ENVIRONMENTAL SURVEILLANCE AROUND NUCLEAR INSTALLATIONS

VOL. II

Proceedings
of a
Symposium
Warsaw
5-9 November 1973

PROCEEDINGS SERIES

ENVIRONMENTAL SURVEILLANCE AROUND NUCLEAR INSTALLATIONS

PROCEEDINGS OF A SYMPOSIUM
HELD BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
IN WARSAW, 5-9 NOVEMBER 1973

In two volumes

VOL.II

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 1974

FOREWORD

Adequate environmental surveillance is an essential part of the measures taken by the operators of nuclear installations and by the national competent authorities to ensure that such installations do not produce any pollution of the environment through the release of radioactive contaminants under normal operating conditions, and to provide information on which protective action can be taken in the event of an accidental release.

There has been argument and uncertainty about the nature and extent of the environmental surveillance programmes required for different types of nuclear installations; much of this is attributable to a lack of clear definition of the aims of such surveillance.

As part of its programme to assist Member States in controlling and minimizing the environmental effects of nuclear programmes, the International Atomic Energy Agency, in co-operation with the Government of Poland, convened a Symposium on Environmental Surveillance Around Nuclear Installations in Warsaw on 5 - 9 November 1973. It was attended by 200 participants from 26 Member States and by representatives of 8 international organizations. Sixty-one papers were presented in eight sessions covering the objectives of environmental surveillance, pre-operational investigations, environmental monitoring procedures in normal and emergency situations, the interpretation of results, research and supportive studies, and examples of the environmental surveillance programmes conducted at specific installations. A small number of the papers dealt with non-radioactive contaminants.

In the final session three short papers by invited speakers reviewed the problems arising in the establishment of standards and derived working limits and the operation of adequate environmental surveillance systems for both radioactive and non-radioactive contaminants that might be released to the environment in the nuclear industry. A panel of selected participants then discussed these topics and replied to questions submitted in written form and orally by the other participants.

It was emphasized that clear objectives should be set for any environmental surveillance programme in order to avoid wastage of resources in manpower and equipment. It is obviously more efficient in some cases to rely on the monitoring of gaseous and liquid effluents in order to check the compliance with the authorized limits on releases to the environment. However, as was pointed out by many speakers, it is not sufficient, especially in populated areas, to rely entirely on effluent monitoring. Properly designed environmental monitoring programmes are also needed to confirm that there are no unsuspected releases, or unsuspected pathways of exposure, and to reassure the public that adequate care is taken for their protection. Environmental measurements can also contribute to increased scientific knowledge of the behaviour of radionuclides in the environment.

The present book contains all the papers and discussions of the Symposium. The Agency gratefully acknowledges the assistance and co-operation of the Polish authorities, which helped greatly towards the success of the meeting.

EDITORIAL NOTE

The papers and discussions incorporated in the proceedings published by the International Atomic Energy Agency are edited by the Agency's editorial staff to the extent considered necessary for the reader's assistance. The views expressed and the general style adopted remain, however, the responsibility of the named authors or participants.

For the sake of speed of publication the present Proceedings have been printed by composition typing and photo-offset lithography. Within the limitations imposed by this method, every effort has been made to maintain a high editorial standard; in particular, the units and symbols employed are to the fullest practicable extent those standardized or recommended by the competent international scientific bodies.

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RESEARCH AND SUPPORTIVE STUDIES

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ENVIRONMENTAL LEVELS OF IODINE-129

J.M. MATUSZEK, J.C. DALY, S. GOODYEAR,
C.J. PAPERIELLO, J.J. GABAY
Radiological Sciences Laboratory,
New York State Department of Health,
Albany, N.Y., United States of America

Abstract

ENVIRONMENTAL LEVELS OF IODINE-129.

Until recently, ^{129}I releases to the environment have not been considered a serious problem by the nuclear industry. The environmental safety analyses for all private nuclear power facilities presently operating in the United States of America limit the halogen gas-effluent releases only for ^{131}I concentration. Controls for ^{129}I releases are not included in the Technical Specifications. Data collected since 1969 by the New York State Departments of Health and Environmental Conservation indicated significant concentrations of ^{129}I in deer, bovine and small animal thyroids collected from the environs of a fuel reprocessing facility in New York State. Animal thyroid levels ranged up to 3700 pCi/g tissue. Surveillance for ^{129}I in milk was begun in September 1971. The levels in milk samples collected from holding tanks of individual dairy herds surrounding the nuclear fuel reprocessing plant were as high as 2.3 pCi/litre. The milk levels decreased in the spring, apparently because the cattle were shifted from contaminated feed to open pasture. Water samples collected off site since June 1971 from streams draining the reprocessing plant site showed ^{129}I concentrations up to 8.3 pCi/litre. The long half-life (17 million years) for ^{129}I causes any release to become a permanent biospheric contaminant, permitting wide-ranging contamination of many food products. Milk products, such as condensed milk or formula preparations for infants, would be of special concern, since ^{129}I levels would not be reduced by physical decay and may actually be increased by milk processing operations. Micro-dosimetric calculations based on the high-LET emissions in ^{129}I decay result in dose estimates as high as 800 mrem/a at the cell/colloid interface of an infant thyroid were the child to consume milk at the observed concentrations. Estimates of the dose to human thyroid based on ^{129}I activity levels equivalent to those found in animal thyroids would result in values up to 6300 mrem/a at the cell/colloid interface. Activity levels of ^{129}I in fish flesh indicate that the fish→man pathway could prove dose-limiting. Recommendations are offered for reducing the potential public health hazard.

1. INTRODUCTION

The first privately owned nuclear fuel reprocessing plant, Nuclear Fuel Services, Inc. (NFS), began operations in West Valley, New York, in April 1966. It served as the pilot plant for a newly developing private industry, the reprocessing of spent fuel from nuclear power reactors. A waste burial facility is also operated on the site. NFS has temporarily suspended dissolutions since December 1971 to renovate and expand the capacity of the plant.

The New York State Department of Health (NYSDH) expanded its routine radiological surveillance in 1966 to include monitoring of NFS [1]. Responsibility for this surveillance has since been transferred to the New York State Department of Environmental Conservation (NYSDEC), but the analytical program remains with the Radiological Sciences Laboratory (RSL) in the Health Department.

Federally owned fuel reprocessing plants which had been in operation for several years served as models for the discharge limits to be applied to NFS. Technical Specification control limits [2] for radioiodine releases were based on ^{131}I, a limitation which permitted a ready alternative for dissolver off-gas treatment when the silver nitrate reactors failed to operate acceptably [3]. A chemical scrubbing system with added mercuric nitrate, though far less efficient for radioiodine removal, could be used [3] if fuel elements were "cooled" to permit the ^{131}I levels to decay to sufficiently small quantities. Apparently the NFS staff and the U. S. Atomic Energy Commission (AEC) felt that the 1.7×10^7 year half-life of ^{129}I would render its specific activity in the environment so low as to be inconsequential. The assumption now appears incorrect.

In December 1968, NFS began to process high-burn, commercial power-reactor fuel. This step heightened the State's concern for potential impact of ^{129}I releases to the environment. The program for ^{129}I measurements was expanded, and the AEC was urged to include control limits for ^{129}I release in the NFS Technical Specifications [5]. A forthcoming plant expansion, coupled with greater burn-up for fuel from newer power reactors, will further increase the potential health hazard. Surveillance has therefore been continued during shutdown in order to obtain baseline data on ^{129}I levels. The U. S. Environmental Protection Agency (EPA) has provided important technical help, including sample analyses and supplemental research programs [6-8], as well as some financial support.

2. MEASUREMENTS

2.1 Methodology

This report includes data from measurements made by the RSL [9]; the EPA's Northeastern Radiological Health Laboratory in Winchester, Massachusetts [8]; Teledyne Isotopes (a private research laboratory holding contracts with NYSDH and NYSDEC)[9]; and the University of Buffalo (under contract with NYSDEC) [9]. All sample collections were made by New York State staff (either NYSDH or NYSDEC) except for six bovine thyroids collected by EPA in Boston, Massachusetts, for baseline measurements. Lack of a research reactor in the Albany area has necessitated the use of the EPA and the University of Buffalo laboratories for neutron activation analysis.

The general experimental techniques for neutron activation analysis and liquid scintillation counting have been described previously [8,10,12,13]. Standard spectrometric methods were used for direct sample counting in a NaI(Tl) well-crystal or on an intrinsic-Ge solid-state detector.

Neutron activation analysis is the most specific and most sensitive assay for ^{129}I, but because at its relatively high cost and the requirement of proximity to a reactor, liquid scintillation counting or direct counting on a NaI(Tl) detector is desirable for surveillance purposes. The latter techniques, however, are less sensitive and suffer from ^{131}I and ^{125}I interference, the ^{125}I being particularly difficult to resolve because of its 60-day half-life.

The high-resolution, intrinsic-Ge detector appears to provide a better alternative to activation analysis for surveillance measurements. It is as specific for ^{129}I as activation analysis. Though it is less sensitive, its sensitivity has recently been improved at the RSL by using a radiochemical separation method, with solid-source sample mounting [10]. All ^{129}I (and ^{125}I) measurements at the RSL are now performed with the intrinsic-Ge detector, either directly or with preconcentration.

Standardization and intercalibration of counting systems was a problem until recently due to the lack of an adequate ^{129}I standard. The U. S. National Bureau of Standards has now provided a certified ^{129}I standard, which was used to recalibrate the RSL counting systems and on which all of the RSL results in this report have been recalculated, making them 8% higher than previously reported [9,10,15]. All of the EPA results appear to be a factor of 1.8 lower than the values calculated from the NBS standard, but they have not been corrected in this report.

Sample activity levels are considered to be below the detection limit of the system (< values) when the error at the 95% confidence level (2σ) is greater than or equal to the calculated value.

The analytical data in Tables I-VII[1] include counting errors measured for each result except those from the EPA. The EPA laboratory did not report any statistical errors for individual values, only a single error estimate for all values reported. For convenience, that total error estimate is here applied to individual EPA measurements.

2.2 Sample Selection

Deer thyroids (Table I) were first collected as environmental indicators of the NFS site area because they were readily available from deer killed for other measurements, such as ^{90}Sr and ^{137}Cs. Though they serve well as early indicators, it is difficult to differentiate between gaseous and liquid NFS discharges of ^{129}I on the basis of deer thyroid results [9,11], since the high fences around the NFS site proved only a minor barrier to deer desiring access to the creeks below the outfall of the plant or even to the various lagoons on the site.

Small animals, such as rabbits and woodchucks, have a more limited forage range, and ^{129}I levels in their thyroids would be a better indicator of the impact of stack discharges at each location (Table II) [9].

Water samples (Tables III and IV) might ordinarily serve as dietary as well as environmental indicators. However, no public water supplies draw on the stream system collecting NFS effluents.

[1] The dates in the tables are given in the order month/day/year.

TABLE I. ^{129}I ACTIVITY LEVELS IN DEER THYROIDS

Date Collected	Location (miles)	^{129}I (pCi/g)
9/24/68[a]	onsite	<80
9/25/68[a]	"	120±80
9/25/68[a]	"	<80
3/18/69[a]	"	<80
3/18/69[a]	"	180±90
3/18/69[a]	"	210±100
3/18/69[a]	"	<80
3/18/69[a]	"	<80
10/7/69[a]	"	1,200±200
10/8/69[a]	"	<80
3/18/70[a]	"	3,700±300
3/18/70[a]	"	850±90
3/19/70[a]	"	2,500±200
3/19/70[a]	"	<80
3/20/70[a]	"	3,100±300
8/19/70[a]	4.5 NE	<80
9/16/70[a]	3.5 N	<80
12/2/70[a]	onsite	<80
12/3/70[a]	"	<80
12/3/70[a]	"	580±90
12/3/70[a]	"	<80
12/3/70[a]	"	250±40
12/3/70[a]	"	<80

Date Collected	Location (miles)	^{129}I (pCi/g)
3/23/71[a]	onsite	480±50
3/23/71[a]	"	<80
3/23/71[a]	"	240±20
3/23/71[a]	"	420±40
3/23/71[a]	"	1,400±100
3/23/71[a]	"	620±60
11/15/71[b]	"	180±4
11/15/71[b]	"	194±3
12/8/71[b]	"	920±6
12/8/71[b]	"	712±4
12/8/71[b]	"	450±5
12/17/71[b]	1.0 SE	694±5
1/22/72[b]	18 NNW	33±1
3/21/72[b]	onsite	360±5
3/21/72[b,c]	"	313±6
3/21/72[b,c]	"	322±6
3/22/72[b]	55 WSW	0.4±0.2
6/16/72[b]	3.3 SE	0.5±0.1
7/7/72[b]	7.0 WNW	1.0±0.2
12/19/72[b]	onsite	64±6
12/19/72[b]	"	27±3
3/27/73[d]	"	20±3
3/27/73[d]	"	65±3

Notes:

a. Liquid scintillation counting - Teledyne Isotopes.
b. NaI(Tl) well-crystal counting - RSL.
c. Second count one year later showed no identifiable decay from ^{125}I.
d. Intrinsic-germanium detector counting - RSL.

Fish and algae samples (Table V) provide information concerning ^{129}I buildup in the aquatic environment. Fish also appear to contribute a significant dose as part of the local diet (see 3.3).

Because the impact of ^{129}I contamination of food is expected to be greatest on infants [13], the grass-cow-milk pathway was considered likely to be dose-limiting [9]. Bovine thyroids (Table VI) were collected in addition to the small animal thyroids to provide indicators of contamination of dairy farms by ^{129}I from the stack effluents from NFS.

The importance of the cow-milk pathway for infant thyroid doses from ^{129}I led to the development of a surveillance method for ^{129}I levels in milk [10]. Results from analysis of milk and water samples have been reported [9,11,15], but the milk data are summarized here (Table VII) as a convenience for this discussion. To facilitate comparison of ^{129}I concentrations as a function of distance from the NFS, the data are arranged primarily by location rather than date.

TABLE II. ^{129}I ACTIVITY LEVELS IN SMALL ANIMAL THYROIDS

Sample	Date Collected	Location (miles)	Method	^{129}I (pCi/g)
Rabbit[a]	12/14-18/70	0.2 to 6	LS(TI)[d]	<80
Rabbit	12/22/70	1.8 NW	"	1,100 ± 200
Rabbit[b]	12/22/70-1/26/71	0.2 to 3	"	<80
Woodchuck	4/19/71	0.2 NW	NA(EPA)[e]	210 ± 16
Woodchuck	4/19/71	1.8 NW	"	2 ± 1
Rabbit	4/29/71	1.2 NE	"	52 ± 4
Rabbit	5/3/71	1.8 NW	"	91 ± 7
Woodchuck	5/7/71	2.8 WNW	"	10 ± 2
Woodchuck	5/7/71	1.2 NE	"	39 ± 3
Woodchuck	5/12/71	1.0 NW	"	30 ± 3
Woodchuck	6/22/71	1.8 NW	"	24 ± 3
Rabbit	12/16/71	1.0 NW	"	0.53 ± 0.04
Rabbit	12/16/71	1.0 NW	"	6.2 ± 0.5
Rabbit	12/29/71	1.8 NW	"	4.5 ± 0.3
Rabbit	2/28/72	2.0 S	"	2.5 ± 0.2
Woodchuck	8/4/72	7.5 E	NA(UB)[f]	<3.7
Woodchuck	8/4/72	1.9 NW	"	<2.4
Woodchuck	8/4/72	1.9 NW	"	<2.3
Rabbit[c]	3/2/73	2.1 NW	"	<1.5

Notes:

a. Six separate samples.
b. Seven separate samples.
c. Composite of three samples.
d. Liquid scintillation counting-Teledyne Isotopes.
e. Neutron activation analysis-EPA.
f. Neutron activation analysis-University of Buffalo.

2.3 Interferences

Care must be taken in interpreting the several sets of data included in the tables. As mentioned previously, activation analysis is most sensitive and specific, but results can be in error when ^{129}I levels become very small relative to ^{127}I concentrations.

The intrinsic-Ge detector has recently shown the presence of ^{125}I in liquid effluents from the burial site. All of the water samples collected earlier from Buttermilk Creek may therefore have contained undetected ^{125}I. However, the ^{125}I activity in recent samples is less than 0.5 pCi/liter, which should also be an upper limit for previous offsite water samples.

Deer thyroid samples collected onsite may also have contained some ^{125}I, but the effect is thought to be small. Samples were normally stored for several weeks until the collection period was completed (usually the end of the hunting season), and counting was generally 3 or 4 months after collection. Recounts of four deer thyroids several months after the initial analysis showed

TABLE III. ^{129}I CONCENTRATIONS IN ONSITE WATER SAMPLES

Sample	Date Collected	^{129}I (pCi/l)
PLANT WASTES		
Waste Lagoon	6/67	750 ± 60[a]
Erdman Brook	4/14/72	0.9 ± 0.3[b]
French Drain	5/8/72	7.3 ± 0.3
Surface Stream #1	5/8/72	0.6 ± 0.3
Surface Stream #2	5/8/72	0.4 ± 0.3
Weir	5/18/72	120 ± 10
Lagoon #3	10/5/72	53 ± 12
Lagoon #4	7/19/73	38 ± 13[c]
BURIAL SITE WASTES		
Burial Drainage	11/13/72-7/9/73(6)[d]	<0.3
Burial Lagoon	1/3/73	^{125}I[e]
Burial Lagoon	3/5/73	^{125}I[e]

Notes:
a. Liquid scintillation counting by Teledyne Isotopes.
b. All values by liquid scintillation counting at the RSL unless otherwise specified.
c. ^{125}I also found (see text).
d. Value in parentheses indicates the number of individual samples measured during the indicated period.
e. Only ^{125}I was found in these samples.

no identifiable change in activity. Several deer thyroids, analyzed by EPA's liquid scintillation method 3 months after measurement at the RSL, did not show statistically significant levels of ^{125}I.

Cattle and small animals at the locations from which samples were collected could not have had access to the burial lagoons. Therefore, bovine and small animal thyroids could not have contained ^{125}I nor could the milk samples.

3. RESULTS AND DISCUSSION

The data reported here were obtained on samples collected primarily by New York State staff and analyzed by the RSL, though analyses have also been performed by other laboratories. Some of the data have been used previously to describe the impact of NFS on the environment [8,9,11,15,16]. This report is a comprehensive evaluation of all the results obtained.

Any assessment of the environmental impact of fuel reprocessing is affected by several factors, including chronological and spatial patterns of deposition, adequate definition of the source level, and dose modeling.

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TABLE IV. ^{129}I CONCENTRATION IN OFFSITE SAMPLES

Date Collected	^{129}I (pCi/l)
Buttermilk Creek (2.7 miles downstream)	
10/19/70-1/4/71(10)[a]	<16[b]
6/7/71	<1.1[c]
6/14/71	1.5 ± 1.2
6/21/71	<2.0
6/28/71	<1.1
7/6/71	1.9 ± 1.1
7/19/71	2.1 ± 0.9
7/26/71	2.7 ± 1.7
8/2/71	<0.3
8/9/71	4.3 ± 0.3
8/16/71	7.2 ± 0.3
8/23/71	4.4 ± 0.3
8/30/71	4.6 ± 0.3
9/7/71	6.5 ± 0.3
9/13/71	4.4 ± 0.3
9/20/71	2.6 ± 0.3
9/27/71	2.1 ± 0.3
10/4/71	4.1 ± 0.3
10/12/71	4.2 ± 0.3
10/18/71	6.7 ± 0.3
10/26/71	4.1 ± 0.3
11/1/71	8.3 ± 0.3
11/8/71	1.5 ± 0.3
11/15/71	0.6 ± 0.3
11/22/71	1.2 ± 0.3
11/29/71	1.4 ± 0.3
12/6/71	1.0 ± 0.3
12/13/71	1.0 ± 0.3
12/20/71	1.1 ± 0.3
12/27/71	<0.3
1/3/72	<0.3
1/10/72	0.3 ± 0.3
1/24/72	<0.3
1/31/72	0.3 ± 0.3
2/7/72	0.8 ± 0.3
2/14/72	0.6 ± 0.3
2/22/72	0.7 ± 0.3
2/28/72	1.5 ± 0.3
3/6/72	1.1 ± 0.3
3/13/72	<0.3
3/20/72	<0.3
3/27/72	0.8 ± 0.3
4/3/72	<0.3
4/10/72	1.2 ± 0.3
4/17/72	<0.3
4/24/72	0.6 ± 0.3
5/1/72	0.6 ± 0.3
5/8/72	0.3 ± 0.3
5/15/72	<0.3
5/22/72	0.9 ± 0.3
5/30/72	1.5 ± 0.3
6/5/72	1.2 ± 0.3
6/12/72-1/8/73(28)	<0.3 to <0.9
1/5/73	3.1 ± 0.7
1/22/73-7/9/73(23)	<0.3 to 0.8
8/6/73	0.7 ± 0.1[d]
8/13/73	1.2 ± 0.2[d]
8/20/73	1.1 ± 0.4[d]
8/27/73	0.13 ± 0.08[d]
Cattaraugus Creek (5.6 miles downstream)	
11/4/70-12/30/70(28)	<16[b]
6/2/71-7/7/71(5)	<1
7/21/71	6.9 ± 0.4
7/28/72-7/3/73(98)	<0.3 to 0.9

Notes:

a. Values in parentheses indicate the number of individual samples measured during the indicated period.

b. Liquid scintillation counting by Teledyne Isotopes.

c. All values by liquid scintillation counting at the RSL unless otherwise specified.

d. ^{125}I also found (see 3.1 in text).