BRH/NERHL 70-3

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An INVESTIGATION of AIRBORNE RADIOACTIVE EFFLUENT from an OPERATING NUCLEAR FUEL REPROCESSING PLANT

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	Title
BRH/NERHL 70-1	An Estimate of Radiation Doses Received by Individuals Living in the Vicinity of a Nuclear Fuel Reprocessing Plant (PB 191 907)
BRH/NERHL 70-2	Liquid Waste Effluents from a Nuclear Fuel Reprocessing Plant (in process)
BRH/NERHL 70-3	An Investigation of Airborne Radioactive Effluent from an Operating Nuclear Fuel Reprocessing Plant (Former Title: Simultaneous Stack and Environmental Monitoring of Airborne Radioactivity at an Operating Nuclear Fuel Reprocessing Plant)
BRH/NERHL 70-4	Calibration and Field Tests of ^{85}Kr Detectors for Environmental Monitoring (in process)

An INVESTIGATION of AIRBORNE RADIOACTIVE EFFLUENT from an OPERATING NUCLEAR FUEL REPROCESSING PLANT

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FOREWORD

The recent growth in the utilization of nuclear reactors for electrical generating plants has brought with it the need for nuclear reprocessing plants that extract usable uranium and plutonium from spent fuel elements. The Bureau of Radiological Health performs a technical review and evaluation of the public health factors for all nuclear facilities and provides technical assistance to State health departments responsible for assessing population exposure and radiation levels in the environment. The Northeastern Radiological Health Laboratory, under the aegis of the Division of Environmental Radiation, has undertaken an environmental survey of the first commercial operating fuel reprocessing plant in the United States.

The technical reports series of the Bureau's regional laboratories and its Division of Environmental Radiation is used to publish the results of research projects and technical evaluations of nuclear facilities. These reports are distributed to State and local radiological health program personnel, Bureau technical staff, Bureau advisory committee members, radiation safety officers, libraries and information services, industry, the press, and other interested individuals. These reports are also included in the collection of the Library of Congress and the Clearinghouse for Federal Scientific and Technical Information.

I encourage the readers of these reports to inform the Bureau of any omissions or errors. Your additional comments or requests for further information are also solicited.

John C. Villforth

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Director
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PREFACE

The projected increase in the utilization of nuclear power for electrical generating plants has resulted in both State and Federal public health agencies placing increased program emphasis on the surveillance of nuclear power plants. The Bureau of Radiological Health provides recommended nuclear facility surveillance program information for the guidance of health agencies. In order to provide a better technical basis for environmental surveillance recommendations, a series of field studies have been conducted at operating nuclear facilities to obtain better data on radionuclides in plant effluents and their subsequent distribution in the environment.

This field study around the Nation's first commercial nuclear fuel reprocessing plant was for the purpose of identifying the movement of critical radionuclides through the atmospheric and aqueous environments in relationship to the fuel reprocessing procedures. Technical data obtained from this study will be used to: (1) develop minimum and optimum requirements for environmental surveillance around nuclear fuel reprocessing plants, (2) designate the radionuclides and environmental pathways of greatest dosimetric significance, and (3) provide estimates and measurements of the radiation dose to populations in the area of a facility.

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CONTENTS

TABLES

FIGURES

ABSTRACT

This paper describes studies carried out at an operating nuclear fuel reprocessing plant for the purposes of characterizing the stack effluent, measuring the environmental levels of activity due to components of stack release, and evaluating instrumentation and methodology used to sample both at the stack and in the environment.

Four field sampling stations, located in the vicinity of the plant perimeter, and a stack sampler simultaneously monitored ^{85}Kr, ^{129}I, and ^{3}H (gaseous and water vapor) during two dissolution cycles. Particulates were monitored at the stack and one field station. Measurements are presented and discussed in terms of emission level versus specific plant operations, primarily the dissolution cycle, In addition, observed and theoretical dilution factors are compared and, based on meteorological considerations, show reasonable correlation.

The instrumentation used in this study includes thin-window geiger detectors and flow-through ionization chambers for ^{85}Kr; bubblers, traps, and grab samplers for ^{3}H; and resin traps for ^{129}I. Choice of methodology and instrumentation is discussed with emphasis placed on a system usable in determining dose to a population in the plant vicinity.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the following for their contributions.

Northeastern Radiological Health Staff, BRH

Development of the ^{129}I Sampler
Paul B. Hahn

^{85}Kr - ^{3}H Calibration and Analyses
Emilio J. Troianello

Implementation of Study
A. A. Bertuccio
J. T. Murphy
C. Rosenberg
G. C. Nicholsen

Radiological Engineering Laboratory, DER, BRH

Meteorological services during the study
David B. Smith

Nuclear Facilities Branch, DER, BRH

Assistance in planning and implementing the study
James E. Martin, Deputy Chief

Participants from other agencies are listed below and their help is gratefully acknolwedged.

Richard B. Keely - In Charge of Health and Safety
Nuclear Fuel Services, Inc.

Kurt Anderson - Senior Sanitary Engineer
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New York State Health Department

Walter R. Lorenz - Radiation Specialist
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AN INVESTIGATION OF AIRBORNE RADIOACTIVE EFFLUENT FROM AN OPERATING NUCLEAR FUEL REPROCESSING PLANT

INTRODUCTION

Increase in the use and number of nuclear reactors has brought with it the need for nuclear fuel reprocessing plants that extract usable uranium and plutonium from spent fuel elements. Presently, one commercial plant is in operation in western New York, a second plant is under construction in Illinois and others are being planned. The fact that such plants discharge radionuclides to the environment necessitates surveillance around the plant in order to determine the quantity and composition of waste-discharge, the effect of such discharge on the environment where it accumulates, and the level of radiation dose delivered to surrounding populations.

The Bureau of Radiological Health has a responsibility to provide guidance and assistance in developing surveillance programs for nuclear facilities including fuel reprocessing plants. In order to meet this responsibility, a study of a nuclear fuel reprocessing plant and its environs was proposed for the purpose of determining the requirements of environmental surveillance programs for such plants.

The objectives of this study are:(1)

1. To develop minimum and optimum requirements for environmental surveillance programs around nuclear fuel reprocessing plants.
2. To develop a uniform set of surveillance methods that can be adopted by other states and companies in meeting surveillance needs.
3. To identify specific radionuclides that may be released in liquid and gaseous waste discharges and the pathways by which they are dispersed in the environment.
4. To relate the levels of released radionuclides to levels in critical pathways in order to specify the most beneficial sampling and analyses to perform.

This paper describes Nuclear Fuel Services, Inc., (NFS), a fuel reprocessing facility, with respect to its gaseous discharge and reports the results of a preliminary airborne effluent study carried out at NFS during a field trip in June 1969. These results include:

1. Characterization of important radioactive components of the stack effluent, including krypton 85, iodine 129, and tritium in both the gas and water vapor forms. Relative emission rates of both krypton 85 and tritium during typical dissolution cycles are discussed.

2. Simultaneous monitoring of stack release and environmental levels of activity using krypton 85 as a tracer. Observed dilution factors are presented and compared to dilution factors calculated from diffusion equations.

3. Evaluation of the environmental survey instrumentation and methodology used for the study. Instrumentation and sampling devices used in this study include thin-window geiger detectors, flow-through ionization chambers, evacuated tanks, bubblers, and ion exchange resin traps. Emphasis was placed upon a choice of system usable for environmental surveillance to determine population dose estimates.

NUCLEAR FUEL SERVICES — A DESCRIPTION

Nuclear Fuel Services, Inc. (NFS) is located in western New York approximately 30 miles southeast of Buffalo at Ashford, in Cattaraugus County. The plant is centered on a 3,000-acre site (fig. 1).

A. GEOGRAPHY AND METEOROLOGY (2,3)

The NFS plant is located on an undulating plain surrounded on the south, east, and west by hills that reach a height of 500 feet above the plant level. North of the plant, the rolling plain extends several miles with two ravines intersecting the plain (fig. 1). The Buttermilk Creek runs through the plant property and forms one ravine approximately 100 feet in depth going from southeast to northeast. The second ravine is formed by the Cattaraugus Creek. This ravine is 300 feet deep in an east to west direction approximately 3 miles north of the plant.

Annual wind rose data indicate that the predominant wind directions are from the northwest and from the south. These wind rose data are based on actual measurements at the plant site and on estimates derived from general wind patterns aloft which were adjusted for local terrain.

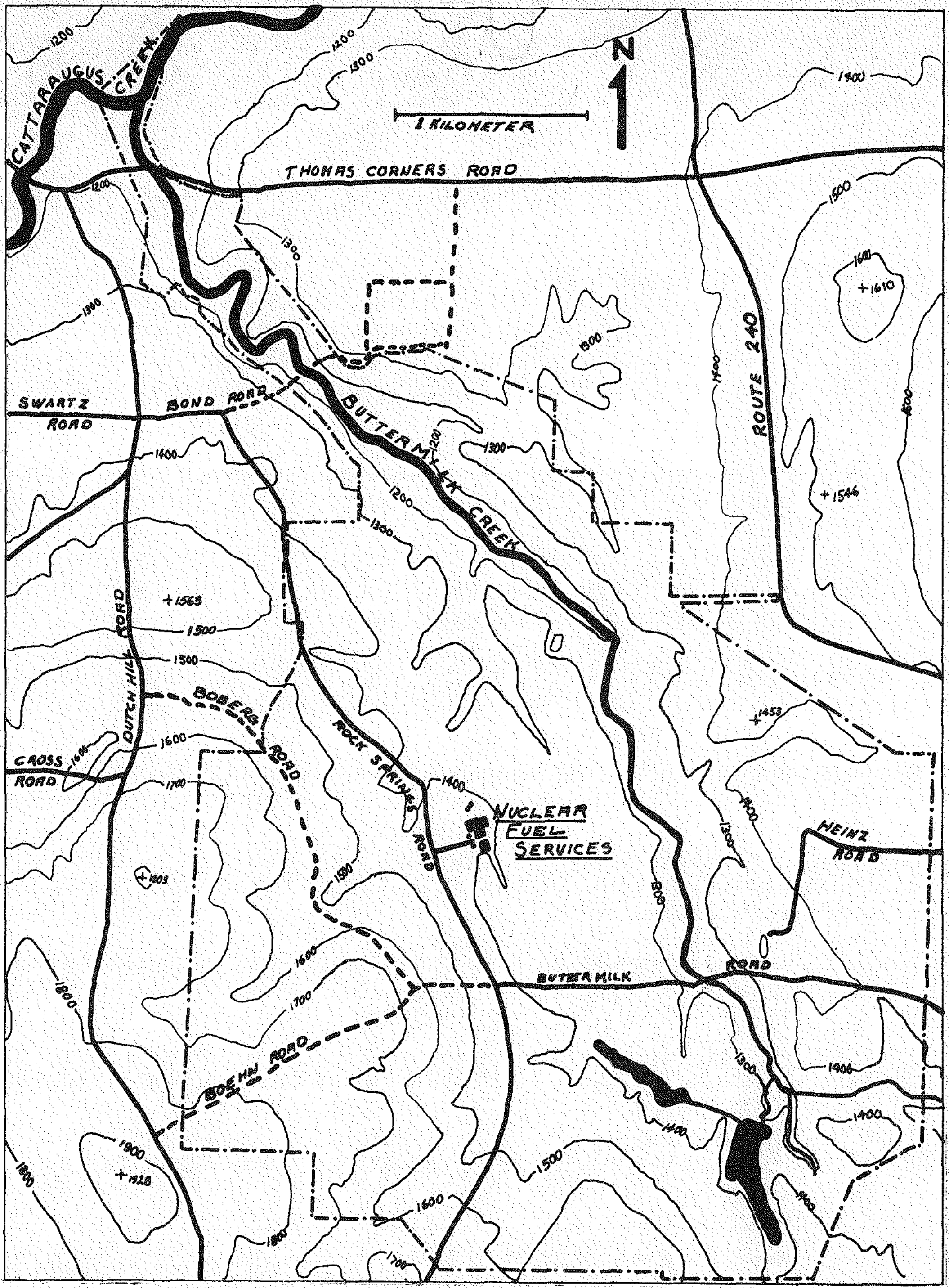

Figure 1. Map of Nuclear Fuel Services.

B. REPROCESSING PLANT DESCRIPTION AND PREDICTED AIRBORNE RELEASES (2,4)

NFS reprocesses fuel using the chop-leach-solvent extraction process. The plant operating capacity at the present time is about one metric tonne of fuel per day. Spent fuel is aged for at least 150 days at the reactor and then undergoes reprocessing in the following steps:

1. Mechanical Chopping: The elements are chopped into small pieces and placed into baskets.

2. Dissolution: Baskets of chopped fuel elements are placed in a dissolver vat. Boiling nitric acid leaches the fuel from the elements leaving a residue of cladding hulls.

3. Solvent Extraction: The solution obtained from the dissolution cycle is put through several extraction cycles which result in the separation of uranium and plutonium from the fission products.

4. Purification: The uranium and plutonium products are further purified by additional solvent extraction and ion exchange processing.

5. Recovery Cycle: The solvent used in the extraction cycle is purified for reuse by chemical washing. The nitric acid is purified by evaporation and concentrated for reuse by fractionation.

Tritiated water is liberated from the dissolution bath and the various evaporation processes. ^{85}Kr, ^{131m}Xe, ^{133}Xe and gaseous tritium are released in small amounts during the chopping; the remaining major portion of these nuclides are released during the dissolution cycle. ^{129}I and ^{131}I are released in small amounts during chopping, dissolution, and extraction and possibly in smaller amounts from other reprocessing steps. (Airborne particulates originating during chopping are expected to be almost completely retained by an "absolute" filtration system.) The predicted release of airborne activity from reprocessing to the off-gas system is shown in table 1.

All gaseous activity is released from a 60 meter stack. The volume of air discharged from the stack is approximately 40,000 cfm. The stack discharge consists of filtered air from the processing complex off-gas lines as well as from the tank farm, fuel storage area, various labs, hoods and ventilation systems.

C. NFS STACK SAMPLING AND MONITORING SYSTEMS

The stack sampling and monitoring system used by NFS is shown in figure 2. The sampling system is located in close proximity to the sampling port to minimize losses. The system contains a filter for particulate collection, a charcoal bed for iodine collection and operates under isokinetic conditions.

The monitoring system is located in the plant some distance from the stack. The system is tied to the stack by 210 feet of piping. The complete system monitors particulates, ^{131}I noble gas activities.

Table 1. Predicted airborne activities from reprocessed fuel[a]

Radionuclide	Activity Released from fuel (curies)	Activity Discharged from stack (curies)[b]
^{85}Kr	6.3×10^{3}	6.3×10^{3}
^{129}I	2.2×10^{-2}	1.1×10^{-4}
^{131}I	1.8×10^{0}	9.0×10^{-3}
^{131m}Xe	1.0×10^{0}	1.0×10^{0}
^{133}Xe	3.8×10^{-3}	3.8×10^{-3}
^{3}H	5.0×10^{1}	5.0×10^{1}

[a] Fuel 1 tonne at 2×10^{4} megawatt days burnup and aged for 150 days.

[b] Activity discharged from the stack is equal to activity released from fuel for all nuclides with the exception of the radioiodines which are retained with an estimated efficiency of 99.5% by a silver reacting scrubber.

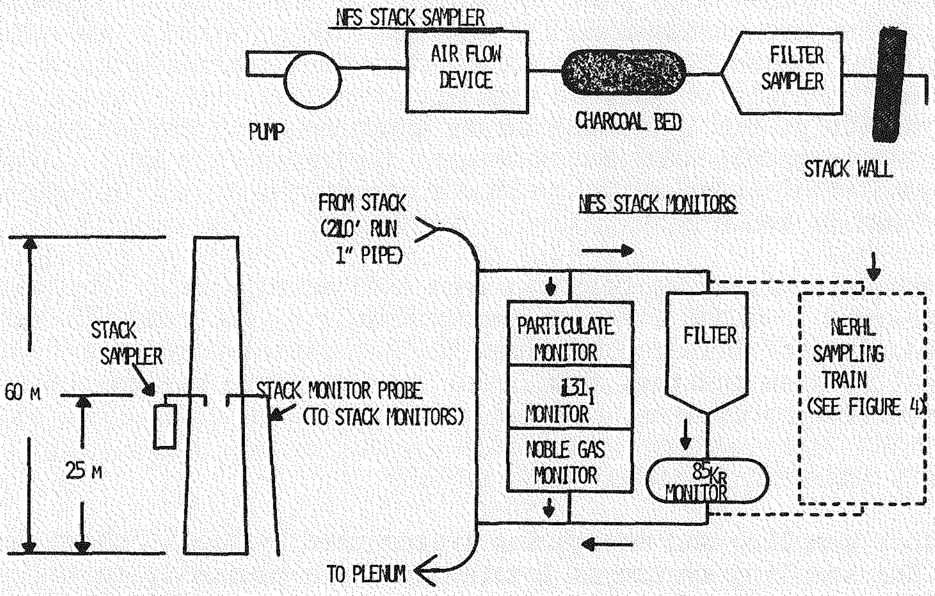

Figure 2. Location of NFS stack monitoring system and NERHL sampling train.

INSTRUMENTATION

A. CHOICE OF SPECIFIC RADIONUCLIDES TO BE MEASURED

The objective of the June 1969 field trip was to make simultaneous measurements of specific radionuclides at the stack and in the environment in order to characterize the stack effluent and its dispersion to the environment. In preparation for this study, instruments and samplers were developed for measurement of the major long-lived radionuclides being emitted from the stack. ^{85}Kr, ^{129}I, and ^{3}H are the major airborne nuclides emitted during normal operation of the NFS plant (table 1). and these radionuclides were studied during the field trip.

B. DETECTION SYSTEMS

The detection systems developed for the NFS study will be described briefly in this section. A detailed analysis of the calibration and characteristics of all field ^{85}Kr detection systems used in this study is presented in a complementary report, "Calibration and Field Testing of ^{85}Kr Detectors for Environmental Monitoring Applications"(5).

1. Ionization Chambers

Flow-through ionization chambers, in conjunction with vibrating-reed electrometers, were used to monitor ^{85}Kr at the stack and in the field. Maximum sensitivity was obtained by designing the detection system to compensate for factors that affect sensitivity. These factors are volume, humidity of the sampled air, the presence of naturally occurring radon, tritiated water, and external background.

Radon was minimized by use of charcoal traps at reduced temperature preceeding the ionization chamber. A charcoal trap (at -78° C) selectively held up radon gas while allowing krypton gas to flow through with minimal hold-up time. Particulate radon daughters were removed by a 0.8 μ pore size millipore filter. Under field conditions it was impractical to reduce background due to external radiation with lead shielding. The tritium, most of which was tritiated water vapor, was eliminated by placing a drying bed, such as Drierite[1], in the upstream sampling line. This drying bed also eliminated any possible high humidity condition in the chamber which could create high background readings due to leakage across the input resistor of the electrometer.

The ionization chamber systems used at NFS consisted of a 1.0 liter chamber for stack sampling and two field systems--one containing a 2.8 liter chamber and the other a 4.3 liter chamber. All three

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[1] Anhydrous calcium sulfate

systems had a charcoal trap and drying bed in the upstream sampling line. Table 2 shows the minimum detectable concentrations for the ion chambers under actual field conditions. The minimum detectable con-concentrations (MDC)[2] is defined as that level of ^{85}Kr where the net count is equal to two times the total sample standard deviation (s_t). The total standard deviation is comprised of the standard deviation of the net count (s_c) and the standard deviation resulting from the instrumental and procedural variations (s_i). Average backgrounds and their normal fluctuations for actual sampling locations used during the study are also shown in table 2.

The calibration factors for the three chambers at 20° C and 760 mm Hg are as follows:

Chamber Size	$\frac{\text{amps}/\mu\text{Ci}}{\text{cc}} \pm 2\, s_t$
1.0 liter	$5.1 \times 10^{-9} \pm 13\%$
2.8 liter	$2.0 \times 10^{-8} \pm 8\%$
4.3 liter	$3.6 \times 10^{-8} \pm 8\%$

Table 2. Minimum detectable ^{85}Kr activity for flow-through ionization chamber under field conditions

Chamber	Location	MDC µCi/cc	Average Background current amps $\pm 2\, s_t$
1.0 liter	Stack	1.5×10^{-5}	$1.4 \times 10^{-13} \pm 20\%$
2.8 liter	Field	3.9×10^{-8}	$2.8 \times 10^{-15} \pm 16\%$
4.3 liter	Field	2.4×10^{-8}	$3.3 \times 10^{-15} \pm 16\%$

2. Geiger Detectors

The GM tubes were calibrated for use in field monitoring of ^{85}Kr. Three of these detectors are end window tubes with a pancake geometry such that the window area is quite large, while the internal volume remains small. The fourth tube is a thin walled probe. Table 3 describes the tube specifications.

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[2] Also called 'Minimum Significant Measured Activity" by B. Altshuler and B. Pasternack, Health Physics Vol. 9:293 (1963).

Table 3. GM Detector specifications[a]

Tube Type	Window dia. cm	Window Thickness mg/cm^2	Tube dimensions cm
EON 8008T Double Window	4.4	1.4-2	4.4 dia x 1.2
EON 8001T Single Window	4.4	1.4-2	4.4 dia x 1.1
AMPEREX 18546 Single Window	5.1	3.5-4	5.8 dia x 4.5
EON 5108E Probe	--	85% - 30 15% - 172	1.5 dia x 14.1

[a] Data taken from manufacturer's specification sheets

The field system used at NFS consisted of the single window detectors coupled to either a scaler or chart readout. The chart readout system was battery operated and portable and it was easily carried by one man.

Table 4 shows the minimum detectable concentration (MDC) for the geiger detectors. The average background and its estimated fluctuations is also shown. The two detectors listed are the single window detectors used during the NFS field trip. A "short count" is the counting time used during the study to monitor, in increments, one dissolution process and characterize the ^{85}Kr concentration over short periods of time within the dissolution period. Analysis of data from the field trip shows that the MDC of these two detectors is primarily dependent upon instrument errors and statistical counting errors, both of which are quite large. The results indicate, however, that some improvement in MDC is realized by extending the counting time.

The calibration factors for the geiger detectors discussed, assuming immersion in an infinite cloud of ^{85}Kr, are as follows:

Detector	cpm/µCi/cc $\pm$ 2 s_t
EON 8001T	$3.6 \times 10^8 \pm 13\%$
AMPEREX 18546	$4.8 \times 10^8 \pm 14\%$

Table 4. Minimum detectable concentration of ^{85}Kr for geiger detectors

Type	Conditions	MDC (μCi/cc)	Avg. background (cpm $\pm$ 2 s_t)
EON 8001T	Actual field conditions[a] (Short count)	4.0×10^{-8}	50 ± 8
AMPEREX	Actual field conditions (Short count)	4.4×10^{-8}	76 ± 10

[a] Conditions
Counting time BKG 10 min.
Counting time Gross 2 min.
Inst. error - 2 s_i = 8 cpm

C. SAMPLING SYSTEMS

Samplers used in the NFS study were developed specifically for tritium, krypton, and iodine collection. All sampler systems were simple in design and easy to operate in the field.

1. Bubblers

Gas washing bottles (bubblers) are efficient devices for collection of tritiated water vapor. The washing bottle was filled with a volume of water and air was bubbled through a fritted disc located at its base. The stack sampling train used during the NFS study (containing 200 ml of water) included a bubbler. The bubbler sampler was subjected to a flow rate of 3 liters per minute during a total sampling time of less than 300 minutes. Under these conditions, the collection efficiency for water vapor in the air was greater than 98%. The efficiency for tritium gas collection under these conditions is negligible.

The bubbler samples were analyzed for tritium using a liquid scintillation technique requiring a 5 ml sample.

2. Drying Traps

Drying traps using "Drierite" or silica gel will remove water vapor from air passed through the bed. These beds make very efficient samplers for tritiated water vapor in air. At NFS "Drierite" traps were used to sample for tritium at the stack and in the field.

The minimum sample quantity necessary for analysis was 5 ml of water. For average sampling conditions of 60% relative humidity at 20° C the volume of air required for sampling is approximately 60 liters.

3. Evacuated Tank Samplers

Evacuated tanks are routinely used for collecting air samples known as grab samples. With the addition of a sensitive flow control on the tank, it is possible to sample over long time periods and obtain an average or integrated sample. Three sizes of evacuated tanks were used during the study, the sizes being 1.8, 8.2, and 34.0 liters. Attached to each tank inlet was a rotometer and needle valve which regulated flow so that the tanks could sample at a steady rate for periods in excess of an hour. A "Drierite" trap was also attached to each inlet to remove all water vapor from the air sampled. These tanks were used to collect ^{3}H gas and ^{85}Kr.

4. ^{129}I Ion exchange Sampler(6)

Gaseous iodine (I_2) can be collected by absorption onto an ion exchange resin, and this method of collection was utilized in the design of the ^{129}I sampler. The sampling apparatus consisted of a 1.5 cm diameter U-tube filled with approximately 20 ml of dry anion-exchange resin.[3] A pre-weighed I_2 crystal was inserted upstream to the resin to act as a carrier for the ^{129}I gas and to facilitate recovery and analysis of the ^{129}I collected on the resin. The efficiency of the sampling system was greater than 99% for a 2-hour sampling time at a flow rate of 3 liters per minute.

The samples were analyzed by chemical removal of the ^{129}I from the resin and measuring by either of two methods: liquid scintillation counting of the ^{129}I beta or by neutron activation analysis and counting the gamma emissions of the induced ^{130}I activity.

5. Filter Samplers

NFS operates an isokinetic filter sampler at the stack (see figure 2). This sampler operates continuously and filters are changed at weekly intervals. The filter used in this sampler is a 47 mm diameter, Gelman Type E. The sample flow rate through the filter is 0.6 cfm.

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[3] Dowex 1 - X8, Cl form anion resin.

Filters were obtained from the plant and analyzed for particulate activity. A high volumn sampler was used in the field during the study to determine the particulate activity in the environment.

FIELD TRIP IMPLEMENTATION

A. SITE SELECTION

In the spring and summer of 1969, NFS processed fuel from the Yankee Reactor, Rowe, Massachusetts. The complete fuel load consisted of 20 metric tons of fuel divided into 23 batches. The processing schedule averaged one dissolution every two days (7). Our study of the stack effluent consisted of monitoring the stack and the environment simultaneously during two dissolutions, specifically the dissolution of batch #13 on June 12 and batch #14 on June 14, 1969.

Five sampling stations were employed. One station was located in-plant at the stack and four stations were located in the field. The four field station locations were selected using the following criteria: a) stations equally spaced, b) lateral spread of stations large enough to give coverage of stack plume fluctuations during the sampling period, and c) stations located near the property line if practical.

Station location was decided upon one hour before dissolution to allow for equipment set up. The final decision was based on the meteorological prediction of plume behavior for the three-hour dissolution cycle. This prediction was derived from local wind speed, direction, and stability conditions. Figure 3 shows the sampling station locations chosen for both dissolutions. Each station distance and azimuth with respect to the plant is also shown.

B. COMPOSITION AND OPERATION OF SAMPLING STATIONS

The in-plant sampling station consisted of one train that contained the necessary systems for detecting and sampling ^{85}Kr, ^{3}H, and ^{129}I. Basically this sampling train was a closed loop connected into the existing NFS stack monitoring system already described in figure 2. The sampling station enabled sample collection of ^{129}I, ^{3}H both in the gas and liquid states and ^{85}Kr. Monitoring capability consisted of ^{85}Kr detection. Figure 4 is a schematic of the in-plant sampling train showing the specific components and their relative placement.

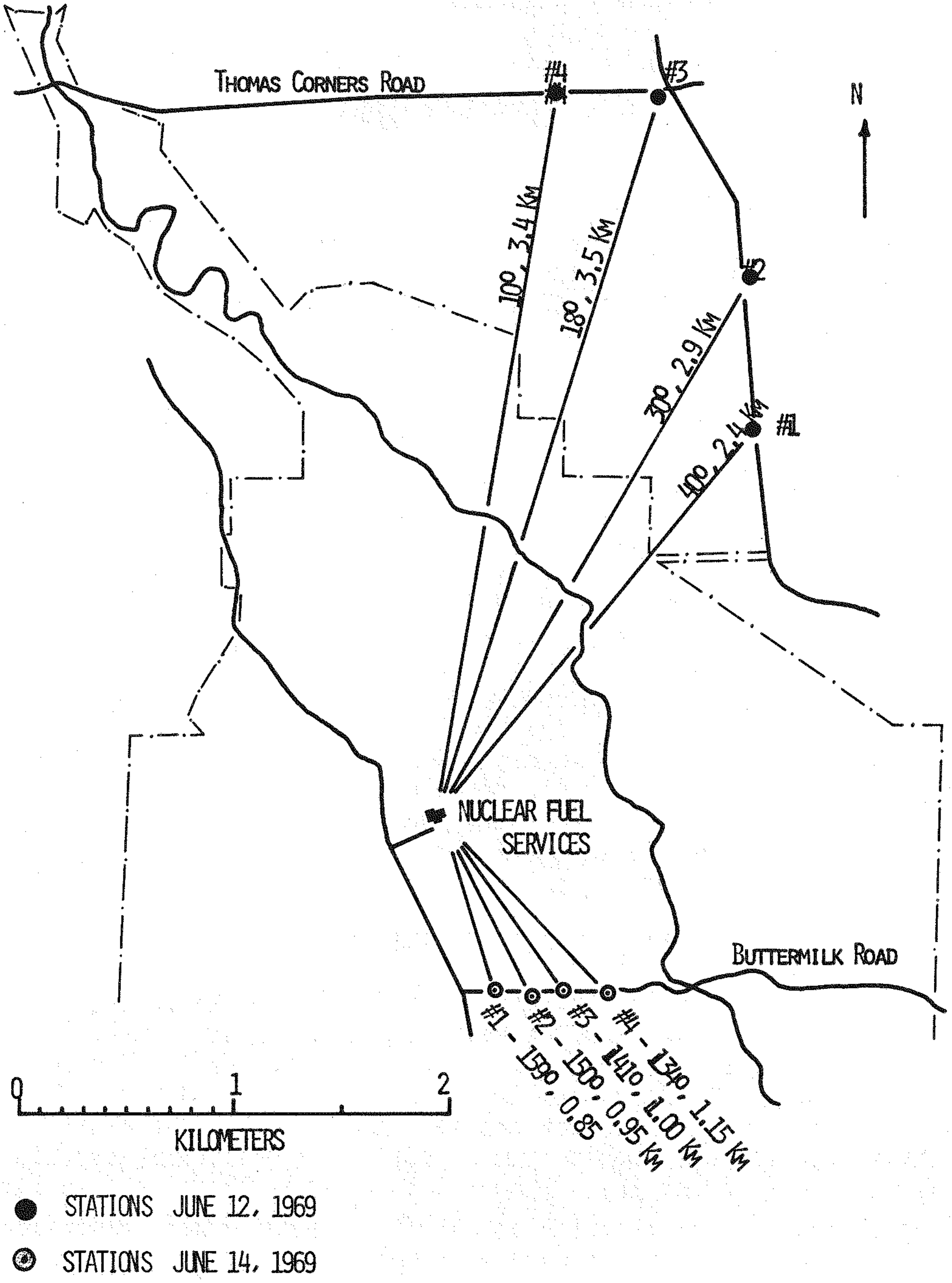

Figure 3. Sampling station locations

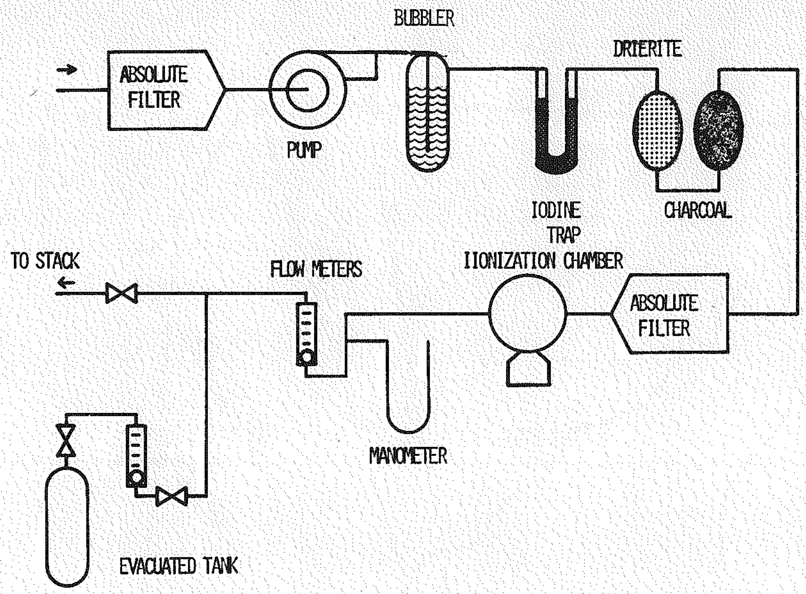

Figure 4. NERHL in-plant sampling train

The train functioned in the following manner:

Train Component	Function
Filter	Remove particulates and radon daughters
Bubbler Trap	Remove ^{3}H water vapor
Iodine Trap	Remove iodine
Drierite Trap	Remove moisture
Carbon Bed	Remove radon
Filter	Remove carbon particulates and residual radon daughters
Ionization Chamber	Monitor ^{85}Kr
Evacuated Tank	Sample ^{85}Kr and ^{3}H gas

Relative to ^{85}Kr, the ^{3}H gas activity is so small (<1%) that the ionization chamber response to ^{3}H is negligible. The stack sampling train enables characterization of the ^{85}Kr output during dissolution as well as collection of integrated samples of tritium and iodine.

The four field stations were classified as follows: two primary and two secondary stations. Each primary station included the following devices:

1. Ionization Chamber with carbon trap - ^{85}Kr Monitor
2. Geiger Detector - ^{85}Kr Monitor
3. Drierite Bed - ^{3}H (water) sampler
4. Evacuated Tank with "Drierite" Trap - ^{85}Kr, ^{3}H gas sampler

In addition, a high volume particulate sampler was placed at one primary station. The monitors operated throughout the dissolution cycle and the tanks sampled from 45 minutes to an hour during the early part of the dissolution cycle. Secondary stations consisted of an evacuated tank sampler. Referring to figure 3, the primary and back-up stations were distributed in the following manner:

Date	Primary	Secondary
6/12/69	Station #2, 3	Station #1, 4
6/14/69	Station #2, 4	Station #1, 3

RESULTS AND DISCUSSION

This section contains all data collected during the study at NFS. Errors are expressed in terms of sample standard deviation. When possible the total sample standard deviation (s_t) is used. The total sample standard deviation consists of the sample standard deviation of both the net count (s_c) and procedural and instrument variations (s_i). In cases where the procedural or instrument variation is unknown, the data are presented with only the associated sample standard deviation of the net counts (s_c). Data are presented in the following order: 1) in-plant data, 2) field data, 3) comparison of in-plant versus field data using meteorological analyses, and 4) annual environmental concentrations predicted from long term meteorological measurements.

A. IN-PLANT STACK DATA

1. ^{85}Kr - Ionization Chamber

Figures 5 and 6 show the measured stack output of ^{85}Kr for the dissolution of batch #13 on June 12, and batch #14 on June 14, 1969, respectively. The general curve shapes show that ^{85}Kr output reaches a maximum approximately 10 minutes after dissolution is initiated, then decreases to background levels within three to four hours. Significant fluctuations in the curves are due to changes in the rate of dissolution. This rate is controlled by the dissolving bath temperature, which can be altered throughout the cycle. The area under both curves above 10^{-5} µCi/cc includes greater than 99.9% of the total ^{85}Kr discharged during the respective dissolution cycles. Integration of the output curves, using a stack flow rate of 1.13×10^{9} cc.min (7), gives measured ^{85}Kr discharges as follows:

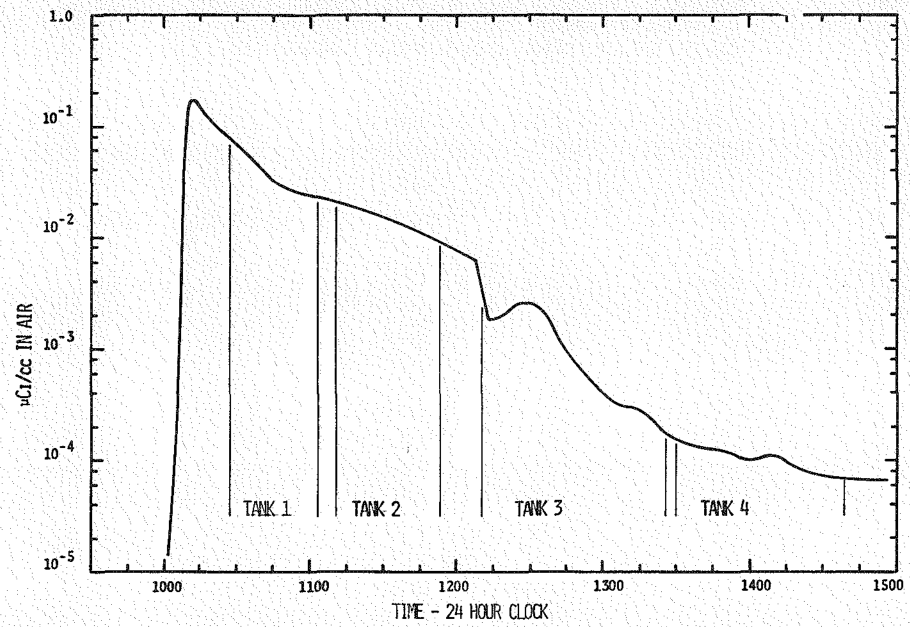

Figure 5. ^{85}Kr response of 1.0 liter ionization chamber to stack effluent during dissolution 6/12/69

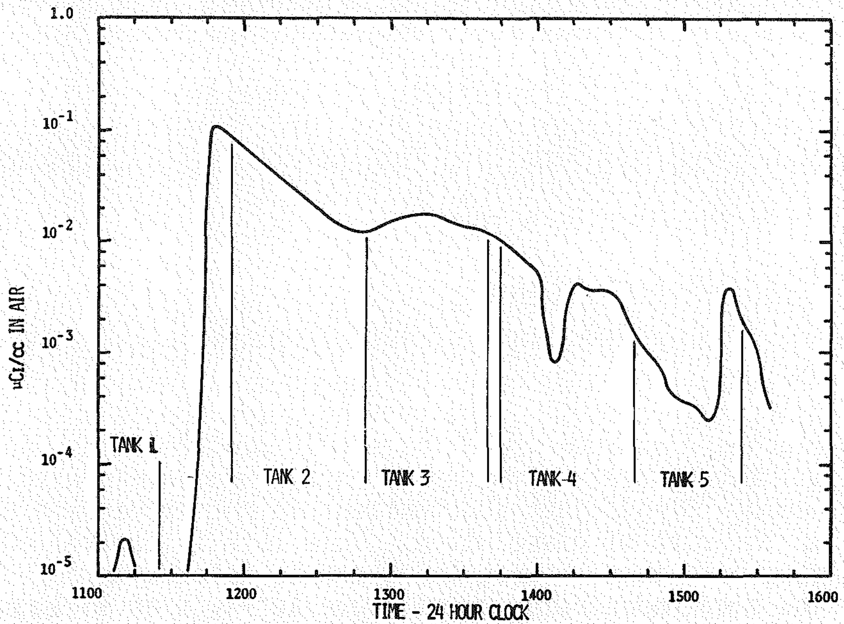

Figure 6. ^{85}Kr response of 1.0 liter ionization chamber to stack effluent during dissolution 6/14/69

Batch #13 - 6/12/69 5117 ± 512 curies
Batch #14 - 6/14/69 4730 ± 473 curies

^{85}Kr output can be predicted from burnup data on the individual batches using the following equation (8).

$$A = \frac{\left(e^{-\lambda t_1}\right)\left(\frac{1}{t_2}\right)\left(\frac{1-e^{-\lambda t_2}}{\lambda}\right)(Y)(\lambda)(BU)(T)}{\left(1.854 \times 10^{-24}\ \frac{MWD}{MEV}\right)\left(E\right)\left(2.22 \times 10^{12}\ \frac{dpm}{Ci}\right)} \qquad \text{(Eq. 1)}$$

Where t_1 = post irradiation time (days)
t_2 = in core irradiation time (800 days)
Y = yield factor for ^{85}Kr
λ = 1.26×10^{-7} min^{-1}
BU = burnup $\frac{\text{Megawatt days}}{\text{Metrictonne Uranium}}\ \frac{\text{(MWD)}}{\text{(MTU)}}$
T = weight of batch MTU (Tonne)
E = MEV/fission

The NFS data for the two batches observed are as follows (7).

Batch #13	Batch #14
BU = $2.57 \times 10^4\ \frac{MWD}{MTU}$	BU = $1.66 \times 10^4\ \frac{MWD}{MTU}$
T = 0.79 MTU	T = 1.06 MTU
t_1 = 480 days	t_1 = 960 days

For the above values, Y and E were assumed based on Yankee Fuel Burnup (8) as follows:

Batch #13	Batch #14
Y = 2.1×10^{-3}	Y = 2.3×10^{-3}
E = 201 MEV fission	E = 201 MEV fission

This results in a predicted yield of ^{85}Kr for each batch of:

Batch #13 - 6/12/69	5700 Ci
Batch #14 - 6/14/69	4750 Ci

These values compare well with the measured output considering the prediction equation is based on past observations of Yankee Fuel and does not consider the specific placement in the reactor core of the two batches of fuel measured at NFS.

It is expected that the measured ^{85}Kr output from a dissolution will show a small deficit since some ^{85}Kr is released during the chopping operation. Chopping outputs were not measured during the study, but estimates based upon NFS stack monitor measurements indicate 20 to 50 curies of ^{85}Kr were released per batch during the chopping operation.

2. ^{85}Kr Tank Data

1.8 liter tank samples were obtained during intervals throughout the two dissolution cycles. The sampling interval for each tank is indicated in figures 4 and 5 by vertical bars and the tank number is noted for each time segment. As shown, four samples were obtained from batch #13 and five samples were obtained from batch #14. During sampling an effort was made to obtain integrated samples by sampling at a constant rate. This required adjustments of a needle valve periodically throughout the sampling interval. As the evacuated tanks filled with the sample, more frequent adjustments became necessary and uniformity of flow rate became more difficult to maintain. Tank samples were analyzed by gamma counting, using dual 4" x 5" NaI(Tl) crystals with summed pulses displayed on a multichannel analyzer.

Table 5 shows the average concentrations of ^{85}Kr in the stack effluent for the nine tank samples along with the average concentrations obtained by integrating the ionization chamber output curves (figures 5 and 6) for the various tank sampling intervals.

The results indicate good correlation between the tank samples and the equivalent ionization chamber values. Samples 6/12-1 and 6/14-1 show the least correlation due to a tendency for the tanks to sample at a greater rate during the beginning of the sampling interval. These two tanks were sampling during time intervals when the rate of change of the stack concentration was maximum.

Table 5. Evacuated tank ^{85}Kr stack concentrations

Date-tank no.	Time interval	Tank samples µCi/cc[a]	Ion chamber equivalent µCi/cc[b]
6/12-1	1027-1103	4.2×10^{-2}	3.6×10^{-2}
6/12-2	1110-1153	1.5×10^{-2}	1.4×10^{-2}
6/12-3	1210-1325	1.2×10^{-3}	1.2×10^{-3}
6/12-4	1330-1438	1.1×10^{-4}	9.8×10^{-5}
6/14-1	1125-1155	1.8×10^{-4}	2.5×10^{-4}
6/14-2	1155-1250	3.1×10^{-2}	3.2×10^{-2}
6/14-3	1250-1340	1.3×10^{-2}	1.4×10^{-2}
6/14-4	1345-1440	4.0×10^{-3}	3.9×10^{-3}
6/14-5	1440-1525	9.6×10^{-4}	9.8×10^{-4}

[a] 2 s_c error = ± 12%

[b] 2 s_t error = ± 16%

3. ^{3}H Water Vapor - Bubbler Samples

Two bubbler samples were obtained during each dissolution. Analysis of a "Drierite" trap down stream from the bubbler in the sampling train indicates that the bubbler collection efficiency, for tritiated water, was greater than 98% in all cases. The values obtained are shown in Table 6.

Table 6. Stack tritiated water vapor during dissolution

Date-sampler	Time interval	Avg. act. ± 2 s_c µCi/cc in air	Total stack output-curies
6/12 Bubbler	0845-1200	3.09×10^{-7} ± 10%	6.8×10^{-2}
6/12 Bubbler	1215-1400	5.18×10^{-7} ± 12%	6.2×10^{-2}
6/14 Bubbler	1045-1340	2.36×10^{-7} ± 8%	4.7×10^{-2}
6/14 Bubbler	1342-1535	3.47×10^{-7} ± 10%	4.4×10^{-2}

The estimated tritium available per batch of fuel dissolved was approximately 40 to 50 curies (2). If a steady state is assumed for tritium stack output, the above outputs for a 24-hour period will be 0.6 curies for batch #13 and 0.4 curies for batch #14. This is about 1.5% of the total available tritium. If dissolution is the major source of tritium, then the total stack output will be less than the steady-state estimate. A more sophisticated tritium balance evaluation is necessary before any precise characterization of tritium can be made. However, the above data indicate that the major portion of tritiated water vapor is carried through the reprocessing cycle.

4. ^3H Gas - Tank Samples

The nine evacuated tanks samples which were analyzed for ^{85}Kr were also analyzed for gaseous ^3H. The technique used for this analysis was oxidation of the hydrogen gas with copper-oxide at elevated temperature and then recovery of the water which was counted by liquid scintillation techniques. The recovery procedure has not, at this time, been calibrated to insure predictable recovery of the oxidized hydrogen. Yield data are therefore questionable and are not included in this report. However, the activity/gram of water formed for all samples has been determined. From this, it can be concluded that ^3H gas was present during dissolution. Table 7 shows the activity per gram of water of each sample analyzed. Although the yields of hydrogen gas obtained have large errors, the apparent activity associated with ^3H in the gas phase is quite small compared with ^3H in the water vapor phase.

Table 7. Specific activity of ^3H gas in stack effluent during dissolution

Date-tank no.	Time interval	Activity µCi/gm water-formed
6/12-1	1027-1103	1.3×10^{-4}
6/12-2	1110-1153	2.6×10^{-2}
6/12-3	1210-1325	3.8×10^{-3}
6/12-4	1330-1438	6.8×10^{-4}
6/14-1	1125-1155	5.4×10^{-4}
6/14-2	1155-1250	1.4×10^{-3}
6/14-3	1250-1340	9.0×10^{-2}
6/14-4	1345-1440	2.7×10^{-2}
6/14-5	1440-1525	2.8×10^{-3}

5. ^{129}I Ion Exchange Sampler

The iodine sampler functioned properly during the sampling of two dissolution cycles at the stack. However, the bubblers located upstream of the ion exchange resin removed a major portion of the gaseous ^{129}I before it reached the ion exchange sampler. The iodine removal ability of the bubbler was anticipated, but not in the magnitude that it occurred. Both the bubblers and ion exchange samplers were analyzed for ^{129}I recovery. Results showed that the bubbler retained 60 to 90% of the ^{129}I sampled.

Two ion exchange and bubbler samplers were used during each dissolution. Table 8 shows the average stack ^{129}I concentrations during the two dissolution as well as the total stack output activities for the sampling period. The measured stack outputs for the sampling period are as follows:

Batch #13 - 6/12/69	1.1×10^{-3} curies
Batch #14 - 6/14/69	2.2×10^{-3} curies

These values are greater than predicted output (Table 1). This was expected since the iodine scrubbers were not operating during the dissolution cycles monitored. The output activities represent between 5 and 10% of the total ^{129}I available from the dissolved fuel. The stated output activities, based on sample analysis, indicate the minimum output that occurred during the dissolutions since losses probably occurred in the piping and ductwork between the stack and the sampler.

Table 8. ^{129}Iodine - measured activities at the stack

Date-sample[a]	Time interval	Avg. Concentration[b] µCi/cc	Stack output µCi
6/12-1	0845-1200	3.9×10^{-9}	8.4×10^{2}
6/12-2	1210-1400	2.5×10^{-9}	3.1×10^{2}
6/14-1	1045-1340	7.3×10^{-9}	1.4×10^{3}
6/14-2	1342-1535	6.2×10^{-9}	7.9×10^{2}

[a] Composite of ^{129}I in bubbler and ion exchange sampler

[b] Estimate of overall error S_c <20%.

6. Particulates - Filter Sampler

Four filters were obtained from NFS for analysis of particulate activity. The filter sampling intervals as well as the number of batches and weight of fuel chopped and dissolved are given below:

Filter sampling interval	Batches	Metric tons fuel
6/5 - 6/12/69	2	∿1.8
6/12 - 6/19/69	3	∿2.9
6/19 - 6/26/69	3	∿2.7
7/3 - 7/10/69	2	∿1.1

Table 9 gives activity per cc of air sampled for gross α, β and specific radionuclides. All activities, in the stack with no dilution, are less than the "allowable concentrations for unrestricted area" (10 CFR 20 PAR 20.106) which indicates that levels at the NFS property line would be quite small.

B. FIELD DATA

1. ^{85}Kr - Ionization Chambers

Ionization chambers, operated in the field during the NFS study, used a charcoal trap upstream of the chamber to remove radon from the gas stream. the 130 gm charcoal traps were maintained at dry ice temperatures, -78° C, throughout the run to insure holdup of the radon. The radon traps were not calibrated prior to the NFS study, so to insure complete holdup of the radon, the trap size was overestimated. Data results indicate that because of the overestimate, the ^{85}Kr holdup time in the charcoal trap increased creating a twofold effect. First the krypton concentration peaks were shifted on a time scale by 30 minutes. Second, the peaks were broadened to a large extent. It is impossible to reconstruct the ^{85}Kr input peaks from altered output peaks of this nature.

Subsequent calibration of the charcoal traps indicated 20 grams of charcoal at -78° C will hold up radon for 34-48 hours while the ^{85}Kr holdup was reduced to approximately 28 seconds. This is an acceptable holdup time which will allow proper analysis of the input ^{85}Kr peaks observed in the field with the environmental monitoring ionization chamber.

2. ^{85}Kr - GM Detectors

Typical response curves for field GM detectors are shown in figures 7, 8, and 9. Figure 7 is the detector response for station 2 on 6/12/69. The detector used was the Amperex 18546 and the curve

Table 9. Air particulates from NFS stack

	μCi/cc $\pm$ 3 s_c[a]			
Radionuclides	6/5 - 6/12	6/12 - 6/19	6/19 - 6/26	7/3 - 7/1
Gross α	$2.8 \pm 0.3 \times 10^{-14}$	$6.5 \pm 0.7 \times 10^{-14}$	$6.6 \pm 0.4 \times 10^{-14}$	N.A.[b]
Gross β	$3.8 \pm 0.1 \times 10^{-12}$	$1.0 \pm 0.5 \times 10^{-11}$	$1.3 \pm 0.5 \times 10^{-11}$	$2.0 \pm 0.1 \times 10^{-11}$
^{106}Ru	$3.1 \pm 0.4 \times 10^{-13}$	$1.1 \pm 0.1 \times 10^{-12}$	$7.6 \pm 0.8 \times 10^{-13}$	$1.6 \pm 0.1 \times 10^{-12}$
^{137}Cs	$7.4 \pm 0.2 \times 10^{-13}$	$2.1 \pm 0.1 \times 10^{-12}$	$1.8 \pm 0.1 \times 10^{-12}$	$2.6 \pm 0.1 \times 10^{-12}$
^{134}Cs	$2.0 \pm 0.1 \times 10^{-13}$	$5.3 \pm 0.2 \times 10^{-13}$	$6.2 \pm 0.2 \times 10^{-13}$	$8.6 \pm 0.2 \times 10^{-13}$
^{90}Sr	$2.4 \pm 0.6 \times 10^{-13}$	$6.2 \pm 0.9 \times 10^{-13}$	$8.1 \pm 1.5 \times 10^{-13}$	$1.0 \pm 0.1 \times 10^{-13}$
^{144}Ce	$1.2 \pm 0.2 \times 10^{-12}$	$3.0 \pm 0.9 \times 10^{-12}$	$4.3 \pm 0.4 \times 10^{-12}$	$4.6 \pm 0.6 \times 10^{-12}$
^{95}Zr/Nb[c]	$< 3 \times 10^{-13}$	$< 3 \times 10^{-13}$	$< 3 \times 10^{-13}$	$6.5 \pm 0.8 \times 10^{-13}$
^{54}Mn	$< 1 \times 10^{-13}$	$< 1 \times 10^{-13}$	$< 1 \times 10^{-13}$	$1.9 \pm 0.8 \times 10^{-13}$
^{60}Co	$3 \pm 2 \times 10^{-14}$	$1.8 \pm 0.2 \times 10^{-13}$	$6 \pm 2 \times 10^{-14}$	$1.2 \pm 0.1 \times 10^{-12}$
^{238}Pu	$5.9 \pm 2.0 \times 10^{-15}$	$1.6 \pm 0.3 \times 10^{-14}$	$9.0 \pm 3.0 \times 10^{-15}$	$2.7 \pm 0.4 \times 10^{-14}$
^{239}Pu	$1.7 \pm 0.3 \times 10^{-14}$	$3.2 \pm 0.4 \times 10^{-14}$	$2.2 \pm 0.4 \times 10^{-14}$	$3.9 \pm 0.6 \times 10^{-14}$
^{234}U	$< 2 \times 10^{-15}$	$< 2 \times 10^{-15}$	$< 2 \times 10^{-15}$	$< 2 \times 10^{-15}$
^{238}U	$< 2 \times 10^{-15}$	$< 2 \times 10^{-15}$	$< 2 \times 10^{-15}$	$< 2 \times 10^{-15}$

[a] When the sample count rate did not exceed the 3 s_c counting error, the activity was considered to be non-detectable and a less than value is reported.

[b] A heavy black deposit on the filter made gross alpha impracticable.

[c] Calculated concentration assumes equilibrium of ^{95}Zr--^{95}Nb at time of collection.

was generated based on two minute average from the ratemeter output. Figures 8 and 9 show the responses of two detectors located at station 2 on 6/14/69. It is interesting to note the similarity of these two curves. Figure 8 is the response of the Amperex 18546 in a face-up position, 3 feet above the ground. Figure 9 is the response of the EON 8001T in a vertical position (i.e., the detector was facing the stack). This detector was also three feet above the ground. The two detectors were separated by 15 feet. The similarity of the response curves indicates the plume was well mixed at the sampling station during the sampling interval. Table 10 shows the ^{85}Kr levels derived from such curves at the field sampling stations during the two dissolution cycles monitored.

The average activity was determined by integrating peak values and averaging these values over the sampling time interval. Peak activity is the maximum activity detected during the sampling interval based on two minute observation. The values of activity for station 2 on June 12, 1969 underestimate the average activity because sampling did not commence until 38 minutes after dissolution began, and therefore a portion of the ^{85}Kr cloud had already passed by the sampling station.

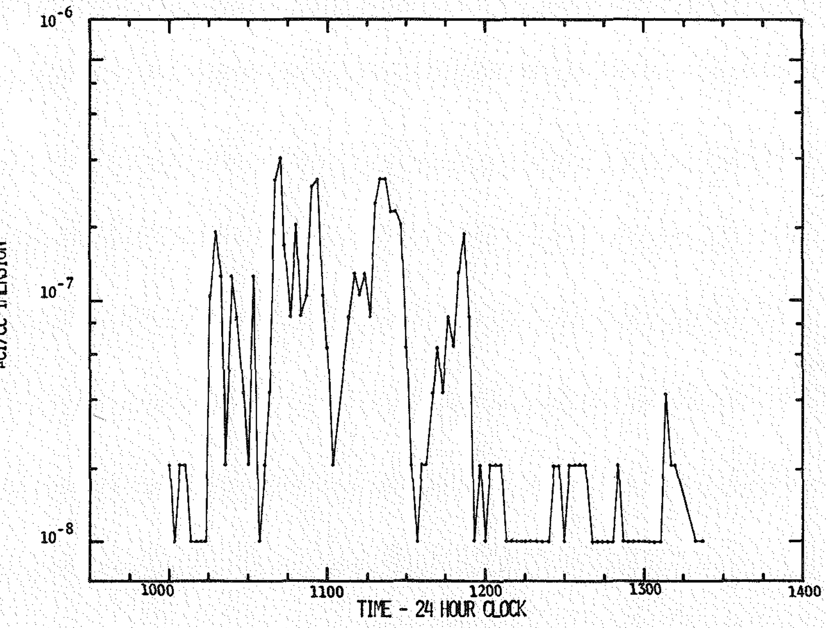

Figure 7. ^{85}Kr response of Amperex 18546 detector, field station #2 6/12/69

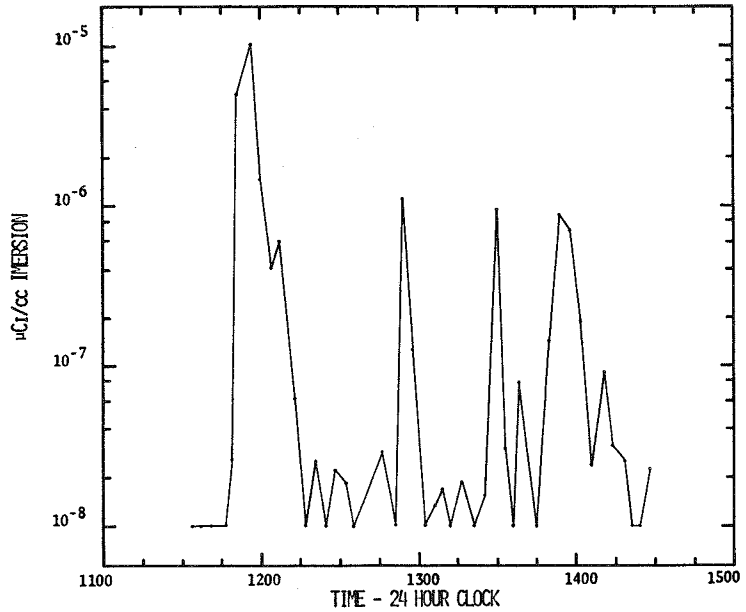

Figure 8. ^{85}Kr response of Amperex 18546 detector, field station #2 6/14/69

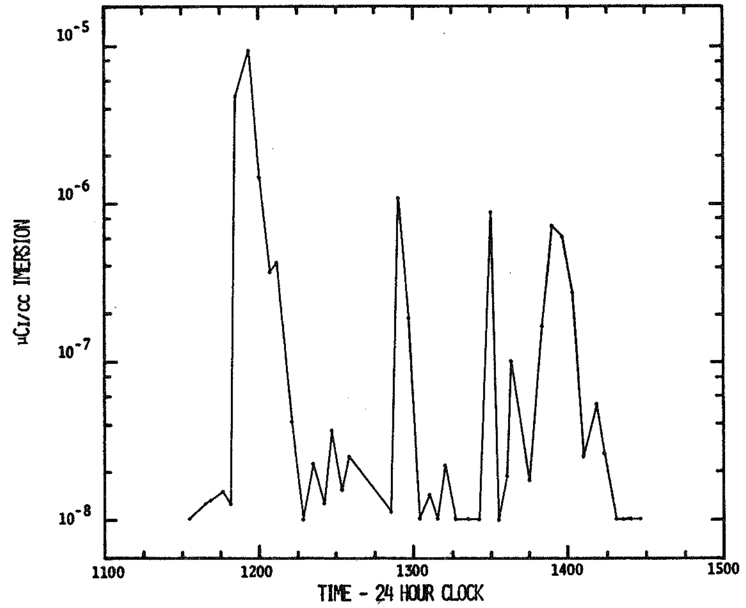

Figure 9. ^{85}Kr response of Eon 8001T detector, field station #2 6/14/69

Table 10. GM detectors - ^{85}Kr field levels

Date-station	Time interval	Avg. activity ± 2 S_t[a]	Peak activity ± 2 S_t
6/12 - 2[b]	1000-1320	6.4×10^{-8} ± 13%	3.9×10^{-7} ± 10%
6/12 - 3[c]	1048-1320	1.7×10^{-8} ± 14%[d]	1.3×10^{-7} ± 14%
6/14 - 2[c]	1145-1427	4.8×10^{-7} ± 11%	9.3×10^{-6} ± 8%
6/14 - 4[b]	1145-1427	2.8×10^{-7} ± 11%	2.4×10^{-6} ± 8%

[a] Average activity for the time period indicated. Allowable continuous property line average is 1×10^{-7} μCi/cc for ^{85}Kr (10 CFR 150-20, Table 2)

[b] Amperex 18546 with ratemeter output

[c] EON 8001T with scaler output

[d] Underestimate - Monitoring started 38 minutes after start of dissolution.

3. ^{85}Kr - Tank Samples

Tanks were operated at the four field stations each day, and sampled during the early part of the dissolution cycle. On June 12, 1969, the tank size used was 34 liters; on June 14, the tank size was 8.2 liters. A "Drierite" trap was attached at the inlet to remove moisture from the sampled air. These tanks were operated in the same manner as the in-plant evacuated tanks, i.e., with needle valve controls to maintain a constant sampling rate.

Table 11 shows the average ^{85}Kr activities detected from the tank samples as well as the equivalent activities based on GM detector measurement. The tank values agree well with the GM equivalent values for two stations. Station 4 on June 14, 1969, shows disagreement between the tank and geiger values. Investigation shows that this tank ceased sampling during the middle of a large rapid upswing in activity at this station. This upswing makes the exact shut-off time critical in terms of a few seconds. Since the tank sampling times are known only to the nearest minute, a large variation in average activity is not surprising.

Table 11. ^{85}Kr activity - evacuated tank samples

Date-station	Time interval	Avg. activity[a] µCi/cc	Equivalent GM- Values[b] µCi/cc + 2 s_t
6/12 - 1	1010-1215	3.98×10^{-8}	---
6/12 - 2	1015-1210	1.18×10^{-7}	$1.1 \times 10^{-7} \pm 12\%$
6/12 - 3	1022-1220	7.65×10^{-7}	---
6/12 - 4	1025-1215	6.49×10^{-7}	---
6/14 - 1	1130-1232	2.52×10^{-8}	---
6/14 - 2	1138-1230	1.29×10^{-6}	$1.4 \times 10^{-6} \pm 8\%$
6/14 - 3	1120-1200	3.30×10^{-7}	---
6/15 - 4	1125-1154	3.01×10^{-7}	$7.7 \times 10^{-7} \pm 11\%$

[a] $2\ s_c \overset{=}{<} 20\%$

[b] For field stations at which equivalent geiger data available

4. ^{3}H Water Vapor -"Drierite" Traps

"Drierite" samples were obtained at one station during each of the two dissolution cycles observed. Tritiated water concentrations are given in terms of Ci per cc of ambient air. The concentration values were determined by two different methods. The first method relates the ^{3}H activity per cc of water to air and uses measurements of relative humidity at the time of sampling. The relative humidity value used for June 12, 1969 measurement was a daily average given by the U. S. Weather Bureau for the area. The value used on June 14, 1969, was a measurement obtained at the sampling site during dissolution. The second method relates total ^{3}H activity to the air volume sampled. Table 12 shows the value obtained by both methods. The data substantiates the fact that daily area relative humidity averages do not necessarily apply to local situations.

5. ^{3}H Gas - Evacuated Tanks

The eight field tank samples were analyzed for gaseous ^{3}H in the same manner as the analyses performed on the in-plant tank samples. There was no measurable tritium gas present in any of the samples.

Table 12. ^{3}H concentrations in sampled air

Date-station	Time interval	Average concentration μCi/cc ± 2 s_c	Method
6/12 - 3	1010-1417	1.44×10^{-11} ± 23%	Relative humidity[a]
		3.47×10^{-11} ± 19%	Air volume
6/14 - 2	0845-1435	3.37×10^{-11} ± 20%	Relative humidity[b]
		4.51×10^{-11} ± 19%	Air volume

[a] Based on U. S. Weather Bureau data for Buffalo.

[b] Measurement obtained at sampling site during the sampling run.

6. Particulate - High Vol. Filter Sampler

A particulate filter sampler was operated in the field during each dissolution observed. The filter used in the sampler was a polystyrene fiber mat with an efficiency of 99.9% for 0.3 µ particulates. The total air volume sampled each day was 363 and 430 cubic meters on June 12 and June 14, 1969, respectively. Analysis of the two filter samples showed no significant elevations in activity above background. This result is expected because of the low particulate activity levels observed at the stack.

C. METEOROLOGICAL DATA AND APPLICATIONS

1. Stack to Sampling Station Dilution Factors

Krypton measurements obtained simultaneously in-plant and in the field can be used to calculate dilution factors for the stack effluent. These factors can be compared to theoretical predictions of dilution based on diffusion equations.

The diffusion equation used to calculate the theoretical dilution factors for the NFS study was The Pasquill diffusion equation (9).

$$X = \frac{Q(10^{-6})}{\pi\sigma_y\sigma_z\mu} \exp\left[-\frac{1}{2}\left(\frac{y}{\sigma_y}\right)^2\right]\left[-\frac{1}{2}\left(\frac{H}{\sigma_z}\right)^2\right] \qquad \text{(Eq. 2)}$$

where: χ = concentration at sampling station (μCi/cc)
Q = stack output (μCi/sec)
μ = wind speed at the top of stack (meters)
σ_y = horizontal dispersion coefficient (meters)
σ_z = vertical dispersion coefficient (meters)
y = cross wind distance from plume centerline (meters)
H = effective stack height (meters)

σ_y and σ_z are dependent upon the downwind distance of the sampling station and the atmospheric stability during sampling. Stability conditions are often classified according to six basic categories, ranging from extremely unstable, Class A, to moderately stable, (inversion) Class F (10).

Equation 2 is applicable for short term observations when the following conditions exist: continuous emission from the stack; constant effluent concentration; field observations at ground level within 1 to 2 σ_y of the plume centerline; and stability conditions unchanging during the ground level observation period. These conditions were approached during a portion of each dissolution period and comparisons between actual measurements of dilution and theoretical predictions have been made.

Figure 10 contains source concentration and downwind direction information for the dissolution of batch #13 on June 12, 1969. Figure 10a shows the stack output concentration of ^{85}Kr versus time. Ten minute averages were used for the calculation. The output concentration was quite variable during the first half of the dissolution cycle but steadied to a small almost linear decrease during the second half of the cycle. With respect to stack output concentration, the best condition for determining the dilution factor existed from 1040 to 1200 hours. Figure 10b shows the down wind direction versus time. The solid line denotes down wind direction measured at the top of the stack and the broken line is wind direction as measured 5 meters above ground level in the vicinity of the field station.

Station #2 was at an azimuth of 30° (from figure 2) with respect to the plant stack. During the time period 10:40 to 12:00 hours, the average downwind direction from the stack was close to this azimuth. This means the plume centerline during this time period passed near station #2. The theoretical dilution factor was calculated for station #2 during the time interval of 10:40 to 12:00. Based on 10 minute averages the down wind direction during this period was at an azimuth of 25°. The stability condition throughout this period was "slightly unstable" or C stability. For these conditions, the parameters for the theoretical determination were as follows:

$Q = 3.61 \times 10^{5}$ µCi/sec from 1040 to 1200
$\mu = 5.4$ m/sec
$H = 60$ meters
$\sigma_y = 280$ meters
$\sigma_z = 160$ meters

The predicted station #2 concentration from Equation 2 was:

$$X = 2.9 \times 10^{-7} \ \mu\text{Ci/cc}$$

which gives a theoretical dilution factor of:

$$d = \frac{\text{Stack Conc.}}{\text{Station Conc.}} = 6.7 \times 10^{4}$$

The GM detector data for station #2 corrected for lagtime, i.e., from 1050 to 1210 showed average concentration of 9.8×10^{-8} µCi.

This gave a measured dilution factor of:

$$d_m = 1.9 \times 10^{5}$$

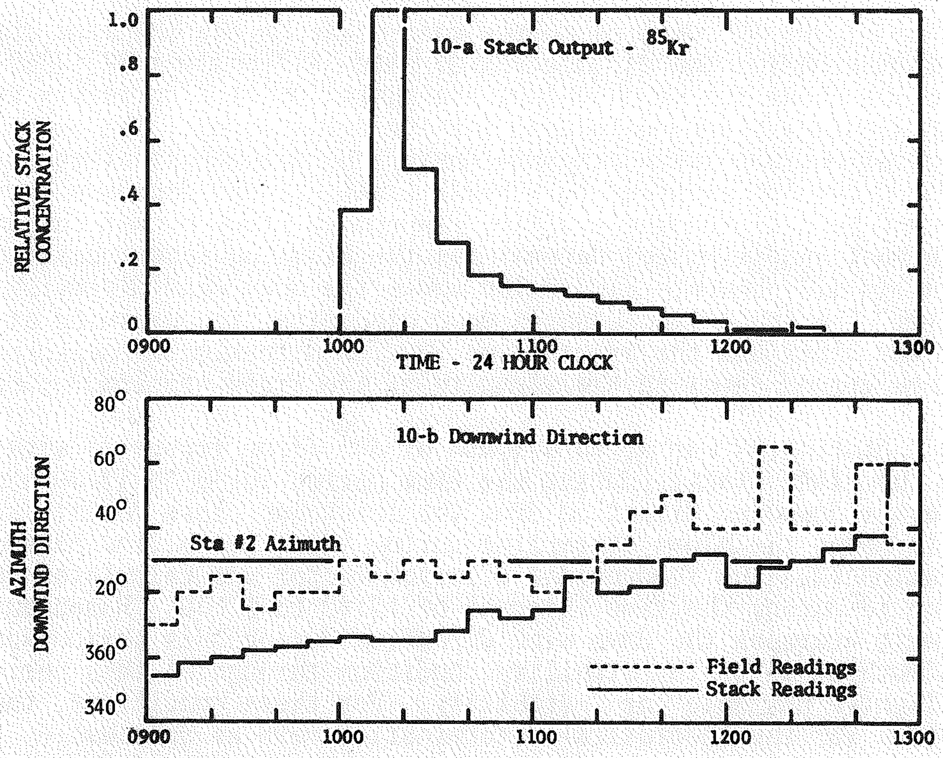

Figure 10. Meteorological data 6/12/69

The theoretical dilution factor is less than the measured value by a factor of three which is fairly good agreement for the first estimate. The first estimate did not take into account the effect of terrain and local alterations in wind direction between the stack and station #2.

During the dissolution of batch #14 on June 14, 1969, field sampling was done closer to the plant with few trees between the plant and the stations. The terrain was flat. A dilution factor was calculated during this run and compared to the theoretical estimate. Figure 11 shows the source concentration and wind direction information for this run. Figure 11a shows the relative stack conconcentration of ^{85}Kr versus time. Five minute averages were used because of the rapid fluctuations that occurred during the sampling period. The nearest the stack output concentration came to a steady state was during the time interval of 1245 to 1340. Figure 11b shows the downwind direction for five minute intervals. Again the solid line denotes measurements at the top of the stack and the broken line is wind direction as measured 5 meters above the ground in the vicinity of the field sampling stations (Refer to figure 2). Wind direction was erratic early in the sampling period but became more consistent during the time period 1245 to 1340. The wind direction at the stack

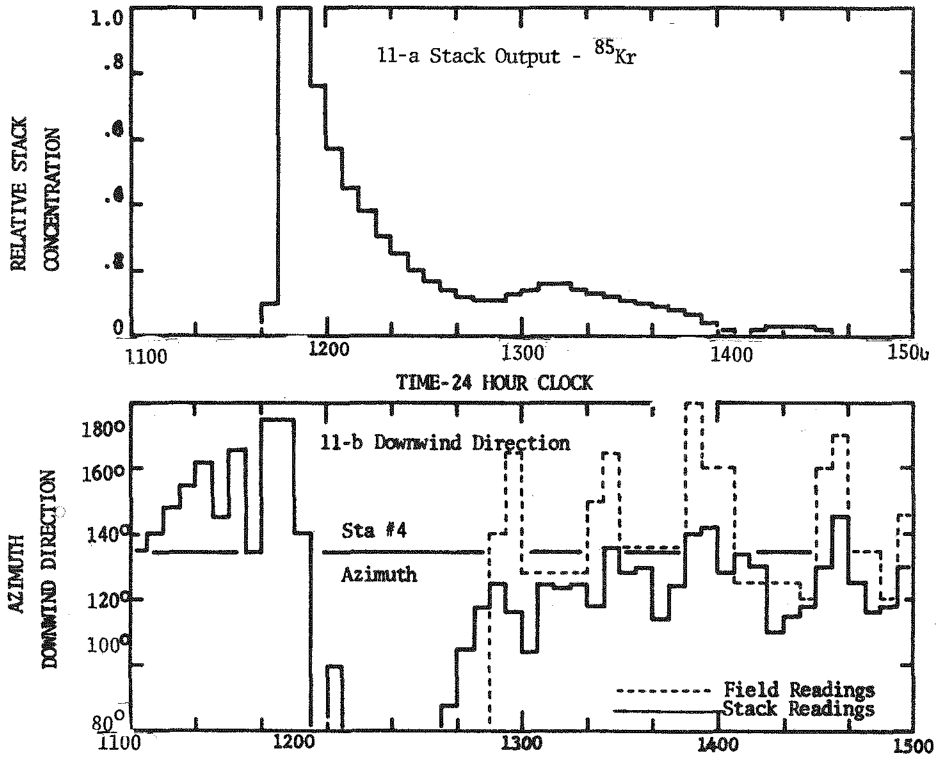

Figure 11. Meteorological data 6/15/69

height of 60 meters was to the southeast and the wind direction 5 meters above the valley floor was toward the south during this time period. Station #4, located at an azimuth of 135° with respect to the stack, was near the plume centerline during this time period and thus, this station position was used in the calculation of diffusion.

The dilution calculations were obtained for the time period 1245-1340. The average wind direction during this period was at an azimuth of 125°. The stability condition throughout this period was "slightly unstable" or C stability. For these conditions the parameters for the theoretical determination were as follows:

$Q = 2.6 \times 10^5$ μCi/sec
$\mu = 3.6$ m/sec
$h = 60$ meters
$y = 179$ meters
$\sigma_y = 120$ meters
$\sigma_z = 70$ meters

The predicted station concentration from Equation 2 was:

$$X = 6.1 \times 10^{-7} \text{ μCi/cc}$$

which gave a theoretical dilution factor of:

$$d = \frac{\text{Stack Conc.}}{\text{Stack Conc.}} = 2.2 \times 10^4$$

The GM detector data at station #4, corrected for lagtime, i.e., from 1250 to 1345, showed an average concentration of 3.2×10^{-7} μCi/cc. This gave a measured dilution factor of:

$$d_m = 4.2 \times 10^4$$

The theoretical dilution factor is less than the measured value by a factor of two. Again this is good agreement for the first estimate which did not account for terrain effects.

The associated error of the above theoretical predictions is large (30-60%) since the predictions are based upon wind speed, direction, and a visual estimation of stability as opposed to precise meteorological measurements and adjustments for local terrain.

The comparisons demonstrate that the actual dispersion conditions north and south of the plant do not differ significantly from predictions based on diffusion theory. Therefore, theoretical predictions can be used to give reasonable estimates of average concentration values of stack effluent in these two directions.

2. Predicted ^{85}Kr Concentrations Outside Exclusion Area

Average and maximum concentrations of ^{85}Kr outside the plant property can be determined using theoretical diffusion equations. The average concentration is defined as the yearly average concentration outside the property line in the predominant wind direction. The maximum concentration is the average 24 hour concentration outside the property line when a single dissolution occurs during which the meteorological conditions are such that minimum dilution occurs directly under the centerline of the plum.

Based on convention, and stability data obtained during the entire Yankee fuel reprocessing operation from May 1969 to July 1969, the average stability conditions are 33% B condition, 33% C condition and 33% E condition(7). The distances from the stack where the maximum ground concentration exist for these conditions are:

B stability - 380 meters
C stability - 670 meters
E stability - 3000 meters

The average property line distance from the stack is 1500 meters. This means that the maximum concentration occurs within plant property when B and C stability conditions occur. Therefore, the highest yearly average concentration outside plant property will occur between 1500 and 3000 meters depending upon which of the three conditions has the dominant effect upon annual dispersion. The predominant wind direction is from the south, based on wind rose data shown in Table 13. From this data, the largest average concentration will occur in the north octant with a frequency of 22%.

The average annual concentration can be calculated from the above information by use of the following equation (11):

$$\bar{\chi} = \left(\frac{2}{\pi}\right)^{\frac{1}{2}} \frac{fQ \times 10^{-8}}{\sigma_z \bar{\mu}\left(\frac{2\pi x}{n}\right)} \quad \exp\left[-\frac{1}{2}\left(\frac{H}{\sigma_z}\right)^2\right] \qquad \text{(Eq 3)}$$

where f = frequency of wind in sector
n = number of sectors
χ = down wind distance (meters)
all other parameters are the same as in Equation #2.

Assuming a reprocessing schedule of one metric tonne of 20,000 MWD fuel each day at a rate of 250 days per year with a yield of 6000 curies of ^{85}Kr per metric ton of fuel, the average stack output per second will be:

$$A = 4.7 \times 10^4 \ \mu\text{Ci/sec.}$$

Table 13. NFS wind rose data

Wind direction	Percent occurance (a)	(b)	(c)
N	8.5%	8%	3.3%
NE	3.0%	3%	0.5%
E	3.5%	3%	1.7%
SE	13.0%	16%	24.1%
S	22.0%	22%	36.8%
SW	19.0%	19%	8.4%
W	10.5%	10%	12.7%
NW	20.5%	19%	12.5%

(a) predicted by upper winds (2)
(b) stack observations for 1968 (7)
(c) Yankee Campaign, spring 1969 (7)

Table 14 shows the values used in Equation #3 and gives the down wind concentration at 1500 and 3000 meters for the three stability classifications. The highest average annual concentration occurs at the property line (1500 meters) in the north octant and is 1.7 x10^{-10} μCi/cc. This estimate is low with respect to the allowable level of ^{85}Kr in an unrestricted area which is 1 x 10^{-7} μCi/cc (10 CFR 150.20). The value of 1.7 x 10^{-10} μCi/cc is defined as the maximum average annual concentration for the population group located in the predominant down wind direction at a distance from the stack greater than 1500 meters but less than 3000 meters. This definition assumes the following:

a) The population is located in the defined area for a year.

b) The plant dissolution schedule is typical for the year (250 dissolutions of one tonne of 20,000 MWD per dissolution).

c) The stability conditions during dissolution have the distribution of 30% B, 30% c, and 30% E stability.

d) The actual dispersion of the stack effluent does not differ significantly from diffusion theory (equation 3) for the above stability conditions.

Table 14. Diffusion parameters for determination of annual ^{85}Kr concentration

Distance = 1500 meters (property line)

Stability	B	C	E
Q μCi/sec	4.7×10^4	4.7×10^4	4.7×10^4
$\bar{\mu}$ meters/sec	3	5	1
H meters	60	60	60
σ_z meters	170	88	28
f	0.22	0.22	0.22
n	8	8	8
$\bar{\chi}$ μCi/cc	1.3×10^{-10}	1.3×10^{-10}	2.5×10^{-10}

Annual Average $\bar{\chi} = \frac{1}{3} B + \frac{1}{3} C + \frac{1}{3} E = 1.7 \times 10^{-10}$ μCi/cc

Distance = 3000 meters

Stability	B	C	E
Q μCi/sec	4.7×10^4	4.7×10^4	4.7×10^4
$\bar{\mu}$ meters/sec	3	5	1
H meters	60	60	60
σ_z meters	360	170	43
f	0.22	0.22	0.22
n	8	8	8
$\bar{\chi}$	3.2×10^{-11}	3.9×10^{-11}	2.9×10^{-10}

Annual average $\bar{\chi} = \frac{1}{3} B + \frac{1}{3} C + \frac{1}{3} E = 1.2 \times 10^{-10}$ μCi/cc

The maximum average 24 hour concentration can be estimated using the same approach utilized in determining the maximum average annual concentration. At the present time the plant dissolves only once in a 24 hour period. Based on 1 metric tonne of 20,000 MWD fuel dissolved, the maximum stack output would be 6000 curies during a three hour period per day. The maximum down wind concentration will occur outside the property line if, during the entire dissolution period, E stability exists and the wind speed is 1 meter per second. If during these conditions the wind direction is stationary, the maximum average concentration for the three hour period will occur at a distance of 3,000 meters from the stack.

The concentration value is obtained using a modified form of equation 2 to account for a sampling time longer than a few minutes (10) (in this case, the sampling time is 3 hours). The three hour concentration value is 5.40×10^{-6} μCi/cc. Therefore, the maximum average 24 hour concentration is 6.8×10^{-7} μCi/cc.

The probability of an individual being at the location of the maximum concentration during an entire dissolution when all the necessary meteorological conditions exist as stated above (E stability, etc) is remote but possible. Until actual measurements are obtained the conservative approach would be to assume that the maximum individual in fact receives an exposure to 6.8×10^{-7} μCi/cc for 24 hours at least once a year.

CONCLUSIONS

A. INSTRUMENTATION AND SAMPLERS

Minimum detectable activities of all ^{85}Kr monitors were adequate for stack and field detection of ^{85}Kr during dissolution. The flow through ionization chamber proved to be a practical stack monitor. The flow through characteristic is preferred in stack sampling trains as opposed to GM detector which is an external monitor. High stack concentrations of ^{85}Kr eliminate the need for radon traps, and mechanical shock to the ionization chamber is not a problem since the stack sampling train is a stationary set-up. As a field monitor, an ionization chamber, even with the use of properly selected charcoal traps for radon retention, is not as practical as a GM detection system. The mobility of a sampling station is hindered by the bulk of the complete monitoring device which must include the ionization chamber, a drying bed, charcoal trap with dry ice, flow meters, and pump. Humidity in some field situations necessitates enveloping the ionization chamber and associated electronics in a plastic bag containing a drying agent.

The GM detector system used during the NFS study is very practical as a field unit. The system is compact, light weight, rugged, and not hindered by the normal humidity encountered in a field situation. It is quite mobile since it can be used with a light weight battery pack.

The evacuated tanks used for integrated gas sampling of ^{85}Kr and ^{3}H functioned well both in-plant and in the field. They are very useful when related to source strength but will give erroneous results when used in short term sampling situations under the conditions of rapidly changing source concentration.

The choice between bubblers and drying traps for sampling of tritiated moisture depends primarily on the concentration of tritium sampled and the mode of sample preparation preferred. The bubbler dilutes the tritiated water sample but it can be counted directly with liquid scintillation techniques. Drying traps, such as the "Drierite" trap, collect the sample without dilution and therefore are most useful at lower concentrations. However, the moisture collected must be extracted from the trap and collected before it can be analyzed. For short term sampling at the stack, the bubbler is preferred whereas the "Drierite" trap is best suited for long-term sampling in the field.

The ^{129}I ion exchange trap, although in itself is a very good sampling device, did not function properly in the NFS stack sampling train. This was because of bubbler interference and the large distance between sampling train and the stack. Due to the plating-out characteristics of iodine, representative samples can be obtained only when the ^{129}I sampler is placed in or near the stack sampling port as the first unit in the sampling train.

B. STACK EFFLUENT

The stack data from the field trip characterize the time and concentration behavior of the ^{85}Kr evolved during dissolution. Essentially all of the available ^{85}Kr is released during dissolution of the fuel. The ^{85}Kr releases observed agree with predictions based on yield data.

Tritium measurements at the stack show a large deficit when compared with the predicted stack output during dissolution. Field measurements of tritiated water during each dissolution gave a dilution factor of $\sim 10^4$, which approximates the dilution factors determined by ^{85}Kr measurements. This indicates that during the dissolution observed, evaporation from the low level waste lagoons located in the plant complex contributed very little tritium compared with stack output.

The ratio of gas to water-bound tritium released during dissolution has not been determined but preliminary data indicates that gaseous ^{3}H is less than 10% of the total ^{3}H released.

The ^{129}I results indicate that stack releases are elevated above levels expected when the iodine scrubbers are in operation which was expected. The extent of the elevated levels can not be determined from present data.

C. METEOROLOGICAL CONSIDERATIONS

The comparison between predicted diffusion of ^{85}Kr and the actual diffusion observed during two dissolution cycles is based on one observation during each dissolution. It is concluded from these two observations, that actual plume dispersion north and south of the plant does not differ significantly from the calculated theoretical diffusion pattern based on standard meteorological equations. More measurements are necessary to verify this conclusion for the other stability conditions existing at NFS during fuel reprocessing operations.

The approach used for estimating long term (annual) concentrations of ^{85}Kr in the predominant wind direction is as follows: Wind rose data employed were based upon long term observations. Characterization of the stability conditions were obtained from observations of local conditions during the sampling periods. The predominant wind direction conformed with the valley center line which permitted the use of an average dispersion model since the terrain is reasonably flat for some distance beyond the plant boundary in this direction. Better estimates of diffusion could be obtained with more observations of the stability conditions, in order to obtain precise annual stability distributions.

Data are not available on dispersion east and west of the plant. Due to the rapid rise of the valley walls in these two directions, use of dispersion equations is not valid. It is conceivable that the plume, when carried east or west of the plant, would collide with the valley walls and yield a dose of ^{85}Kr from one dissolution cycle that would give a larger annual concentration level than estimated for the predominant wind direction north of the plant. The only way to find out what actually occurs east or west of the plant is by measurement.

D. RECOMMENDATIONS

Based upon data and findings of this report, it is recommended that the following activities and studies be undertaken.

1. Iodine scrubbers should be used in the offgas lines of a fuel reprocessing plant regardless of fuel age in order to remove the long lived ^{129}Iodine.

2. Stack output characterization should be extended to better define ^{129}I and ^{3}H outputs. Investigation of other possible pathways for introducing ^{3}H into the environment should be undertaken to determine the relative contribution with respect to stack output.

3. Environmental measurements should be made with a suitable number of continuously monitoring ^{85}Kr stations in order to determine long term average concentrations.

4. The ^{129}I to stable iodine ratio, by measurement of suitable samples (e.q., bovine thyroid), should be obtained in the vicinity of NFS in order to determine the extent of ^{129}I buildup in the local environment.

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and in the environment. Four field sampling stations, located in the vicinity of the plant perimeter, and a stack sampler simultaneously monitored ^{85}Kr, ^{129}I, and ^{3}H (gaseous and water vapors) during two dissolution cycles. Particulates were monitored at the stack and one field station. Measurements are presented and discussed in terms of emission level versus specific plant operations, primarily the dissolution cycle. In addition, observed and theoretical dilution factors are compared and, based on meteorological considerations, show reasonable correlation. The instrumentation used in this study includes thin-window geiger detectors and flow-through ionization chambers for ^{85}Kr; bubblers, traps, and grab samplers for ^{3}H; and resin traps for ^{129}I. Choice of methodology and instrumentation is discussed with emphasis placed on a system usable in determining dose to a population in the plant vicinity.

KEYWORDS: Air Surveillance; Airborne Radioactivity; Environmental Radiation; Environmental Surveillance; Fuel Reprocessing Plants; Iodine 129; Krypton 85; Nuclear Facilities; Stack Sampling; Tritium.

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