

# Gross Gamma Radioactivity in Indiana Precipitation

P. L. ZIEMER,<sup>1</sup> L. A. SCHAAL,<sup>2,3</sup> J. E. NEWMAN, C. L. RHYKERD,<sup>2</sup> and J. E. CHRISTIAN,<sup>1</sup> Purdue University

## Introduction

Worldwide sampling of water, soil, air and precipitation has shown that long term stratospheric fallout levels of strontium-90 and cesium-137 are closely correlated with the distribution of high velocity westerly winds and associated precipitation (1). These relationships focus particular attention on the geographical areas of the mid-latitudes such as the eastern half of the continental U. S. and western Europe. In such areas long term atmospheric fallout from post weapons tests is believed to be concentrated in two major ways: first through the amount of precipitation over a long period of years, and second, by the resulting runoff from precipitation. The principle gamma emitters that have been identified in fallout are zirconium-95, niobium-95, iodine-131, cesium-137, barium-140, and lanthanum-140. Knowledge of how to estimate and locate high concentrations of long-lived isotope fallout accumulation is an important consideration in agricultural production and marketing procedures.

The Public Health Service (4) has pointed out the growing need for more federal, state, and municipal agencies to assess the radioactivity level in environmental samples such as air, food, vegetation, milk, and water. The Public Health Service has further stated that in most cases it is desired to measure first the gross activity of the sample and then, where indicated, the activity of specific nuclides. Such measurement of gross activity at various installations throughout the country is presently complicated by a number of factors such as choice of standards, techniques of sample preparation, and type of instrument used for counting.

The determination of fallout activity in precipitation is presently carried out under the auspices of the U. S. Public Health Service Radiation Surveillance Network (5, 6). Precipitation is collected at each station using funnels with collection areas of 0.4m.<sup>2</sup> One-half liter of the precipitation is evaporated to dryness and the residue sent to a central laboratory for gross beta analysis for fission products. This method has certain disadvantages such as the time involved in carrying out the evaporation and the possible loss of volatile nuclides during evaporation.

This paper describes a rapid and sensitive method for determining the gross activity of precipitation samples by the method of large-volume liquid scintillation counting.

---

<sup>1</sup> Bionucleonics Department, School of Pharmacy and Pharmacal Sciences.

<sup>2</sup> Agronomy Department, Agricultural Experiment Station.

<sup>3</sup> Weather Bureau, U. S. Department of Commerce.

### Sample Collection and Processing

Commencing April 1, 1962, and continuing through March, 1963, precipitation catches in excess of 1 inch of water per 24 hours were collected from 29 official U. S. Weather Bureau standard 8 inch gages, geographically distributed over Indiana. From April, 1963, through March, 1964 sampling was continued at 14 of the stations. A total of 278 samples were collected over the 2 year period. Collection as indicated provided samples of approximately 1 liter in volume. Catches were transferred directly to 1 liter polyethylene bottles at the weather stations and mailed to the Bionucleonics laboratories at Purdue for the activity determinations. The only sample processing prior to determining the gross activity was a measurement of the volume. If the volume slightly exceeded 1 liter, the excess was discarded. If the volume was less than 1 liter, activity-free distilled water was added to bring the volume up to 1 liter. Finally, the exterior of each bottle was washed with distilled water so as to remove any residual surface contamination which might have accumulated.

### Instrumentation and Counting Techniques

All samples were counted directly in the bottles in which they were received. These polyethylene containers were introduced into the 8 inch long by 4 inch diameter sample chamber of the Purdue Small Animal Counter (PUSAC). Dunavant and Christian (2) have described this large volume liquid scintillation counter in detail. The counter was a 4 pi system consisting of an 8 inch long by 16 inch diameter annular tank with central sample chamber. The tank was filled with approximately 23 liters of a scintillator solution (p-terphenyl and POPOP in toluene). Four 5-inch Dumont type 6364 photomultipliers were mounted at one end of the tank. Shielding was accomplished by means of a cylindrical rolled iron shield 5 inches thick. The output pulses from the photomultipliers were fed to a preamplifier, an amplifier, a single channel analyzer, and a scaler.

The instrument was operated in the integral counting mode for maximum sensitivity. Each sample was counted for 10 minutes (three 200-second counts) preceded by and followed by background counts. Backgrounds were determined with a 1 liter sample of pure distilled water in the counting well so that no correction was necessary for background depression from the sample mass.

Samples were counted approximately 10 days after collection depending on the time interval between collection and receipt by our laboratory. No attempt was made to correct for decay during this interval of short lived materials.

### Instrument Calibration

Most work on gross measurements of fallout reported in the literature is given in terms of gross beta activity. It would be just as convenient to report gross gamma activity if an appropriate means of calibration is established.

For a mixture of fission products, a complex decay scheme results. A beta particle is liberated in each disintegration, but gamma

ray photons are produced in only about one-half the fission product disintegrations, the fraction varying somewhat with time after explosion (3). Furthermore, the mean energy of the gamma photons does not remain constant with time. Thus, a precise calibration for fission products is essentially impossible for either the betas or the gammas in a fission product mixture. However, for periods of practical interest, the mean energy of the gamma ray photons may be taken as essentially constant at about 0.7 Mev. (3).

Cesium-137 was chosen as the calibration standard for the gross gamma measurements because its gamma energy (0.662 Mev) is very close to the average gamma energy for mixed fission products. Also, cesium-137 is itself one of the important long-lived fission products and offers the additional advantage of having a simple and well-known decay scheme. For purposes of the gross gamma measurements, a curie of gamma activity was defined as that activity which resulted in  $3.7 \times 10^{10}$  gamma photons per second.

The calibration standard was prepared by placing 1 ml of a 920 gamma/sec/ml standard solution<sup>1</sup> in one of the polyethylene bottles and diluting to 1 liter with distilled water containing 0.1 g/1CsNO<sub>3</sub> as a carrier. Thus the standard was identical to the precipitation samples with respect to physical size, volume, density, and container material. Using this standard, the counting efficiency for the system was found to be 28.2%.

### Results and Discussion

The deposition of radioactivity in an area may be determined from the concentration of activity in the precipitation samples as follows:

$$D = \frac{CP}{1000}$$

where D is the deposition in nanocuries per square meter, C is the concentration in picocuries/liter, and P is the depth of the precipitation in mm. The individual values of deposition and depth of precipitation were totaled for each month, and the average concentration for the month,  $\bar{C}$ , was determined by:

$$\bar{C} = \frac{\sum D}{\sum P} \times 1000$$

Table 1 summarizes the average yearly deposition and concentration values for each of the 29 stations for the period from April, 1962, through March, 1963. Deposition and concentration values for the 14 stations participating in the April, 1963 to March, 1964 collection period are also listed in Table 1.

The highest average monthly deposition over the two year period was 271.6 nc/m<sup>2</sup> which occurred in March, 1963 at Franklin, Indiana. Franklin also showed the highest yearly total deposition of 439.5 nc/m<sup>2</sup> for the period April, 1962 to March, 1963. This high deposition is a reflection of the amount of rainfall occurring in Franklin whose 13 samples represented 528.6 mm of water. The highest average monthly

<sup>1</sup> Prepared from Nuclear Chicago Model RS-137 radioactive standard solution.

TABLE 1  
Total Deposition and Average Concentration of Gamma Radioactivity in Indiana Precipitation April, 1962, to March, 1964  
April, 1962 — March, 1963 April, 1963 — March, 1964

Station Number	Location	No. Samples	Depth of Prec. (mm)		Total* Deposition	Ave. + Conc.	No. Samples	Depth of Prec. (mm)		Total* Deposition	Ave. + Conc.
			Prec.	(mm)				Prec.	(mm)		
1.	Portage	3	88.9		100.2	1125	6	231.1		166.6	721
2.	Wanatah	6	205.5		50.2	244	3	110.2		28.8	261
3.	South Bend	16	347.0		211.1	608					
4.	Kendallville	5	132.8		78.6	592					
5.	Collegeville	1	41.1		34.3	835					
6.	Culver	4	126.5		98.2	776					
7.	Warsaw	4	136.9		130.9	956					
8.	Fort Wayne	6	157.5		100.5	638					
9.	Lafayette	11	402.6		247.1	614					
10.	Kokomo	4	134.1		87.0	649					
11.	Upland	9	329.7		292.2	886					
12.	Farmland	10	289.5		197.9	684					
13.	Crawfordsville	12	394.7		171.2	434					
14.	Whitestown	5	190.5		95.4	501					
15.	Knights town	6	265.4		227.3	856					
16.	Terre Haute	10	332.2		214.5	646					
17.	Spencer	5	240.0		139.9	583					
18.	Franklin	13	528.6		439.5	831					
19.	Greensburg	11	316.9		195.2	616					
20.	Edwardsport	8	268.7		270.7	1007					
21.	Bedford	9	413.8		269.4	651					
22.	Seymour	9	360.9		339.3	940					
23.	Owensville	3	112.0		140.4	1254					
24.	Evansville	12	406.4		224.8	553					
25.	Tell City	10	352.0		239.7	681					
26.	Dubois	5	141.0		289.6	657					
27.	Henryville	7	274.1		1057	1057					
28.	Clarksville	10	349.7		194.3	556					
29.	Madison	13	418.6		292.3	698					

\* (nanocuries per square meter)

+ (picocuries per liter)

concentrations observed were 2084 pc/l for a rainwater sample collected in May, 1962 at Owensville and 3838 pc/l for a snow sample collected in January, 1963 at South Bend.

The precision of the counting method, based on three 200-second sample counts and two 200-second background counts was  $\pm 45$  pc/l.

Values for the maximum permissible concentration (MPC) for the various fission products in drinking water have been established by the National Committee on Radiation Protection. These values range from 2000 picocuries/liter for iodine-131 to 20,000 picocuries/liter for lanthanum-140 based on continuous consumption of such water for periods up to a year. Levels of gamma fallout radioactivity in Indiana precipitation were thus well below MPC values during the period April, 1962 through March, 1964.

#### Literature Cited

1. Alexander, L. T., et al., 1961. Strontium-90 on the Earth's Surface Summary and Interpretation of Worldwide Soil Sampling Programs. U. S. Atomic Energy Commission, Office of Technical Information TID-6567.
2. Dunavant, B. G., and J. E. Christian. 1960. A Large-Volume 4 Pi Liquid Scintillation Detector. *Int. J. Appl. Radiation Isotopes* 8, 223-227.
3. Knapp, H. A., 1959. Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy, Congress of the United States, Eighty-Sixth Congress, First Session on Fallout from Nuclear Weapons Tests. pp 1969-1976.
4. U. S. Department of Health, Education, and Welfare, 1960. *Radiological Health Data*, No. 6, 54.
5. *Ibid.*, 1962, 3, No. 1, 7.
6. *Ibid.*, 1962, 3, No. 4, 105.