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MEASURING RADIOACTIVITY IN THE ENVIRONMENT – THE QUALITY OF THE DATA

RADIOACTIVE WASTE

KEYWORDS: *environment, accuracy, radiochemical analysis, radioactivity, milk, comparative evaluations, radiation monitoring, strontium-89, radioisotopes, iodine-131, strontium-90, cesium-137, barium-140*

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Laboratory intercomparison studies involving samples of most environmental media are conducted on a continuing basis by the U.S. Environmental Protection Agency's (EPA) National Environmental Research Center-Las Vegas. These studies enable the EPA to assess the precision and accuracy of radioassay procedures for environmental samples. The data from a recent six-month study to assess the quality of radionuclide measurements in milk indicate that the measurements of 30 federal, state, and private laboratories compare favorably with those of 10 reputable international laboratories. However, the data also indicate a need for improved analytical performance, the adoption of standard reference methods, more frequent instrument calibration, and better quality control in all radioanalytical procedures.

INTRODUCTION

Environmental radiation measurements are made daily by federal, state, local, and private agencies. The data obtained from these measurements are utilized by the U.S. Environmental Protection Agency (EPA), the U.S. Atomic Energy Commission (USAEC), and other agencies for such purposes as estimating dose and health effects, establishing standards and guides, and conducting regulatory activities. It is therefore imperative that the precision and accuracy of the data be assured so that policy decisions concerning environmental quality are based on valid and comparable data.

The quality assurance program of the EPA is designed to encourage the development and implementation of quality control procedures at all levels of sample collection, analysis, data processing, and reporting. As an integral part of the EPA's program, the Quality Assurance Branch of the National Environmental Research Center-Las Vegas (NERC-LV) distributes calibrated radionuclide solutions for instrument calibration and chemical yield determinations, and conducts a number of laboratory performance studies involving the analysis of radionuclides in environmental media. Any laboratory involved in environmental radiation monitoring may participate in these programs.

The intercomparison studies program enables participating laboratories to maintain checks on their analyses and assists them in documenting the validity of their data. In addition, this program enables the EPA to obtain an overall estimate of the precision and accuracy of environmental radiation measurements, or more precisely the precision and accuracy of laboratory radioassay procedures of environmental samples.

Studies currently in progress involve samples of most environmental media, including milk, air, water, soil, food, urine, and noble gases. Participants include state, federal, and international laboratories. There are also 18 nuclear facilities, or their contractors, participating in one or more of the cross-check programs. The continuing nature of these programs with numerous participants enables the Quality Assurance Branch to assess the quality of environmental data.

For the purpose of illustration, results of a milk cross-check program were chosen to demonstrate the need for and results of an on-going quality assurance program. Such a program is vital for all phases of environmental radiation monitoring, including the monitoring of waste disposal sites and procedures, the basic principle

TABLE I

Summary of Milk Analysis Data, July to December 1973

	July	August	September	October	November	December
Strontium-90						
Known pCi/liter	65	158	72	119	135	103
1 σ limit pCi/liter	3	8	4	6	7	5
1 σ/known %	4.6	5.1	5.6	5.0	5.2	4.9
N	25	14	20	23	22	22
$\bar{X}$ pCi/liter	61.0	148.2	74.2	106.1	115.2	93.1
s pCi/liter	12.9	21.8	22.2	19.9	27.0	16.4
s/known %	19.9	13.8	30.8	16.7	20.0	15.9
Cesium-137						
Known pCi/liter	116	122	84	91	126	101
1 σ limit pCi/liter	6	6	5	5	7	5
1 σ/known %	5.2	4.9	6.0	5.5	5.6	5.0
N	37	23	27	36	30	30
$\bar{X}$ pCi/liter	115.2	121.6	83.0	88.0	121.1	100.1
s pCi/liter	11.6	10.2	11.5	9.6	11.8	9.4
s/known %	10.0	8.3	13.7	10.5	9.4	9.3
Strontium-89						
Known pCi/liter	98	96	85	133	123	89
1 σ limit pCi/liter	5	5	5	7	6	5
1 σ/known %	5	5	6	5.3	4.9	5.6
N	21	14	14	19	14	14
$\bar{X}$ pCi/liter	86.5	61.5	74.5	113.2	107.4	85.9
s pCi/liter	25	29.58	14.9	24.3	19.9	12.5
s/known %	25.5	30.8	17.5	18.3	16.2	14.1
Iodine-131						
Known pCi/liter	115	144	123	493	104	177
1 σ limit pCi/liter	6	7	6	25	5	9
1 σ/known %	5.2	4.9	4.9	5.1	4.8	5.1
N	33	23	22	35	28	26
$\bar{X}$ pCi/liter	125	149.8	121.3	488.7	102.2	177.2
s pCi/liter	18.3	16.5	8.8	31.3	13.9	11.6
s/known %	15.9	11.5	7.2	6.3	13.4	6.5
Barium-140						
Known pCi/liter	148	152	146	104	0	0
1 σ limit pCi/liter	7	8	7	5		
1 σ/known %	4.7	5.3	4.8	4.8		
N	32	20	24	32		
$\bar{X}$ pCi/liter	153.8	161.6	155.3	107.4		
s pCi/liter	24.8	20.9	43.1	19.2		
s/known %	16.8	13.8	29.5	18.5		

being the same for all radiation programs. To illustrate the nature of the program and to indicate the current status of some environmental measurements, results showing what has been learned from the intercomparison study with milk are discussed.

PROCEDURES AND RESULTS

To assess the quality of milk analysis, data from the milk cross-check program were compiled for a six-month period beginning in July 1973 and ending in December 1973. Four-liter milk samples containing ^{89}Sr, ^{90}Sr, ^{137}Cs, ^{131}I, and ^{140}Ba were distributed each month to approximately 30 participating laboratories. All participants received samples containing these radionuclides; however, all did not submit results for each analysis. This resulted in a different number, N, for the various radionuclides. The data for these studies are summarized in Table I.

A comparison may be made between the experimental standard deviation, s, calculated from the data supplied by the participating laboratories, and the standard deviation, σ, expected for various analyses listed in Table II.

TABLE II

Limits Established by the Analytical Quality Control Service (Refs. 1 and 2)

Radionuclide	1 σ Limits for Single Determination	
	For Levels ≤100 pCi/liter (pCi/liter)	For Levels >100 pCi/liter (%)
^{131}I	5	5
^{137}Cs	5	5
^{140}Ba	5	5
^{89}Sr	5	5
	For Levels ≤30 pCi/liter	For Levels >30 pCi/liter (%)
^{90}Sr	1.5	5

For the purpose of comparison, the experimental and expected standard deviations, expressed as a percent of the known values, are shown for each month in Table I. These data were compared with those from a recent study conducted by the World Health Organization, International Reference Center for Radioactivity (IRC) (Ref. 3) in which 10 international laboratories participated in an intercomparison study with milk containing ^{90}Sr and ^{137}Cs. The IRC data are summarized in Table III.

TABLE III

Results for a Liquid-Milk Intercomparison Study by the World Health Organization, International Reference Center

General Mean (pCi/liter)		Sample No. A 1010		Sample No. A 338	
		^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
		39.0	126	19.3	23.9
Standard Deviation	pCi/liter	8.9	14	3.1	7.6
	%	23	11	16	32

Data compiled by the Quality Assurance Branch at NERC-LV (Table IV) reveal that during the six-month period, from July to December 1973, 30% of the participants were within the ±3σ control limits for the ^{89}Sr analysis, 45% for ^{90}Sr, 48% for ^{140}Ba, 63% for ^{131}I, and 69% for ^{137}Cs.

TABLE IV

Results of Milk Cross-Check Analyses, July to December 1973

σ Limits	Radionuclide				
	^{89}Sr	^{90}Sr	^{131}I	^{140}Ba	^{137}Cs
	Laboratories Within σ Limits (%)				
≤1	18.5	18.0	25.0	23.9	34.7
≤2	23.1	38.0	47.9	39.6	55.4
≤3	30.1	45.0	63.2	47.8	69.4
>3	69.8	55.0	36.8	52.2	30.7

DISCUSSION

Although these data are somewhat inconclusive, some generalizations may be drawn:

1. The expected relationship (i.e., with increasing sample activity the accuracy of the analysis should improve) was not seen at the levels of activity used in the studies.
2. During the six-month period for which the data were analyzed, the results did not approach the established limits.[2,3]

Exceptions were the ^{131}I samples in October and December. One reason for such inadequate per-

formance may be the expansion of the milk cross-check program to include new participants; the number of participants more than doubled during the first half of 1973. Hopefully, as these laboratories become more familiar with the analysis, the accuracy of the results will improve. Studies over a long period of time will be required to determine if any changes in the limits are warranted. These results, however, do indicate that a more detailed look should be taken at our present criteria for sample analysis, and they emphasize the need for the adoption of standard reference methods, better calibration procedures, and more frequent calibration of instrumentation.

Two facts are readily apparent from these data. First, the participants in the EPA NERC-LV national cross-check program are performing as well as reputable international laboratories (see the σ/known and s/known values of Tables I and III). Second, there is a need for considerable improvement in the laboratory analysis of both beta- and gamma-emitting radionuclides in milk. Other studies currently being conducted by the Quality Assurance Branch indicate that this is also true for alpha and beta emitters in water. The fact that less than half of the participating laboratories can satisfy the current criteria for strontium analysis in milk is particularly distressing.

That only 63% of the laboratories can satisfactorily analyze for ^{131}I in milk at levels exceeding 100 pCi/liter is of considerable interest and concern in view of the current USAEC guidelines for reactor effluent monitoring,[4] which require measurements of ^{131}I at levels below 0.5 pCi/liter.

It is believed that much improvement in the quality of radiation data can be accomplished through the adoption of standard reference methods and protocols for establishing equivalency to the reference methods, more frequent calibration and maintenance of counting instruments, improved quality control practices within each laboratory, and greater participation in laboratory intercomparison studies conducted by the National Bureau of Standards, USAEC, and EPA. As discussed earlier, the above practices are applicable to waste management, milk analysis, and all programs involving radiation measurements.

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