is composed of a listing of documents, reports, open-literature publications, and technical communications archived at Oak Ridge National Laboratory to facilitate evaluation of Chernobyl fallout data for model validation. The identification numbers given at the end of each citation are for the purpose of document retrieval.

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