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AERIAL RADIOLOGICAL MEASURING SYSTEM

AERIAL RADIOLOGICAL MEASURING SURVEYS
OF THE NUCLEAR FUEL SERVICES PLANT,
WEST VALLEY, NEW YORK, 1968 and 1969

U. S. Atomic Energy Commission

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AERIAL RADIOLOGICAL MEASURING SURVEYS
OF THE NUCLEAR FUEL SERVICES PLANT,
WEST VALLEY, NEW YORK, 1968 and 1969

by

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EG&G, Inc., Goleta, California
April 1971

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This report is part of a series on the continuing activities of the Aerial Radiological Measuring System operated for the U. S. Atomic Energy Commission. The ARMS program is managed by the Civil Effects Branch, Division of Biology and Medicine, and is a cooperative effort among various offices and divisions of the Commission

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ABSTRACT

Two Aerial Radiological Measuring surveys, made in October 1968 and September 1969, of the 600 square mile area surrounding the Nuclear Fuel Services plant in West Valley, New York, are analyzed and reported. Within the primary fenced boundaries of the facility, which include a fuel-processing plant and high-level waste burial grounds, the ground exposure rates inferred from the airborne gamma count rate measurements were approx. 100 μ R/hr during the 1968 survey and approx. 1 mR/hr during the 1969 survey. These measurements, which are averages over an approx. 1200-ft-diam. effective field of view, are consistent with the concurrent plant operations. The primary sources of observed radiation were ^{137}Cs , ^{134}Cs , and ^{60}Co , and the other nuclides known to be present were of insufficient additional intensity to be observed.

Within the secondary fenced boundaries, ^{137}Cs and ^{134}Cs gamma radiation was detected along a system of waterways used for low-level liquid waste disposal, at sufficient intensity in places to exceed the background count rate by up to a factor of three. Concentrations of water-borne cesium in these streams have been measured by other investigators and are 1000 times too small to produce the radiation levels observed from the ARMS aircraft. A mechanism in which the radioactivity is strongly concentrated in the exposed silt material surrounding the streams is shown to be capable of explaining the observed radiation levels.

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Chapter 1

INTRODUCTION

1.1 ARMS PROGRAM

The Aerial Radiological Measuring System (ARMS), operated by EG&G, Inc., Las Vegas, Nevada, for the U. S. Atomic Energy Commission, performed two radiation surveys of the area surrounding the Nuclear Fuel Services, Inc., (NFS), plant in West Valley, New York: from 13 October to 15 October 1968, and from 13 September to 21 September 1969. These surveys were made as part of a continuing nationwide ARMS program started in 1958 to determine radiation levels in locales maintaining or utilizing radioactive materials. The present surveys represent the second and third such performed in the West Valley (Buffalo) area since the NFS plant began operation in 1966.¹

1.2 NUCLEAR FUEL SERVICES, INC.

Nuclear Fuel Services, Inc., is a private company with plants in West Valley, New York, and Erwin, Tennessee. The West Valley plant provides transportation and reprocessing of spent nuclear fuel, disposal of radioactive waste, and isotope recovery. West Valley, New York, is located about 30 miles south of Buffalo in an area of sparse population; in fact, there is no town or village within 25 miles with a population of greater than 10,000.

The operations of NFS, West Valley, the world's first commercial nuclear fuel reprocessing plant, have been described² and hence will only be sketched here. On removal from a reactor, fuel elements are stored for at least 3 to 4 months to allow for the natural decay of short-lived fission products.* After the cooling period, elements are transported to West Valley in specially designed shielded casks. The fuel elements are removed from the shipping casks under water and by remote control. Again by remote control, the cladding materials are removed and the unclad fuel is chopped up and then dissolved in a nitric acid solution. The fission products and the uranium/plutonium mixture are separated by chemical means, which are also used to separate and concentrate the uranium and plutonium.

Gaseous, liquid, and solid radioactive wastes are generated during the various stages of the fuel reprocessing, and they are disposed of as detailed on the following page.

*The fuel elements processed to date have been stored for eight months or longer.

Gaseous Effluents: Gaseous fission products,* such as ^{85}Kr and ^3H (produced in ternary fission), are released during the dissolution process. After filtering and monitoring, the residual low-level gases are exhausted through a stack into the atmosphere. The escaping residuals consist primarily of ^{85}Kr , which, because of its inert chemical nature, does not respond well to filtration. The ^3H is exhausted as tritiated water (HTO) which appears as vapor in the gaseous effluent and is also present in the liquid waste.

Liquid Wastes: Liquid wastes are disposed of according to intensity: high-level liquids are stored permanently in underground steel-in-concrete tanks; and low-level liquids are held in special storage lagoons until they are valved into a passing brook for dilution and dispersal. Fission products, particularly ^{134}Cs , ^{137}Cs , ^{106}Ru - ^{106}Rh , and ^{95}Zr - ^{95}Nb , as well as tritiated water have been identified in the liquid waste.^{3,4}

Solid Wastes: Solid wastes consist of two categories: general low-level waste, and high-level material such as leached fuel-element hulls and discarded plant equipment. Because the high-level materials are constructed largely of zirconium and steel alloys, long-lived nuclides produced by neutron bombardment are likely to be present: ^{54}Mn and ^{60}Co (steel) and ^{95}Zr - ^{95}Nb (from zirconium).* Long-lived fission products are also present in the solid waste.

1.3 ARMS EQUIPMENT AND PROCEDURES

The ARMS aircraft and its on-board radiation detection equipment were used to measure the radiation from the NFS facility and the surrounding area. The ARMS equipment and procedures have been discussed in detail⁵ and, hence, will only be described briefly here.

Three physical phenomena make the ARMS program viable: 1) most radioactive species or their daughters emit γ radiation, 2) most of the γ radiation characteristic of radioactivity is capable of penetrating several hundred feet of air, and 3) the γ -ray energy spectrum of a radionuclide is unique to that nuclide. Furthermore, those species whose radiation cannot be detected from a distance, α , β , and soft γ emitters, usually occur in conjunction with a hard γ emitter. Therefore, given adequate γ radiation detection equipment, which is commercially available, airborne surveys are efficient methods of determining the intensity and probably identity of aerial and/or ground radiation.

The ARMS surveys are flown in a Beechcraft Twin Bonanza at an altitude of 300 to 500 feet above ground level (AGL) at a ground speed of about 140 knots (235 ft/sec). A typical flight pattern consists of a grid of parallel paths 5 to 25 miles in length with a spacing of $\frac{1}{4}$ to 2 miles. The ground position of the aircraft as well as its altitude AGL are measured and recorded every other second via a radar-navigation-computer system. The position and altitude measurements are accurate to ± 600 and ± 5 ft, respectively, although greater position accuracy can be inferred.

The aerial radiation measurements are of two distinct types and both are made simultaneously: gross γ count (intensity) measurements and γ spectral (identity) measurements. The detector system, common to both, consists of an integrated

*Gaseous ^{131}I (fission product) and solid ^{24}Na (from neutron irradiation of aluminum) are not expected in the plant wastes, because these short-lived isotopes decay during the cooling period.

package of fourteen 4- by 4-in. NaI(Tl) scintillation crystals, each mounted on its own photomultiplier assembly. The detector system output is directed both to the gross γ count computing system and to the multichannel spectrum analyzer.

The gross γ count system consists of an amplifier-discriminator-computer package which counts and records the total number of γ -rays of energy greater than 50 keV that are detected during a precisely determined 1-second time interval. The gross γ count rate (number of detected γ -rays per second) is measured every other second and is simultaneously and automatically recorded along with the aircraft position and altitude. Gross γ count rates, typical of background, are several thousand per second, implying a statistical uncertainty of less than 2%.

As a backup and complement to the digital readout of the gross γ count data, an analog record is made on a continuous strip chart plot of both gross γ count rate and altitude as a function of distance. By cross referencing the analog with the digital record, aircraft position can be determined more accurately.

Whereas the gross γ count data specify the intensity of radiation as a function of position, the γ spectral data identify the sources of radiation. A pulse height analyzer (PHA) automatically sorts detected γ -rays according to energy, thereby generating a number-per-unit-energy versus energy spectrum. While nuclear γ -rays occur only at well known discrete energies, characteristic of the emitting species, air scatter phenomena tend to smear the detected distribution. Nonetheless, superimposed upon smooth smeared spectra are the discrete characteristic peaks that permit isotope identification. A typical spectrum with identified species is shown in Fig. 1.1. In wide area surveys, the typical acquisition time for a γ -ray spectrum is several minutes, thereby characterizing the average radiological properties of a tract several miles in length. However, if an area of interest is indicated by an increase of the gross γ count data, spectral data acquisition times of only a few seconds are used to isolate the area spatially. If further investigation is warranted, a similarly equipped ground mobile unit with greater areal and energy resolution is available as part of the ARMS program.

Fig. 1.1. Typical ARMS γ -ray spectrum showing naturally occurring radionuclides.

The equipment and procedures discussed, while sophisticated, offer great flexibility even during in-flight operation. Equipment may be tested and calibrated, detector sensitivities may be easily modified to accommodate large dynamic ranges of γ radiation intensity, local areas of interest may be repeatedly scanned from different directions to get a detailed areal radiological profile, and the techniques can be applied equally well to the detection and identification of both diffusely dispersed or highly localized quantities of radioactive materials. Lastly, there exist no terrestrial or aquatic survey techniques capable of monitoring such large areas in such short times.

1.4 REDUCTION AND PRESENTATION OF DATA

The raw data from the gross γ count and the γ spectral measurements are permanently recorded on paper tape, and they must undergo reduction and analysis to characterize the radiological properties of the surveyed area.

Calibration measurements, which give the altitude dependence of the gross γ count rate over a field of uniformly dispersed radioactivity, are utilized to convert the raw gross γ count rate data (cps) plus the recorded altitude information to the biologically more significant exposure rate ($\mu\text{R/hr}$) at 3 ft above-ground, which we refer to here as "ground exposure rate". If all 14 γ -ray detectors are used, an average of 460 cps are measured at an altitude of 300 ft over a uniform source that produces 1 $\mu\text{R/hr}$ ground exposure rate. In practice this conversion factor is obtained by averaging many values, which individually may vary by as much as a factor of 2, depending upon source nuclides and ground roughness. Furthermore, the detectability of uniform ground radiation is subject to variations of up to an additional factor of 2. For example, dense foliage, which varies seasonally, can partially shield the ARMS aircraft from ground radiation; and ground-moisture content, which varies geographically and temporally, can either wash away or shield radioactive materials in the ground.

With such complicating factors, it is easy to understand why spatial variations of ground exposure rates over apparently uniform sources are observed and why the radiological profile will change with time. Despite the fact that the ARMS' system reproducibility is good to a few percent, real natural phenomena give rise to substantially larger fluctuations. Based upon average ground exposure rates observed in the course of many nationwide surveys, exposure rates from 4 to 14 $\mu\text{R/hr}$ are considered normal.

Additional variation of the relationship between the γ count rate observed on the ARMS aircraft and the ground exposure rate is introduced if the radiation sources are not dispersed uniformly over the detectors' field of view. In particular, observed radiation sources are often concentrated within a ground area that is smaller in at least one dimension than the approx. 1200-ft-diam. effective field of view. Then the ground exposure rate near the concentrated source will be higher than the spatially-averaged rate inferred directly from the aircraft's γ count rate. If the source shape and area are known the ARMS measurement can be corrected to approximate either the higher exposure rate that would be measured near the concentrated source or the actual source intensity (Ci/m^2).

These corrections for typical source geometries can be derived and are discussed in Appendix A. As an example the exposure-rate correction factor for a uniform circular field of radiation sources is plotted, versus the radius of the circle, in Fig. 1.2. The ordinate gives the factor by which the average ground exposure rate, as inferred from the aircraft's measurement, must be multiplied to

Fig. 1.2. Correction factor relating exposure rate at 3 ft above the center of a circular uniform area source to the average ground exposure rate over the ARMS aircraft's field of view, inferred from measurements at 300 ft above the same source.

give the actual ground exposure rate that would be measured at the source center. The correction is not highly sensitive to source shape but depends primarily on source area, so long as the source does not become excessively elongated.

For sources of 1000-ft radius or larger, no correction is required. Near the other extreme the local exposure rate above a 10-ft radius source would be approx. 500 times the average ground exposure rate inferred for the whole field of view of the airborne detectors. The factor would be approx. 5000 for a point source.

Similar data can be derived for sources of other geometries, as shown in Appendix A and as used later in this report. Unless otherwise noted, however, all reports of ground exposure rates are based on the assumption of uniform radiation sources over the aircraft's field of view.

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Chapter 2

ARMS SURVEYS OF NFS, INC., WEST VALLEY, NEW YORK

2.1 DETAILED SURVEYS (PRIMARY BOUNDARIES)

Two detailed aerial radiological surveys of the NFS facilities are reported here. The 1968 survey consisted of 11 parallel flight lines, 5 miles in length and $\frac{1}{2}$ mile apart, over a 25 square mile area centered on the NFS buildings. The 1969 survey consisted of 11 flight lines 5 miles in length and $\frac{1}{4}$ mile apart and were flown in a criss-cross pattern centered on the NFS buildings. The discussion immediately following is limited to the actual fuel processing and disposal areas within the primary boundaries of the facility, for which isoexposure maps are given in Fig. 2.1 (1968 survey) and Fig. 2.2 (1969 survey).

The exposure rates measured within the primary NFS boundaries are substantially higher than typical background radiation levels observed elsewhere in the vicinity. Exposure rates of up to 100 $\mu\text{R/hr}$ were measured over the fuel processing buildings and waste burial grounds in the 1968 survey, and ≈ 750 $\mu\text{R/hr}$ were measured during the 1969 survey. This year-to-year difference is discussed below, in terms of plant operations.

The highest ground exposure rates in both surveys were centered around the processing buildings and the waste burial ground, and in each survey the radiation levels fell rapidly towards normal background values near the primary boundaries of the facility, with the one exception discussed below. The ground exposure rates, as measured from the air, appear to exceed normal levels for short distances beyond the primary boundaries owing to the extended field of view of the airborne detectors, rather than to the presence of artificial radioactivity exterior to these boundaries.

Non-background nuclides distinctly identified in the γ spectral data recorded over the primary plant and burial ground were also the same for the two surveys: ^{137}Cs , ^{134}Cs , and ^{60}Co . Both cesium isotopes are common long-lived fission products, whereas cobalt is an activated fuel rod material. As discussed earlier each of these species is expected at a nuclear fuel reprocessing plant. Figure 2.3 shows a typical γ -ray spectrum obtained over the NFS site, with identified nuclides noted.

The radiation levels observed during the two surveys are consistent with normal plant operations, and in fact they correlate with concurrent phases of the plant's operations, as follows:

Fig. 2.1. Isoexposure map of NFS, West Valley primary boundaries, 1968. The 50- and 100- $\mu R/hr$ contours appear to extend beyond the NFS primary boundaries owing to the extended field of view of the aircraft detectors.

Fig. 2.2. Isoexposure map of NFS, West Valley primary boundaries, 1969. The 50- and 100- μ R/hr contours appear to extend beyond the NFS primary boundaries owing to the extended field of view of the aircraft detectors.

SPECTRUM NO. 589
 DATE 09-21-69
 LIVE TIME .21
 INTEGRATED CT. 263407

TYPE ACFT TERRAIN BKG. - GND. DEPO.
 ALTITUDE 300
 AIRCRAFT (ARMS)

Fig. 2.3. ARMS γ -ray spectrum showing artificial radionuclides within primary NFS boundaries.

2.1.1 Burial Grounds

Average ground exposure rates were ~ 60 $\mu\text{R/hr}$ in 1968 (Fig. 2.1) and ≥ 750 $\mu\text{R/hr}$ in 1969 (Fig. 2.2). If no concentrations of radioactive materials are uncovered while awaiting interment, the burial grounds approximate a large-area source, for which uncorrected ground exposure rates are valid. The radiation levels so inferred from the 1968 and 1969 measurements are typical of waste burial operations.

The difference in the inferred 1968-1969 burial ground radiation levels has several possible contributing causes.

1. It represents in part a genuine increase of the radiation-source concentration in the burial grounds. There is, of course, an increase with time of the total radioactive material buried at the site; however, this increase is by itself not large enough to produce the observed count rate differences. The total material buried during all plant operations previous to the 1968 survey was $\sim 23,000$ Ci;¹ between the surveys $\sim 18,000$ Ci were added.² Even neglecting decay of the waste material activity, the increase in waste inventory between the surveys can account only for a factor of ~ 1.8 increase in the observed radiation level.

2. A difference of up to a factor of two can be produced by differing amounts of surface water deposited by rainfall. As discussed earlier the ground water would shield some of the ground sources from detection by the aircraft. The actual rainfall histories pertinent to the two surveys are not known.
3. Count rate excursions of a factor of 10 can easily be produced during normal waste burial operations, when radioactive material is present on the burial ground site but has not yet been covered with earth. As shown in Fig. 1.2 the exposure rate near such a limited-size source can exceed the average exposure rate inferred from the ARMS measurement by more than a factor of 100.

The difference of the burial ground radiation levels of 1968 and 1969 indicate that a waste burial operation was probably underway during the 1969 flights but not during those in 1968. In support of this hypothesis we have investigated the operating status during the surveys. During the 1968 flights no fuel reprocessing operation was underway.³ During the 1969 survey, however, a reprocessing operation was nearing completion and was at a phase in which wastes would be available for burial.⁴ Unfortunately, no concurrent records or photographs of the site are readily available to evaluate whether burial operations were actually in progress.

2.1.2 Processing Plant

The radiation levels observed from the ARMS aircraft, expressed as average ground exposure rates over the processing plant buildings, were 70 to 100 $\mu\text{R/hr}$ during the 1968 survey (Fig. 2.1), and ≈ 750 $\mu\text{R/hr}$ during the 1969 survey (Fig. 2.2). Nothing in the processing plant is large enough to classify as a "large area" source, and, consequently, local exposure rates would have exceeded the above values. Based on the correction factors given in Fig. 1.2 the average exposure rates listed in Table 2.1 would be measured over limited-area features of the processing plant, if each were the source of the observed radiation. These data are not corrected for attenuation by any shielding that may be present.

TABLE 2.1 -- RADIATION LEVELS OBSERVED OVER PROCESSING PLANT BUILDINGS, EXPRESSED AS AVERAGE EXPOSURE RATES OVER THE ENTIRE FIELD OF VIEW AND ALSO OVER CERTAIN LIMITED-AREA FEATURES.*

Feature	Area, square feet	Correction Factor	MR/hr (average)	
			1968	1969
Aircraft field of view	~1,000,000	1	~0.1	~0.8
Whole processing area	~80,000	7	~0.6	~5.0
Fuel-element storage pool	~4,500	26	~2.2	~20.0

*Data are not corrected for any shielding that may be present.

As was the case with the burial ground radiation the higher exposure levels observed in the 1969 flights over the processing buildings also appear to correlate with the different fuel reprocessing phases underway at NFS during the two surveys, (see 2.1.1).

One contributing factor has been isolated that produces at least part of the enhanced radiation levels observed in 1969.* A batch of spent fuel was received by NFS during 1969 that contained substantially more radioactive impurities than usual. During mechanical processing of this fuel these impurities were released into the fuel storage pool, bringing its radioactivity concentration from $\sim 10^{-4}$ $\mu\text{Ci/ml}$ typical of 1968 operations up to $\sim 10^{-2}$ $\mu\text{Ci/ml}$ at the time of the 1969 survey. This concentration would enhance the 1968 γ -ray count rate by at least a factor of three and so must have significantly contributed to the higher levels in 1969.

2.2 DETAILED SURVEYS (SECONDARY BOUNDARIES)

The ground exposure rates were observed to fall rapidly to normal background levels near the primary boundaries of the NFS site, except for two regions of terrain in which the rates were 2 to 3 times higher than average:

1. A ribbon that extends north from the processing plant, then veers to the northwest, with a local maximum about 2 miles NNW of the plant buildings, as shown in Figs. 2.4 and 2.5. This ribbon coincides with the paths of the waterways into which low-level liquid wastes from the fuel reprocessing operation are dispersed. Analysis of the observed radiation is the primary subject of this section.
2. Above-background radiation contours to the west of the processing plant, in the vicinity of a waterway called Quarry Creek, into which no radio-hydrological paths are known to exist (see Fig. 2.5). This "Quarry Creek Anomaly" is discussed at the end of this section.

2.2.1 Liquid-Waste-Dispersal Waterways

Figures 2.4 and 2.5 show isoexposure contours within the secondary boundaries of the NFS facility as derived from the 1968 and 1969 data, respectively. Both figures demonstrate above-background radiation levels that correlate with the paths of Frank's Creek and Buttermilk Creek, with local maxima at confluences between Frank's and Buttermilk and between Buttermilk and Cattaraugus Creeks. This complex of waterways is the system into which low-level liquid wastes are dispersed.

Above-background radiation levels were also inferred for Cattaraugus Creek, downstream from NFS. Since this waterway lies outside the NFS boundaries the presentation of the Cattaraugus Creek data and their analysis are reserved for our discussion of the wide-area surveys, in the next section.

A detailed analysis of the gross γ count rate data unequivocally demonstrates that the enhanced exposure rates were caused by radioactivity in or around Frank's Creek and Buttermilk Creek downstream from NFS. Plots of simultaneous

*We gratefully acknowledge the contribution made by the AEC Compliance staff in calling this factor to our attention.

Fig. 2.4. Isoexposure map of NFS, West Valley properties, 1968.

Fig. 2.5. Isoexposure map of NFS, West Valley properties, 1969.

recordings of radar altitude (i.e., ground clearance) and gross γ count rate, as a function of aircraft position, clearly show count rate maxima when crossing rivers downstream from NFS. River crossings are located at minima in absolute terrain altitude, i.e., at maxima in ground clearance. Typical plots of this type are shown in Figs. 2.6 and 2.7, for 1969 flight paths that crossed Frank's and Buttermilk Creeks, and the plots show sharp increases in the count rate over the streams. Similar crossings of Buttermilk Creek upstream from the plant revealed no increase in the background radiation level.

The flight paths along the downstream waterways were sufficiently far from the buildings and burial grounds, so that the γ -ray spectra were not influenced by pickup of radiation emanating from within the primary fenced boundary. The 1969 γ -ray spectra from flight paths along Frank's and Buttermilk Creeks are shown in Figs. 2.8 and 2.9, respectively. Photopeaks at 0.66 ± 0.02 MeV (^{137}Cs) and 0.79 ± 0.02 (^{134}Cs) are clearly present in both spectra. Identification of the photopeaks as cesium radiation is unambiguous, in spite of the nearly similar photopeak energies of ^{106}Ru - ^{106}Rh (0.622 MeV) and ^{95}Zr - ^{95}Nb (0.724 and 0.756 MeV), owing to the 0.02-MeV spectral resolution of the detectors.

Other investigators^{5,6} have studied water samples from Frank's Creek and Buttermilk Creek. They substantiate the conclusion that ^{137}Cs and ^{134}Cs are major contributors to the γ -ray emissions of the radioactive materials in the water. However, they also report other nuclides, including ^{106}Ru - ^{106}Rh and ^{95}Zr - ^{95}Nb , in concentrations that are below the sensitivity threshold of the ARMS detectors when in the presence of the observed nuclide concentrations. They also report the presence of other nuclides, including ^{90}Sr , which emit no penetrating γ -rays and therefore cannot be detected from the aircraft.

Based upon equations in Appendix A the concentration of the observed radioactive materials at ground level can be derived from the increase in the gross γ count rate if one knows the source geometry. Since the data so strongly demonstrate that the increased activity is concentrated in or around the waterways the source geometry can be approximated by a ribbon-like shape of infinite length, whose width is approximately that of the creek or brook. From U. S. Geological Survey maps and aerial photographs the widths of the various waterways were determined to be about 25 ft for Frank's Creek, 100 ft for Buttermilk Creek, and 200 ft for Cattaraugus Creek. Over Frank's Creek the gross γ count rate increases were about 4000 cps, while over Buttermilk Creek they were as high as 5000 cps. (The background-radiation counting rate was ~ 2000 cps for the limited area surveys.) The increases over Cattaraugus Creek were only a few hundred cps and are discussed separately below.

The source dimensions, the increase in gross γ count rate, the effective initial γ -ray energy (0.66 MeV), and the equations in Appendix A were used to calculate the surface concentration of cesium to be about $70 \mu\text{Ci}/\text{m}^2$ along Frank's Creek and as high as $20 \mu\text{Ci}/\text{m}^2$ along Buttermilk Creek. If the radioactive material is assumed to be uniformly dissolved or suspended in the water the inferred volume concentrations of cesium are approximately 6×10^{-10} Ci/cc and 2×10^{-10} Ci/cc, respectively, as determined by dividing the surface concentrations by the 12-cm mean free path of a 0.66 MeV γ -ray in water.

The on-site measurements by the other investigators^{5,6}, however, showed a cesium concentration in Buttermilk Creek of about 1.0×10^{-13} Ci/cc, which is about three orders of magnitude lower than our inferred volume concentration. From this difference, we conclude that what we observe cannot be coming from radioactive material dissolved or suspended in the water.

Fig. 2.6. Ground exposure rate vs. position and ground elevation vs. position plots showing increased activity over Frank's Creek, downstream from NFS.

Fig. 2.7. Ground exposure rate vs. position and ground elevation vs. position plots showing increased activity over Buttermilk Creek, downstream from NFS.

Fig. 2.8. ARMS γ -ray spectrum showing artificial radionuclides along Frank's Creek.

Fig. 2.9. ARMS γ -ray spectrum showing artificial radionuclides along Buttermilk Creek.

Another radiohydrological model, suggested by observations of both ground-based and airborne surveys, gives better agreement with our measurements. The model hypothesizes that 1) radioactive cesium is strongly concentrated in silt deposits along the NFS waterways, and 2) these deposits were exposed during the ARMS overflights, which occurred during autumn when the water levels are known to be at a yearly low (as reported by the U. S. Geological Survey⁷). There are two observations that represent strong evidence in support of this hypothesis.

1. The ground-based surveys reported ^{137}Cs concentrations in Buttermilk Creek bottom silt as high as 1.8×10^{-10} Ci/g, or about 1000 times the concentration in the water itself.
2. In flying along the waterways, local maxima in the gross γ count rate were detected at three distinct locations: 1) at the confluence of Frank's and Buttermilk Creeks, 2) at a point where Buttermilk Creek meanders sharply, and 3) at the confluence of Buttermilk and Cattaraugus Creeks. All are likely areas for the deposition of waterborne silt.

Thus, when considered in the light of available ground survey data the ARMS observations of increased activity along waterways, downstream from NFS, are best interpreted in terms of the accumulation and concentration of ^{134}Cs and ^{137}Cs in silt, which is deposited along the banks and exposed during periods of low-water levels.

The observed radiation intensity emanating from the vicinity of these waterways was higher by about 30% in the 1969 survey than in 1968. This increase is probably not sensitive to day-to-day differences of the amount of radioactive material discharged into the waterways because, as noted earlier, typical radiation-source concentrations in the water are several orders of magnitude smaller than necessary to produce the observed count rates.*

Instead the intensity increase suggests that higher concentrations of radioactive material existed in the stream beds during the later survey, which would imply a buildup of the accumulated radiation source concentration with time. However, the observed levels of radiation, based on our silt-accumulation model, depend strongly on the amount of water flowing in the streams at the time of the flight in this way: a higher water level would shield a greater part of the silt accumulation from the overflying aircraft. Thus we can explain the 1968 vs. 1969 differences as well by postulating a 30% larger area of silt exposed during 1969, of the same radiation source concentration as existed in 1968. A choice between the two models could be made if water flow rates in the creeks were known; these data are not readily available for either survey.

2.2.2 Quarry Creek Anomaly

An area of above-background activity was indicated west of the primary building complex during the 1969 ARMS survey (first discussed in Section 2.2). It appeared as a local maximum along one flight line, in a position that was not consistent with pickup of stray radiation from the higher-level primary site. The exposure-rate value at the peak of this local maximum was corroborated by an orthogonal flight path, although the actual maximum was not resolved.

*See also "Note added in proof," p. 35, for pertinent results of a similar NFS survey conducted during autumn 1970.

The location of the enhanced exposure region coincided with a tributary of the Buttermilk Creek known as Quarry Creek and, tentatively, the source of the excess radiation was assumed to be in or near Quarry Creek. This assumption was based on our previous evaluation of significant concentration of radioactivity in Frank's Creek and Buttermilk Creek, even though to infiltrate Quarry Creek it would appear that radioactivity would have had to move upstream from these regions of known enhanced activity.

The isoexposure contours of Fig. 2.5 were consequently drawn according to the assumption of Quarry Creek's involvement in the anomalous exposure. In actuality the 1969 survey flights do not exhibit sufficient spatial resolution by themselves to identify the contours unambiguously as drawn. Discussion of the apparent anomaly with AEC personnel has revealed the following.

1. No known paths exist for radioactivity to flow upstream, from Buttermilk Creek into Quarry Creek.
2. Possible leakage from underground liquid waste storage tanks northwest of the NFS buildings into Quarry Creek was investigated by the AEC and found to be nonexistent.
3. Nevertheless, an on-site inspection was made by AEC personnel of the Quarry Creek area at the point where the ARMS overflights reported enhanced counting rates. Local ground exposure rates were found to be approximately three times normal background, apparently corroborating the ARMS measurement.

This anomalous radiation source location and cause are under current investigation by the AEC.

Owing to the apparently corroborated involvement of the Quarry Creek waterway in this above-background region we have permitted the detailed isoexposure contours, which were based on the assumption of this involvement, to stand.

2.3 WIDE-AREA SURVEYS

The flight patterns of the two wide-area aerial radiological surveys flown over the region surrounding the NFS facility were similar, each consisting of 25 parallel flight lines, 25 miles in length with one-mile spacing, thus surveying a 600 square mile area centered on the NFS site. The results of the wide-area surveys are discussed below in two parts: a general discussion of the 600 square mile radiation signature of the NFS vicinity (Section 2.3.1), and a discussion of radioactivity inferred to be in the Cattaraugus Creek, outside the secondary NFS boundaries, as derived from the wide area survey data (Section 2.3.2).

2.3.1 NFS Vicinity Radiation Signature

The general results of the wide-area surveys, in the form of contour-like isoexposure maps, are shown in Figs. 2.10 and 2.11, respectively. Together they define the radiation signature and variations for the NFS vicinity.

The maximum ground exposure rate observed in either survey, excluding the immediate NFS area, is 15 μ R/hr, an individual reading in satisfactory agreement

Fig. 2.10. Isoexposure map of West Valley area, 1968.

Fig. 2.11. Isoexposure map of West Valley area, 1969.

with the normal-background range of 4 to 14 $\mu\text{R/hr}$. Readings of 12 $\mu\text{R/hr}$ or higher, when individually scrutinized, were always found in conjunction with rapid altitude changes, conditions under which the altitude compensation factor is of limited accuracy.

The results of the two surveys are in qualitative agreement, i.e., the high and low exposure rates from survey to survey are reasonably well correlated. Perfect correlation is precluded not only by the natural phenomena already discussed, but also because the shapes of the contour lines are interpolated between discrete flight lines which differed somewhat for the two surveys. Nonetheless, the observed highs and lows do correlate well and, furthermore, they can generally be related to natural geographic features. For instance the higher ground exposure rates observed in the extreme northwest sector of the maps in Figs. 2.10 and 2.11 coincide well with the region around the town of Eden, New York, a relatively developed area. Other smaller developed areas also exhibit slightly higher ground exposure rates than do undeveloped areas, due perhaps to ground cultivation and/or the presence of construction materials (e.g., concrete) exposing greater concentrations of naturally occurring radionuclides.

The results of the two independent surveys are quantitatively different, the 1968 exposure rates averaging about 2 $\mu\text{R/hr}$ less than the 1969 rates. Such an overall pattern is quite consistent with normal meteorological and seasonal variations of ground moisture content, which have already been discussed.

It may be significant that the generally lower exposure rates during 1968 suggest a greater ground water content than in 1969, probably produced by a rainfall just prior to the survey. Such a rainfall would also have produced a higher water level in the creeks during the 1968 survey which, as discussed earlier, would have shielded part of the creek-bed radiation, thus producing the lower levels of waterway radiation intensity observed in 1968. Of course, this unsubstantiated hypothesis is insufficient to isolate the cause(s) of the 1968 vs. 1969 survey differences.

2.3.2 Radioactivity in Cattaraugus Creek

The only indications of any measurable concentration of fission or activation products outside the NFS boundaries arise when a detailed analysis is made of the gross γ count data taken as the aircraft crossed Cattaraugus Creek downstream from NFS. (This analysis is similar to the analysis of the radioactivity content of Frank's Creek and Buttermilk Creek presented earlier in our discussion of radiation levels within the secondary boundaries of NFS.) Because the apparent concentrations of radioactivity in Cattaraugus Creek are small, they are difficult to detect and resolve from natural sources. However, 30 such crossings were made both upstream and downstream from NFS, thereby providing a good statistical sample upon which to base conclusions.

Care must be taken in evaluating the γ count data, because altitude-compensation corrections can obscure or enhance subtle effects. The altitude compensation is based on ground clearance measurements by a narrow-field radar altimeter, which determines the altitude above a point directly beneath the aircraft. The compensation based on this narrow-field measurement is accurate above a flat plain, but it overcorrects the count rate when the aircraft crosses a narrow river depressed below the surrounding terrain. Thus, altitude-compensated data will tend to exaggerate gross γ count rates observed while crossing narrow

waterways. On the other hand, uncompensated data will be overly conservative, since at least part of the field of view is further removed from the detectors. The following conclusions take these limitations into consideration.

Upstream from the NFS facility on the Cattaraugus Creek the uncompensated gross γ count data underwent decreases of 300 to 500 cps as the creek was crossed; downstream the data remained constant within ± 200 cps. The 300 to 500 cps represents a fractional change of about 10% in the upstream count rate and is therefore resolvable from system effects and statistical uncertainties. The compensated data upstream remained constant to within ± 100 cps, while downstream increases from 300 to 2000 cps were observed. By either measure the results demonstrate that a net increase in the gross γ count rate is present downstream from NFS.

Our data show an average increase in gross γ count rate of 500 cps above ambient when crossing the Cattaraugus downstream from its confluence with Buttermilk Creek. If we again were to assume the observed activity to be due to material dissolved or suspended in the water, we would infer surface and volume concentrations of $1 \mu\text{Ci}/\text{m}^2$ and $1.2 \times 10^{-11} \text{ Ci/cc}$, respectively. The other investigators^{5,6} have surveyed the Cattaraugus Creek downstream from NFS, and have reported ^{137}Cs concentrations of $1.0 \times 10^{-14} \text{ Ci/cc}$ in the water and from 2.4 to $6.2 \times 10^{-11} \text{ Ci/g}$ in the bottom silt. Comparison of their and our results again suggests that the detected activity stems from a concentration in the silt rather than in the water (see earlier discussion). This hypothesis is supported by the observation that local maxima in intensity occur where the Cattaraugus meanders sharply, which are likely places for shoreline silt accumulation.

Since the observed intensities of the artificial radioactive sources were small and since wide-area γ spectral data were recorded along long portions of flight lines, no aerial isotope of the Cattaraugus Creek radiation sources could be corroboratively identified. However, ^{137}Cs and ^{134}Cs were identified in Frank's and Buttermilk Creeks, which empty into Cattaraugus Creek, and the ground surveys also report the presence of these species. Consequently, the source identification is positive.

Finally, the decreases in inferred cesium concentrations, from 6 to 2 to $0.1 \times 10^{-10} \text{ Ci/cc}$, along Frank's Creek, Buttermilk Creek, and Cattaraugus Creek, respectively, are consistent with the known dilution factors of these waterways.

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Chapter 3

SUMMARY

An analysis has been presented of data recorded on ARMS survey flights over the Nuclear Fuel Services plant and vicinity, West Valley, New York, during 1968 and 1969. Ground exposure rates ($\mu\text{R/hr}$ at 3 ft above the ground) and, in some cases, concentrations of radioactive materials were inferred from the airborne measurements for three main areas of the facility: 1) the fuel processing buildings and radioactive-waste burial grounds, which lie within the primary fenced boundaries, 2) the system of waterways into which low-level liquid wastes are dispersed, and 3) the remainder of the 600 square mile vicinity of NFS.

3.1 PRIMARY FENCED BOUNDARIES

Maximum ground exposure rates of $100 \mu\text{R/hr}$ were observed during the 1968 survey over the processing buildings and burial grounds, and rates of $\approx 750 \mu\text{R/hr}$ were observed during the 1969 survey. These exposure rates represent averages over the $\sim 10^6$ square-foot field of view of the ARMS sensors, and more intense local maxima can be present. The observed exposure rates correlate with concurrent operating phases during the two surveys: the lower 1968 levels were recorded during a quiescent phase between batches of fuel reprocessing operations, whereas the higher 1969 exposure rates occurred during the last phases of a fuel reprocessing cycle, when much radioactive waste material is separated out. Sources of the observed radiation were primarily ^{60}Co , ^{137}Cs , and ^{134}Cs ; other nuclides were not present in sufficient quantity to resolve in the presence of the ones observed.

3.2 LIQUID-WASTE DISPOSAL WATERWAYS

Above-background radiation levels were observed along the system of waterways into which low-level liquid wastes are dispersed. Source nuclides of the observed radiation were ^{137}Cs and ^{134}Cs . Typical surface and volume concentrations of cesium were derived for the waterways, based on the assumption that the observed sources were dissolved or suspended in the water. These data are shown, together with on-site measurements by other investigators, in Table 3.1. There is a factor of $\sim 10^3$ difference between our inferred concentrations of radioactive cesium and those measured by others. This difference suggests that instead of the radiation sources being in the water, as assumed, there is instead a mechanism which accumulates radioactive material near the creeks.

TABLE 3.1. INFERRED AND MEASURED SURFACE AND VOLUME CONCENTRATION OF ^{137}Cs AND ^{134}Cs IN NFS WATERWAYS. THE DIFFERENCE BETWEEN INFERRED AND MEASURED VALUES SUGGESTS A MECHANISM BY WHICH CESIUM IS ACCUMULATED NEAR THE CREEKS.

Creek	Width (ft)	Count Rate Increase (cps)	Surface Concentration ($\mu\text{Ci}/\text{m}^2$)	Inferred Volume Concentration (Ci/cc)	Measured Volume Concentration (Ci/cc)
Frank's	~25	4000	70	6×10^{-10}	---
Buttermilk	100	5000	20	2×10^{-10}	1×10^{-13}
Cattaraugus	200	500	1	1.2×10^{-11}	1×10^{-14}

This mechanism appears to be a thousand-fold concentration of ^{137}Cs and ^{134}Cs in the silt bottom and banks of the waterways. Such a concentration of cesium has been reported by the on-site investigators. This cesium-bearing silt can produce the levels of radiation observed by the ARMS aircraft when the water level is low enough to expose significant areas of the creek beds, as occurs at NFS mainly during autumn. Both ARMS surveys were conducted during autumn.

3.3 NFS WIDE-AREA SURVEY

Throughout the remainder of the 600 square mile NFS survey area, only typical background-level radiation signatures were observed.

Chapter 4

PRELIMINARY SURVEY RESULTS, 1970 SURVEY

Based on the measurements reported here for 1968 and 1969, a third ARMS survey of the NFS plant was flown, in the fall of 1970. Particular emphasis was paid to the liquid-waste dispersal waterways, and flight lines were directed along the streams to enhance detection of radioactivity in them. The findings fully corroborate and extend the measurements reported here for 1968 and 1969.

Both ^{137}Cs and ^{134}Cs were again unambiguously present during the 1970 survey; the isolated 1970 flight lines enhanced the observable line structure in the spectra. Gross count rates were substantially higher along the rivers than over the surrounding terrain $\frac{1}{4}$ mile on each side of the rivers. The observed radiation levels in 1970 exceeded those observed in 1969 by up to a factor of two, owing in part to the more detailed coverage of the waterways provided in the 1970 survey (See Fig. 4.1). Average radiation levels observed over the streams were from 30% to more than 50% higher than in 1969, which in turn had been ~30% higher than observed in 1968. This monotonic increase over the period 1968 to 1970 lends credence to the possible inference that radioactivity concentrations in the vicinity of the streams are on the increase. As was true for the earlier surveys, however, it is not possible to define unambiguously whether the observed increase is due to an increased concentration of radioactive sources or to a correspondingly lower water level in the streams.

Fig. 4.1. Partial isoexposure contours derived from preliminary data of an ARMS survey flown in autumn 1970.

APPENDIX A

To convert an observed gross γ count rate or a resolvable increase in the same to an effective ground concentration, the following definitions and calculations are used:

I_o	=	number of γ 's emitted per unit area per second
R	=	total number of γ 's detected per second
A	=	total effective area of the detectors
$d\omega$	=	element of emitting surface area
ρ	=	distance from $d\omega$ to detector
μ	=	total γ mass attenuation coefficient in air
μ_o	=	total γ mass attenuation coefficient in surface material
$e^{-\mu\rho}$	=	air scattering attenuation factor
$A/4\pi\rho^2$	=	inverse square attenuation factor
F_b	=	buildup factor to account for detected scattered γ 's
N_c	=	number of γ 's per second per curie (Ci) = $3.7 \times 10^{10}/\text{sec}$
z	=	vertical distance between source and detector (altitude)
ϕ	=	effective absorption factor for the detector array

To calculate the detected count rate, R , the contributions from all the sources must be summed:

$$R = \int \frac{F_b I_o \phi A e^{-\mu\rho} d\omega}{4\pi\rho^2}$$

In general, integrals of this sort cannot be performed in closed form. However, they can usually be transformed into some form of the exponential integral, which is tabulated¹, or otherwise numerically estimated. Such calculations can be carried out for the general geometries of the circular area and the infinite ribbon area, as well as their limiting cases of the point source, the infinite line source, and the infinite plane source.

1. Point source: This special case is trivial, and for a point source intensity of I_p :

$$R = \frac{F_b I_p \phi A e^{-\mu z}}{4\pi z^2}$$

2. Circular plane-area source: A simple change of variable reduces the general expression to:

$$R = \frac{1}{2} F_b I_o \phi A \int (e^{-x}/x) dx$$

between the limits μz and $(\mu^2 z^2 + \mu^2 r^2)^{1/2}$, where r is the radius of the source. The integral is tabulated in terms of the exponential integral.

3. Infinite plane source: This special case is calculable by extending the upper limit of the previous example to infinity.
4. Infinite ribbon source: This most complex case can be expressed as follows:

$$R = F_b I_o \phi A \int_{-\infty}^{\infty} \int_{-w/2}^{w/2} \frac{e^{-\mu \rho}}{4\pi \rho^2} dx dy$$

where w is the width of the source and $\rho^2 = x^2 + y^2 + z^2$. This integral has been numerically evaluated for several widths.

5. Infinite line source: This special case is calculable by suppressing integration over the source width of the above example and expressing the intensity as an intensity per unit length, I_ℓ .

Buildup factors have been described²; and at 0.66 MeV (¹³⁷Cs), they are found to vary from about 2 for a point source to about 4 for an infinite plane source, when the detector is located about one mean free path from the source. (The nominal altitude of 300 ft represents 0.95 mean free paths of a 0.66 MeV γ -ray in air.) Since the integrals can be evaluated, the buildup factors are known, and the effective detector area is approximately $14\pi a^2$ (14 detectors of

radius a), the detected gross γ count rate can be related to the ground intensity. For an easily resolvable rate increase of 500 cps, a general expression for the ground area concentration per 500 cps increase becomes:

$$\#Ci/m^2 = I_o/N_c = \frac{2000\pi}{F_b \phi A N_c \int (e^{-\mu\rho}/\rho^2) d\omega}$$

If the mean free path of the emitted γ 's through the surface material is $1/\mu_o$, the ground volume concentration becomes:

$$\#Ci/m^3 = I_o \mu_o/N_c$$

In water $1/\mu_o$ is about 12 cm for 0.66 MeV γ 's. If it is believed that the radioactivity is concentrated near the surface at a depth of only a few cm, as is likely if the materials are deposited on the surface rather than dissolved, then a shorter range should be assumed.

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