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TM-345C
1104.100

ENVIRONMENTAL MONITORING REPORT
For Calendar Year 1972

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March 23, 1973

Operated by Universities Research Association Inc. Under Contract with the United States Atomic Energy Commissio

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1. Introduction

The National Accelerator Laboratory (NAL) facility is a proton synchrotron with a design energy of 200 GeV (billion electron volts); however, the accelerator is now operated routinely at 300 GeV and it reached a maximum energy of 400 GeV in 1972. The primary purpose of the installation is fundamental research in high-energy physics. It is located in the greater Chicago area (Fig. 1), permitting convenient access for users throughout the country. The 10.6 sq. mi. (27.5 km^2) tract of land comprising the site is in an area which is rapidly changing from farming to residential use. There are many municipalities in the vicinity, resulting in a distinct pattern of high population concentration. Within a 2 mile (3 km) distance from the Laboratory boundaries Batavia (pop. 8,500), Warrenville (pop. 3,000), and West Chicago (pop. 9,900) may be found.

The two major environmental features near the Laboratory are the Fox River to the west which flows south through Batavia with an average of 500 million gallons per day, and the west branch of the DuPage River which passes east of the site flowing south with lower flow rate through Warrenville. The land on the site is relatively flat with a high area (elevation 800 ft. above sea level, ASL) near the western boundary and low point (elevation 715 ft., ASL) toward the southeast. The drainage of the ground water and most of the surface water is toward the southeastern corner of the Laboratory, toward the DuPage River. A somewhat smaller amount drains to the southwest, toward the Fox River.[1] The drinking water in many of the surrounding communities comes from deep wells usually drilled 1200 feet deep into the Cambrian Ordovician aquifer system.[2]

The 2 km (1.2 mi.) diameter main accelerator (Fig. 2) receives 8 GeV protons from a combined function booster accelerator which is fed by a 200 MeV linear accelerator (linac). The linac receives protons from a hydrogen ion

Figure I - Location of National Accelerator Laboratory (NAL)

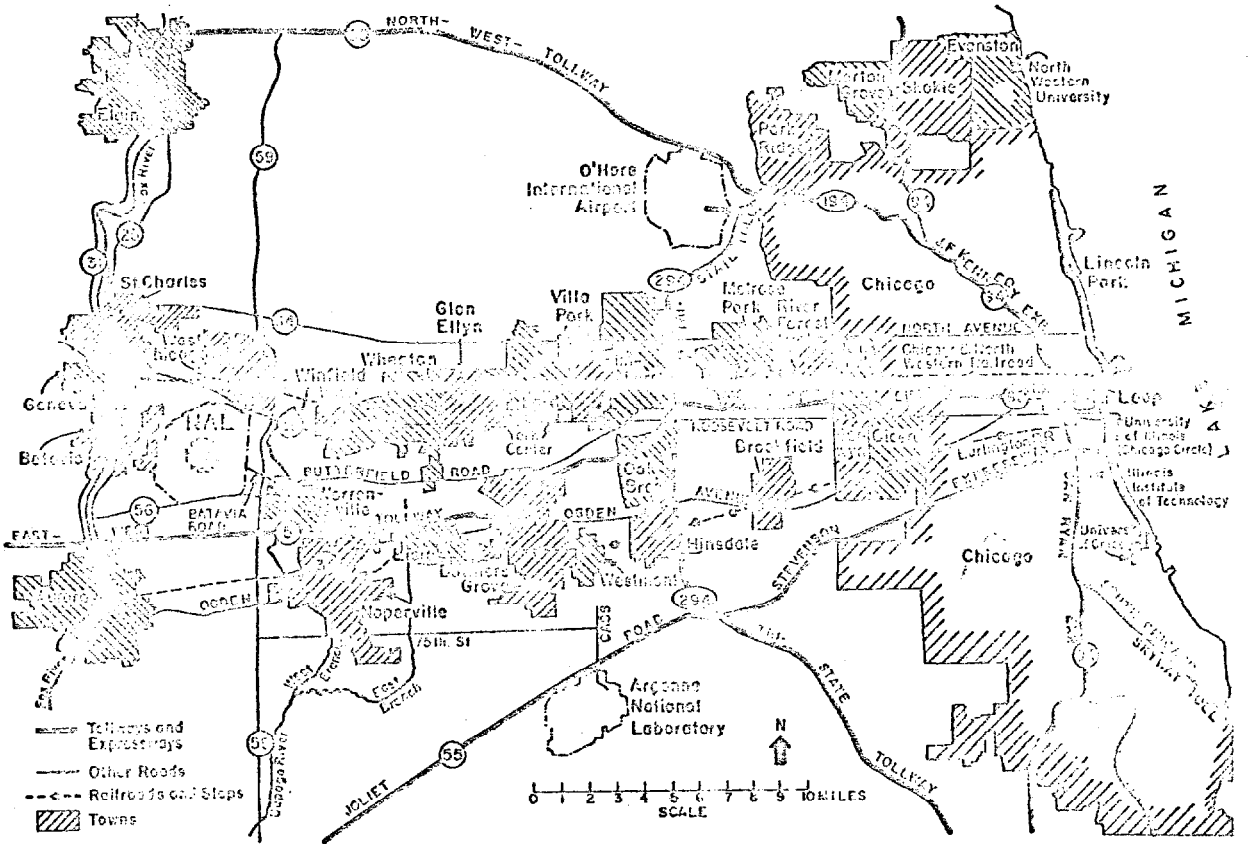
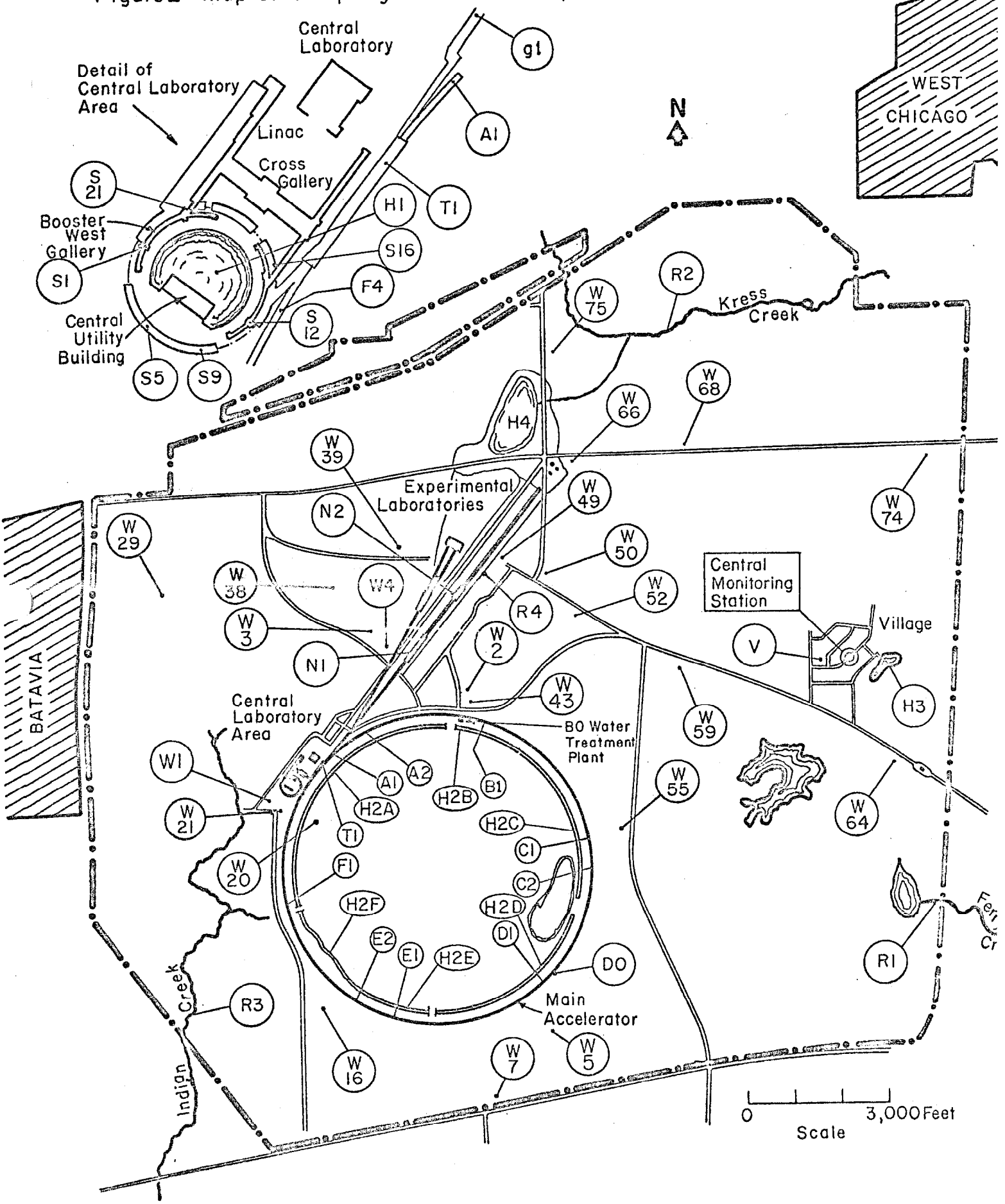


Figure 2- Map of Sampling Locations - Map 001



source via a 750 keV Cockroft-Walton electrostatic accelerator. The beam extracted from the main accelerator can be taken to three different areas (Figs. 3, 4). All three of these areas received proton beams for the first time in 1972. Radioactivity is produced as a result of the interaction of the accelerated protons with matter. Most of this activity is contained in insoluble shields and beam dumps. Operation of the accelerator will produce some radiation which penetrates the shielding as well as some air-borne activity. Also, some radioactivation of the soil will occur. Thus, a broad program of environmental monitoring is being maintained.

An environmental radiation monitoring station, located temporarily in a house in the NAL Village (Fig. 2), has been in operation throughout 1972. Also, a four-wheel-drive vehicle is being especially equipped for environmental radiation monitoring. This mobility is very desirable in view of the long distance from one corner of the site to another, or from the origin of a beam line to its terminus. Water samples are analyzed by an independent testing laboratory as well as in NAL Nuclear Chemistry and Counting Laboratories. Environmental radiation data are collected from a multitude of radiation detectors throughout the accelerator and experimental areas.

The analyses for residual radioactivity are performed specifically for accelerator-produced nuclides as well as for radium and thorium. Since there is no reactor facility on the site, no program has been initiated for monitoring fission product activity such as occurs from ruptured fuel rods or in fall out from nuclear weapons tests. Some analyses of this type are carried out on samples collected near the site by Argonne National Laboratory located 15 mi. (25 km) away.

2. Summary

In 1972 the design energy of 200 GeV was first achieved and the first beam extracted from the accelerator before the scheduled date of July 1. The proton energy was raised to 300 GeV (now the standard operating energy) during the summer

Figure 3 - Sampling Locations Along Beam Lines

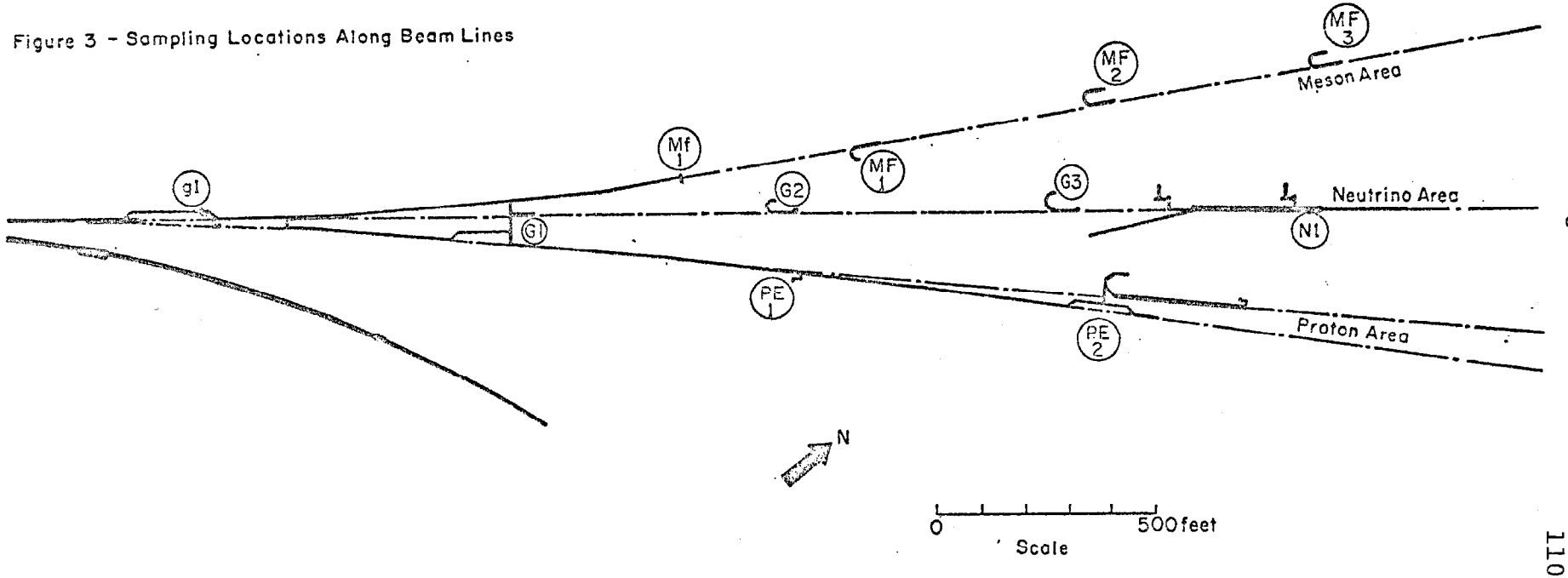
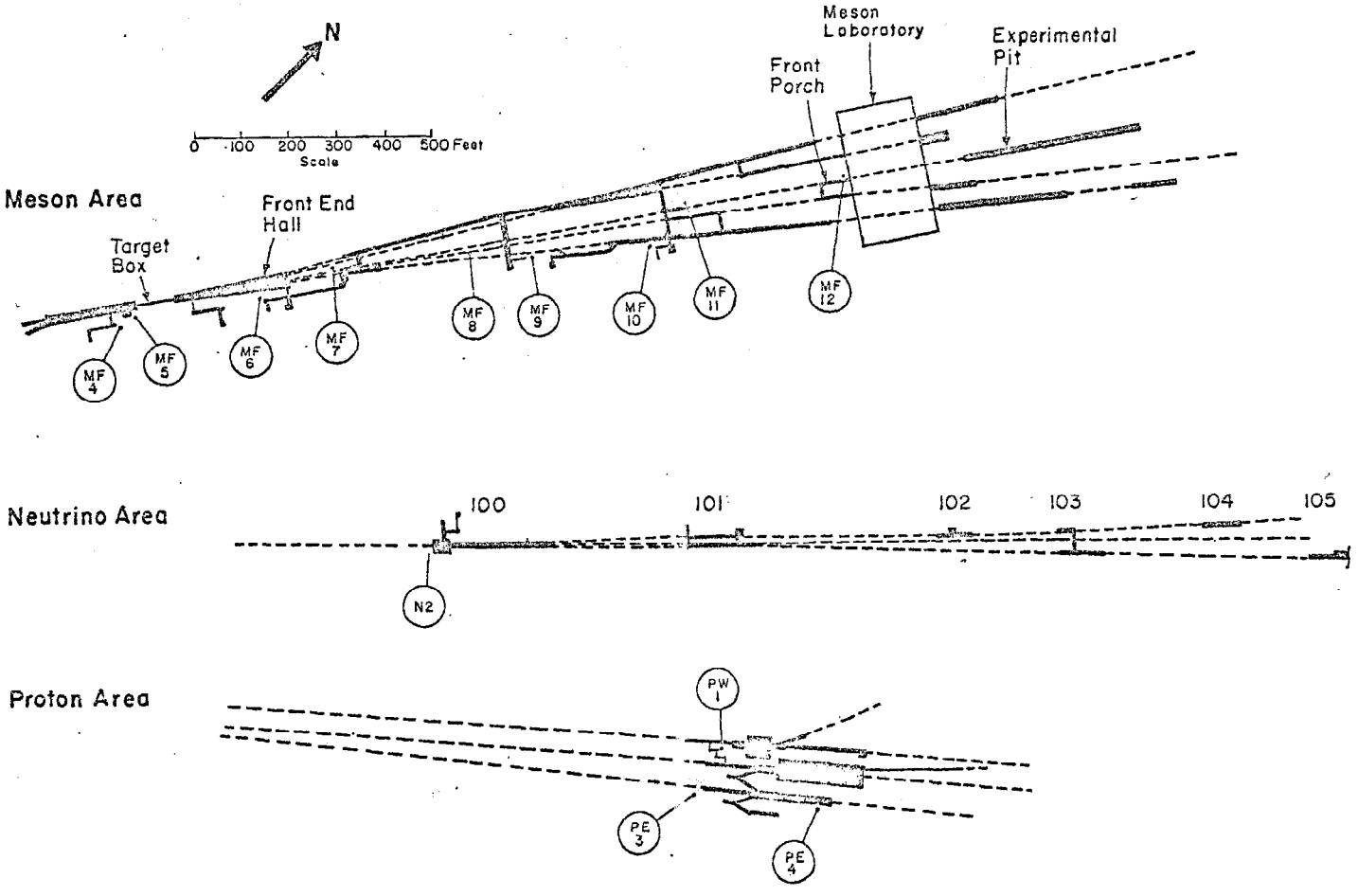


Figure 4- Sampling Locations Along Beam Lines



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and then near the end of 1972, 400 GeV operation was achieved. The Neutrino Area (Figs. 3, 4) received extracted beam for extended periods of time at high proton beam intensities while the Proton and Meson Areas, which are still under construction, received low proton beam intensities starting in September mainly for transport studies. Beam splitters are now being installed to allow the three areas to receive simultaneously portions of the same beam pulse. In addition, using a pulsed bypass system, bubble chamber pictures have been taken at 100, 200, and 300 GeV, using low beam intensities while the bulk of the beam was being used for muon and neutrino production. Since the beam intensity delivered to the Neutrino Area never exceeded one hundredth of the design intensity, the amount of radioactivity produced outside the shields and dumps was proportionally quite small. A measurement of activity produced in soil directly above the target in the Neutrino Area supported this conclusion.

Water samples have been taken from all sumps along the beam lines (Figs. 3, 4) before the beam was extracted in order to establish normal background levels. From that time a number of sumps in the three experimental areas have been sampled on a routine basis to verify that no accelerator produced activities had reached them. Water from the closed loop magnet cooling systems in the Neutrino Area and in the Main Ring (main accelerator) and Booster (booster accelerator) have also been analyzed. The water samples were analyzed independently by the U. S. Testing Company, Inc. of Richland, Washington, and by NAL. Except for a minute amount of tritium in the closed loop in the Neutrino Area, no accelerator produced activity other than 53 day half-life Beryllium-7 has been found in the water. The Be-7 in the water has been concentrated into measurable quantities by capture in the ion exchange resin used to maintain low conductivity in the closed water cooling systems. Be-7 is subsequently released into the effluent from regeneration of the resin, and a

measurement made of its concentration and volume. Since the half-life of Be-7 is short; the relative velocity of the Be-7 ion very low[3], and the effluent is released inside the Main Ring where the Be-7 is trapped essentially forever, it cannot conceivably become a pollutant in domestic water. It does serve the useful function of giving an early warning of the production of 12.3 year half-life tritium.

Air-borne radioactivity was measured for the first time during accelerator operations this reporting period. The location chosen for monitoring was in the vicinity of the target inside the Neutrino Area Target Tube, a location which is probably the worst case in the whole site at the present time, i.e., concentrations are probably highest there. The beam travels through more than 10 meters (approx. 30 ft.) of air between the end of the beam pipe and the target, and the target is inside a 2 meter (6 ft.) diameter tube. The area is interlocked so that no persons are present when the beam strikes the target because the dose rate at a meter from the target is in excess of 1000 rem/hr at that time. Air was drawn into the tritium monitor through a plastic tube, the open end of which was 2 meters inside the Target Tube. The concentration of air-borne activity was found to be a few times maximum permissible concentration and it had a short enough half-life that it fell below maximum permissible levels by the time persons entered the tube.

Penetrating radiation is monitored at the Environmental Radiation Monitoring Station located in the NAL Village. No evidence of accelerator produced radiation has been seen by any detector in the station during the reporting period. The three gamma sensitive monitors (aluminum-argon ion chamber, tissue-equivalent ion chamber and NaI(Tl) crystal) have consistently indicated natural-background-level exposures of approximately 0.011 millirad/hour except for brief periods discussed in Section 4 when radon daughters in rain clouds were detected. The neutron monitors have indicated an average

neutron dose of approximately 0.0005 mrem/hr, which is consistent with the expected cosmic-ray neutron background.[4]

There were no unusual incidents or releases during the reporting period. In addition, there were no nonradioactive materials produced in quantities which could pollute the environment, and there were no abnormal natural occurrences which could have resulted from or have had some impact upon the facility or its operation.

3. Monitoring, Data Collection, Analysis and Evaluation

The three types of accelerator produced radiation meriting monitoring for environmental purposes are discussed below.

3.1 Penetrating Radiation

Operation of the accelerator at full design energy and intensity will inevitably result in production of some penetrating radiation (primarily neutrons and muons) outside the shielding. Although the shielding has been designed to be adequate for foreseeable circumstances, monitoring for purposes of determining actual radiation levels both on and off the site is necessary.

A central monitoring station is maintained in the NAL site "Village" for detecting penetrating radiation. The monitoring equipment consists of five major components.

A. Aluminum-Argon ionization chamber. This chamber is mostly sensitive to muons and gamma rays, and much less sensitive to neutrons. The data is recorded as daily integrals of the ionization current. A continuous strip-chart record of ionization current is also made.

B. Tissue-equivalent ionization chamber. This chamber is sensitive to neutrons as well as gammas and directly ionizing radiations. The data is recorded as daily integrals of the ionization current and as a strip-chart record of ionization current.

C. A 7.6 cm diameter x 7.6 cm (3 in. x 3 in.) NaI(Tl) radiation detector. This device is sensitive primarily to gamma radiation above 100 keV.[5] The data is recorded as daily integrals of the counts and as a strip-chart record of count rate.

D. Bonner spectrometer. This device is an array of moderating hydrogenous spheres with thermal-neutron sensitive Li-6 I(Eu) scintillators located at the center of each sphere. The data is recorded as the daily integral of counts in each detector. It may be unfolded by a computer program to obtain the neutron flux, dose and dose equivalent.

E. Precision reproducible (DePangher) long counter.[6] This device is a BF_3 proportional counter moderated by polyethylene to obtain an essentially energy independent response to neutrons up to about 14 MeV. The count rate from this device is thus a measure of neutron flux. The data is recorded as daily integrals of neutron counts.

A number of short periods of increased radiation levels have been observed with the ion chambers (A, B) and the NaI crystal (C) since the monitoring station was put into operation, but no correlation with accelerator production of penetration radiation has been found. These short periods became more frequent in the past six months and they were studied in some depth. Since the increase was most easily detected using the 7.6 cm x 7.6 cm NaI(Tl) detector, it was decided to record multichannel analyzer spectra from the NaI(Tl) detector to see if the spectrum was altered during the period of increased radiation. It was found that the peaks corresponding to daughters of radium were enhanced relative to the K-40 peak. The energies were measured precisely using a Ge(Li) detector to verify that the gamma rays were emitted by radium daughters.

It soon became apparent that there was a definite correlation between the presence of low-lying rain clouds and increases in radiation level. A one-to-one correlation was made between increases and precipitation as measured with the rain gage recently installed on the site by the State of Illinois Water Survey Division. It remained to collect a rain water sample and observe the activity directly. Since the daughters of interest have half-lives of approximately one-half hour, a fresh sample was collected during a shower for less than five minutes and was counted for 1000 sec starting about one minute thereafter. The quantities of Pb-214 and Bi-214 were found in concentrations of approximately 60 nCi/liter. The daughters of radium observed are also daughters of radon, a noble gas which is known to be released into the atmosphere naturally. Also, some radon is released in the process of burning fossil fuels, especially coal. The concentration of radon in the atmosphere is too low to explain the large increase in radiation level seen,[7] but a mechanism for concentrating the activity is provided by condensation of water vapor at the ionized atomic sites. A literature search revealed that concentrations of Pb-214 and Bi-214 in precipitation have been observed in excess of the above values.[8]

This effort in trying to understand the variations in the background rate may be justified from the point of view of legal protection of NAL. Neighbors using less sophisticated equipment than that available at NAL could erroneously assign the variations in the environmental radiation to the operation of the accelerator.

With the exception of the periods of increased radiation levels from radon daughters (the maximum increase being approximately three times natural background levels) the radiation level has remained relatively stable throughout the reporting period. The value of 0.006 mrad/hr reported in Calendar Year 1971 was measured inside the Environmental

Radiation Monitoring Station, a wooden frame house in the NAL Village. Subsequently, the radiation level has been measured outside using the tissue-equivalent ionization chamber calibrated using a radium source whose strength was determined by direct comparison with a radium source calibrated by the National Bureau of Standards. The values of the radiation level inside and outside the house were 0.008 and 0.011 mrad/hr, respectively. The latter value compares well with the values of the 0.012 to 0.013 mrad/hr reported for this area by the Health and Safety Laboratory[9] and 0.014 to 0.015 mrad/hr reported by Kastner, et al., using a similar chamber.[10]

As mentioned above, the Bonner spectrometer and the precision reproducible (DePangher) long counter are used to determine the neutron flux and dose. The use of the Bonner spectrometer for measurement of neutron backgrounds has been hampered by interference from gamma-ray interactions in the ^{6}LiI(Eu) scintillators. Experiments conducted here have demonstrated that most of the background can be eliminated by using pulse shape discrimination as follows:

The ^{6}LiI crystal is surrounded by a plastic scintillator forming a "Phoswich".[11,12] The electrons from gamma-ray interactions and charge particles from cosmic rays deposit energy in the small diameter ^{6}LiI crystal and the plastic scintillator shroud, and produce fast pulses of light in the plastic scintillator. The pulses from the interactions in the ^{6}LiI crystal give slow (long duration) pulses of light. The two pulses are separated electronically. Only the neutron initiated events ($n + {}^{6}Li \rightarrow {}^{4}He + {}^{3}H + Q$) are counted. Efforts are being made to obtain additional satisfactory Phoswiches and necessary electronics for converting the Bonner spectrometer to the pulse shape discrimination system.

The cosmic ray neutron background was measured using the precision reproducible (DePangher) long counter[6,13] which

differs from other long counters through the use of polyethylene instead of paraffin as the moderator, and by the use of a well defined (reproducible!) geometry. The long counter was calibrated using a Pu-Be neutron source whose neutron emission rate had been previously measured by the manganese sulfate bath technique at the National Bureau of Standards and at Argonne National Laboratory. The results of these two independent measurements agreed to within 1.7 per cent, the accuracy of the measurements. The neutron detection efficiency of the DePangher counter is essentially energy independent below 10 MeV.[6,13] The energies of neutrons emitted by the Pu-Be neutron source fall within this range and provide a good calibration for most purposes, e.g., reactor monitoring. However, the cosmic ray neutron spectrum has neutrons with much higher energies than 10 MeV.

The shape of the cosmic ray neutron spectrum has been extracted from Bonner sphere measurements[4] and it was used in conjunction with the measurements of the DePangher counter to determine the cosmic ray neutron flux and neutron dose equivalent assuming that the light building in which the counter was located did not distort the neutron spectrum significantly. The dose equivalent rate was calculated for bilateral isotropic irradiation of a 30 cm thick tissue slab (approximating the average man), using the neutron flux spectrum and dose equivalent conversion factors of Hajnal, et al.[4]

The calculated average cosmic ray neutron flux at the Environmental Radiation Monitoring Station in Calendar Year 1972 was 0.0104 n/(cm^2-sec) and the dose equivalent rate was 0.00055 mrem/hr (4.8 mrem/year). Correcting to sea level using the dependence of neutron flux on thickness of the atmosphere reported by K. O'Brien[14], the result becomes 0.0081 n/(cm^2-sec) in good agreement with 0.0082 obtained by Hajnal, et al.[4], and other values ranging from 0.0065 to 0.018 measured previously.[4]

3.2 Air-borne Radioactivity

Under normal operation radioactivation of air may occur in the vicinity of certain beam dumps and target boxes. Monitoring of such activation will be carried out for purposes of personnel exposure control. Under no circumstances is the off-site concentration of air-borne radioactivity expected to approach the limits set forth in the AEC Manual, Chapter 0524.

The first monitoring of air-borne radioactivity was carried out during this reporting period. A Johnston Laboratories, Inc. Triton Monitor Model 955B was calibrated using tritiated methane and used to sample gas from the Neutrino Area Target Tube. This location is inside an interlocked enclosure and represents a "worst case" on the site at the present time because the proton beam travels through more than 10 meters of air between the end of the beam pipe and the target, the target is inside a two meter diameter tube which somewhat restricts the flow of air, and the full intensity beam strikes the target. Actually, the plan calls for extending the vacuum pipe and for keeping the radioactive gases inside the building by turning off the ventilating fans during operation. All persons are excluded at this time because of high radiation levels. Since efforts are made to contain rather than exhaust the radioactive gases, concentrations measured by remotely collecting a sample from the Target Tube during operation should be far in excess of those which could be realized for venting the radioactive gas to the atmosphere. For example, if the gas from a 10 meter section of the Target Tube were mixed thoroughly with the air in the rest of the enclosure, a dilution of approximately 300 times would be achieved. Also, further dilution would occur before the gas reached the site boundary about 2000 meters away, and the short half-lives of the radioactive constituents[7, 15] would also reduce the environmental load greatly. The effects of

dilution by ever changing winds has been mentioned elsewhere.[16]

The Triton Monitor detects all radioactive gases. In fact, the far more energetic beta particles (almost two orders of magnitude higher than the 19 keV end point energy from the decay of the triton) emitted by ^{11}C, ^{13}N, ^{15}O, and ^{41}Ar produced in air result in a much higher reading than one gets from the same number of disintegrations of tritium. Thus, the use of tritium for calibration results in an overestimation of the decay rate and, hence, concentration of the radioactive gases. No correction was made for this overestimation which is expected to be greater than one order of magnitude. A table of maximum permissible concentrations (MPC's) for radioisotopes produced in air follows:[7, 15]

Table 1

Some Maximum Permissible Concentrations (Air)

	^{11}C	^{13}N	^{15}O	^{41}Ar	
$E_{\beta\ max}$	0.97	1.20	1.74	1.20	MeV
Radiation Worker (40 hr/wk)	59	40	27	47	$\mu Ci/m^3$
Population at Large	0.026	0.023	0.02	0.02	$\mu Ci/m^3$

Based on these concentrations, the radiation levels observed corresponded to a few MPC's for radiation workers. The concentrations fell below maximum permissible a few minutes after the beam was turned off. By comparison, radiation from the target at the sampling location would have resulted in a lethal dose in approximately one hour. Thus, when problems from radioactive gas appear, they should be easily manageable at this facility.

3.3 Water-borne Radioactivity

During accelerator operations, some radioactivation of the soil will occur.[17, 3] Leaching of these radionuclides into the ground water provides a possible mechanism for transport of NAL produced radionuclides into surface run-off waters and the aquifer. Hence, a broad program of ground water monitoring for radioactivity is maintained.

Monthly water samples are taken at various locations on the site and analyzed for the presence of those radionuclides which have been experimentally determined to be produced and to be leachable from NAL soils in measurable quantities.

The water sampling locations were chosen to sample two ground water systems:

1. Surface and near-surface waters. These samples were taken from sumps which collect water in the vicinity of accelerator components and from on-site streams and industrial holding ponds.

2. Silurian aquifer. These samples were taken from farm wells which tap the 70 foot silurian dolomite aquifer which is a prime water supply for many private residences in the area.

3.3.1 Water Sample Collection

Water samples collected from wells not in regular use are pumped for a sufficient length of time, usually two or three hours, to insure that the water standing in the pipe has been pumped out before a sample is taken. The water in the pipe could conceivably have been there since the last time a sample was taken. Normally, the pipe volume is pumped several times before sampling. Water samples from sumps, creeks, and other surface waters are normally collected by dipping a bottle well below the surface. Several of the sumps inside normally locked

enclosures are sampled by remotely operated peristaltic pumps or the sump pumps themselves.

The samples are treated with concentrated hydrochloric acid to prevent precipitation of radionuclides, prior to shipment to U.S. Testing Company in Richland, Washington. Samples collected from key locations where some radioactivation of water and/or soil may have occurred are counted before shipment using the thallium-activated sodium iodide gamma-ray detector in the NAL Nuclear Counting Laboratory. Also, samples from the closed loop magnet cooling systems are examined in the Counting Laboratory. The shipment each month includes a sample containing known amounts of several of the accelerator produced radionuclides to check the accuracy of the assays made by U.S. Testing Company.

The agreement of the reported concentrations with the known concentrations of radionuclides for these control samples provided verification that the analyses were meeting the specifications agreed upon in the contract with U.S. Testing Company. These specifications, given in Section 4, provide warning of the presence of radionuclides at concentrations far below the maximum permissible.

3.3.2 Analytical Procedures at U.S. Testing Company

Once the samples reach U.S. Testing Company they are subject to the appropriate one of the following analyses:

Type I. Test for Hydrogen-3 (tritium), Beryllium-7, Sodium-22, Calcium-45, Manganese-54, and Cobalt-60. Analysis Type I is performed on almost all samples.

Type II. Radium-226 and Thorium-232. This analysis is performed routinely only on samples from deep wells (greater than 250 feet deep) since no appreciable concentrations of these naturally

occurring radioisotopes have been detected in samples from shallower wells. This test was designed to establish normal background levels in water from surrounding communities and has been essentially completed.[18] Routine sampling of the one deep well on the NAL site for radium and thorium is being carried out to observe any long term changes in the concentrations which might signal a change in the pattern of water flow.

Type III Chemical separation of Calcium-45. If the concentrations of certain radionuclides ever become large, the detection of a low concentration of Calcium-45 in the presence of a high concentration of those radionuclides will be difficult. In those cases a chemical separation will be required before analysis.

Type IV. Tritium only. Because tritium has only a very low energy beta particle emission (19 keV end point), it is normally detected by intimate mixing with liquid scintillator. Analyses are usually used in conjunction with studies of closed water cooling systems.

The procedures used by U. S. Testing Company follow:

3.3.2.1 Gamma-Ray Spectroscopy

For gamma-ray pulse height analysis for National Accelerator Laboratory samples, the sample is homogeneously mixed and a 500 milliliter aliquot is transferred to a polyethylene bottle for a direct count. Ten per cent of all samples are done in duplicate.

A gamma-ray spectrum is obtained by plotting events detected by the analyzer on the ordinate and the pulse height on the abscissa. There is a Gaussian distribution about the gamma energy photopeak and a Compton continuum

extending from zero energy to a point a little below the photopeak. Bremsstrahlung interactions add to the complexity of the lower region of the energy spectrum. Peaks may also be present from pair production and backscatter events.

Although all of a spectrum is characteristic of a particular radionuclide in a given counting geometry, only the photopeak area is currently used to identify and measure the amount of the radionuclide present. If more than one radionuclide is present, their spectra are additive.

Resolution of a complex spectrum of gamma energies into its component parts requires a determination of the Compton or other contribution from each radionuclide present to the photopeak area of each of the radionuclides present.

For the resolution of four components, (in the case of National Accelerator Laboratory samples, Na-22, Co-60, Mn-54, and Be-7) four simultaneous linear equations comprising 4^2 coefficients (Compton and interference correction factors) are used. The gamma spectrometer used is calibrated for approximately 40 different radionuclides.

The system solutions are obtained by an iterative process performed by computer using an especially designed data processing program which converts from counts to number of disintegrations, including Compton and interference correction factors obtained from a calibration of a specified source-crystal geometry.

The gamma-ray detector used in the determinations is a Harshaw Chemical Company 23.8 cm diameter by 20.3 cm deep (9.375" x 8") thallium-activated sodium iodide crystal with a 7.6 cm diameter by 12.7 cm deep (3" x 5") well. The crystal is optically coupled to seven 5.1 cm (2") photomultiplier tubes. The resolution, full width at half maximum, for the peak corresponding to the 662 keV

gamma ray from ^{137}Cs is eleven per cent. A Packard Instrument Company 400 channel analyzer is used to accumulate the pulse height information. Instrument gain and backgrounds are determined every eight hours, or more frequently if counting data or other phenomena indicate significant background change or electronic gain drift. Counting efficiencies are checked once per eight-hour shift at four different energy levels: 0.06, 0.66, 1.16, and 2.50 MeV, using calibrated ^{241}Am, ^{137}Cs, and ^{60}Co sources. Count rates are expected to fall within two standard deviations of the expected values, determined from historical data. Also, the gain of the system must be such that the peak corresponding to ^{241}Am (0.06 MeV) fall within channels 5.98 and 6.02 and the peak corresponding to ^{137}Cs (0.66 MeV) fall within channels 66.06 and 66.26 in order for the unit to be considered calibrated for counting.

Radionuclide standards are purchased from Westinghouse Hanford Corporation, Richland, Wash., with $\pm$ 2 per cent stated accuracy. Efficiencies have been determined for various geometrical configurations based on multiple counts of replicate standards.

3.3.2.2 Alpha Counting

Proportional counters are used for determining the presence of thorium and radium by alpha-particle detection. Either an open window Nuclear Measurements Corporation or a 0.5 mg gold coated window Beckman Sharp Laboratories 5.7 cm diameter (2.25") proportional counter is used. Backgrounds are determined daily by an eight hour count. Operating plateaus are determined weekly and counting efficiencies twice daily using ^{239}Pu sources. Counting efficiencies are expected to fall within two per cent of the expected values as determined from historical data. Deviations

greater than this will require the instrument to be removed from use and returned when the problem is rectified. For the proportional counters, curves of counting efficiency versus precipitate weight on counting planchets have been constructed based on multiple counts of replicate standards at each point on the curves. This method of calibration serves to correct for such things as backscatter as well as air, window, and sample absorption.

Radium in National Accelerator Laboratory samples is separated from other interfering nuclides by coprecipitation with calibrated barium carrier, first as the carbonate, then as the nitrate, and finally as the chromate. The precipitate is transferred to a tared planchet, dried, weighed, then alpha counted immediately. If the first count is three or more times normal background fluctuation, decay counts are made at 3, 15, and 30 days from the date of separation. This method provides for a check on the ingrowth of radium daughters and also provides for the determination of ^{224}Ra and ^{226}Ra. The yield is determined by comparing the final precipitate to the amount added. The activity after conversion to disintegrations per minute from counts per minute is adjusted to 100 per cent with the yield factor.

When a thorium separation is requested for National Accelerator Laboratory samples, the sample is evaporated to near dryness and then the thorium is precipitated as a fluoride. This serves to isolate thorium from elements other than the rare earths and (IV) actinides. Iron, zinc, titanium, molybdenum, and tantalum sometimes coprecipitate as will the alkaline earths. Extraction of thorium with TTA in benzene, at a pH of 1 to 2, leaves rare earth, actinium and other elements extractable in the higher pH ranges remaining in the aqueous phase. Separation from zinc,

protactinium, plutonium, neptunium, iron (III), and tin (III) is accomplished by back-extraction of thorium into 2N HNO_3. After volume reduction the sample is transferred to a planchet for activity determination.

Samples are mounted on the planchets as securely as possible to prevent sample loss both outside and inside the counting chamber. This is accomplished by "flaming" or electrodeposition. Samples are not counted if they are likely to contaminate the counting chamber. In those cases new samples are prepared from additional aliquots. Planchet holders and sample changers are cleaned daily with mild detergent and/or decontamination solution and dried with alcohol. The Beckman detectors have windows which can be replaced should they become contaminated. Background stability is maintained by requiring results to lie within two times the expected standard deviation limits. If background counts fall outside these limits, the instrument is removed for service and returned to use when background determinations are normal. A high background is an indication of contamination; consequently is the first priority. Should decontamination fail to remedy the situation, the unit is checked for component malfunction.

3.3.2.3 Liquid Scintillation Counting

Concentrations of tritium and ^{45}Ca are determined by intimate mixing of the samples with liquid scintillator and counting in a refrigerated unit where the noise from the photomultiplier tubes has been greatly reduced.

Liquid scintillation counting is done using the Packard Instrument Company Model No. 3320 Tri-Carb Liquid Scintillation Spectrometer. Optimum operating

settings are determined prior to use for each radionuclide to be analyzed, using the appropriate source, i.e., ^{3}H and ^{45}Ca for National Accelerator Laboratory samples. One-tenth of all samples counted is background. One-tenth of all samples counted is a standard source. Every unknown is internally "spiked" after the original count, with a calibrated source, and recounted to accurately determine the quenching effect, or else duplicate samples are counted, one of which is spiked. Backgrounds are about 22 counts per minute. Counting efficiencies are expected to fall within four per cent of the expected values as determined by historical data. Deviations outside expected limits result in removal of the instrument from service until the problem is corrected.

Tritium in water is measured by a direct count of the sample in the liquid scintillation counter. Seven milliliters of sample is pipetted into 18 ml of PPO, POPOP, toluene, and Rohm and Haas Company Triton x-100. The mixture is transferred to the deep freeze and is allowed to remain there one hour before counting is begun. This waiting period allows temperature equilibrium to be reached and the decay of excitation from the plastic vials to occur. Duplicate samples are run for National Accelerator Laboratory and one is spiked with a known amount of tritium. This procedure allows both the determination of quenching and counting efficiency; the latter is approximately 55 per cent for 1 ml of sample in 25 ml of scintillation "cocktail".

Calcium-45 in water samples from NAL can also be measured by the above procedure. A known amount of ^{45}Ca is substituted for tritium in the spiked cocktail. However, the determination of ^{45}Ca, which has a very low maximum permissible concentration compared with

tritium (Section 4), is difficult to make in the presence of higher concentrations of tritium and other accelerator produced isotopes such as ^{22}Na. When there are interfering nuclides, a ^{45}Ca separation must be done and is described below.

To an aliquot of sample a known amount of calcium is added along with strontium hold-back carrier. This procedure involves a nitrate precipitation of calcium. The precipitate is dissolved in hydrochloric acid and evaporated to dryness. The salts are dissolved in distilled water and diluted to 10 ml. Calcium recovery is determined by Atomic Absorption Spectroscopy and the sample is counted before and after separation. The counting procedure is the same as above except that in this case the separated ^{45}Ca sample is counted, then spiked and recounted.

3.3.3 Results of Analyses

Samples were analyzed by U. S. Testing Co. from the locations shown on Figures 2, 3, and 4. A description of the waters sampled is given in Table 2, and elevations of sump pit bottoms and well water levels are reported in Table 3. The sampling frequencies, the type of analysis, and the results of the analyses are tabulated in Table 4. The concentration guides on which the specifications for the analyses were based are discussed in Section 4.

No measurable concentrations of accelerator produced radionuclides were found in surface or ground waters. Small concentrations of ^{7}Be and ^{3}H (tritium) were found in closed loop magnet cooling water systems.

Concentrations of long-lived tritium would be expected to increase even without an increase in beam intensity. However, beam intensity may increase still by more than one order of magnitude, hence ^{3}H concentration in the

Table 2
Description of Sampling Locations

Designation	Description	Water System Sampled
A1,A2,B1,C1, C2,D1,E1,E2, F1	Sumps adjacent to Main Accelerator enclosure	Shallow ground water from footings
g1,G1,G2,G3	Sumps along beam line between Transfer Hall and Neutrino Area Front-end enclosure	Shallow ground water from footings
H1	Central Utilities Building Cooling Pond	Industrial cooling water
H2A,H2B,H2C, H2D,H2E,H2F	Main Accelerator Cooling Pond	Industrial cooling water
H3	Village Oxidation Pond	Waste water
Mf1,MF1,MF2,MF3	Sumps along beam line between Transfer Hall and Meson Area Front-end enclosure	Shallow ground water from footings
MF4-MF12	Sumps in Meson Area	Shallow ground water from footings
N1	Sump in Neutrino Area Front-end enclosure	Shallow ground water collected in decay pipe underdrains
N2	Sump in Neutrino Area Enclosure 100	Shallow ground water collected in decay pipe underdrains
PE1,PE2	Sumps along beam line between Transfer Hall and Proton Area Front-end enclosure	Shallow ground water from footings
PE3,PE4,PE5	Sumps in Proton Area	Shallow ground water from footings
R1	Ferry Creek	Surface water
R2	Kress Creek	Surface water
R3	Indian Creek	Surface water
R4	Drainage Ditch between Neutrino and Proton Areas	Surface water
S1,S5,S9,S12, S16,S21	Sumps adjacent to Booster enclosure	Shallow ground water from footings
T1	Sump adjacent to extraction area in Transfer Hall	Shallow ground water from footings
V	NAL Village water supply	Silurian aquifer
W4,W7,W16,W21,W29, W38,W39,W43,W49, W50,W52,W55,W59, W64,W66,W68,W74,W75	Cased farm wells	Silurian aquifer

Table 3

Water Sampling Elevations

A. Approximate Well Water Levels

Designation	Elevation Above Sea Level* (feet)
W7	694
W16	694
W21	693
W29	701
W38	696
W39	701
W43	688
W49	708
W50	701
W52	696
W55	690
W59	689
W64	694
W66	708
W68	706
W74	700
W75	711

* Values obtained from Robert T. Sasman, State of Illinois, Water Survey Division, Preliminary Map #2, June 1969.

Table 3

Water Sampling Elevations

B. Elevations of Sump Pit Bottoms

Designation*	Elevation Above Sea Level (feet)
S1, S5, S9, S12, S16, S21	714
A1, A2, B1, C1, C2, D1, E1, E2, F1	714
g1, G1	714
G2	725
G3	732
N1	726
N2	731
Mf1	714
MF1	719
MF2	724
MF3	729
PE1	714
PE2	714
T1	714

* See Table 2 for description.

Table 4

Results of Water Sample Analyses†

Part A - Surface and Shallow Ground Water

Month	Jan.	Feb.	Mar.	Apr.	May	June
Batch No.	22	23	24	25	26	27
Location						
A1		None	None	None		
A2	None					
B1		None		None		None
C1			None		None	
C2	None					
D1		None		None		
E1			None		None	
E2	None					
F1		None		None		
g1					None	None
G1					None	None
G2					None	None
G3					None	None
H1	None	None	None	None	None	None
H2A		None				
H2B			None			
H2C				None		
H2D					None	
H2E						None
H2F	None					
Mf1					None	None
MF1					None	None
MF2					None	None
MF3					None	None
N1				None	None	None
N2		None	None	None	None	None
PE1					None	None
PE2					None	None
R1			None			
R2				None		
R3					None	
S1	None	None	None	None	None	None
S5		None	None			
S12			None	None	None	None
S21	None	None		None		
T1			None	None	None	

† (Footnotes follow Part B.)

Table 4

Results of Water Sample Analyses†

Part A (Cont.) - Surface and Shallow Ground Water

Month	July	Aug.	Sept.	Oct.	Nov.	Dec.
Batch No.	29	30	31	32	33	34
Location						
A1	None		None		None	
BO	None	None	None			None
B1		None		None		None
C1	None		None		None	
D1		None		None		None
E1	None		None		None	
F1		None		None		None
G1	None	None	None	None	None	None
H1	None	None	None	None	None	None
H2A		None				
H2B			None			
H2C				None		
H2D					None	
H2E						None
H2F	None					
H3	None					
MF4	None					
MF5	None		None	None	None	None
MF6	None					
MF7	None					
MF8	None					
MF9	None					
MF10	None					
MF11	None					
MF12	None					
N1	None	None	None	None	None	None
N2	None	None	None	None	None	None
PE3		None				
PE4		None				
PE5		None	None			None
R1			None			
R2				None		
R3					None	
R4	None	None	None	None	None	

† (Footnotes follow Part B.)

Table 4

Results of Water Sample Analyses†

Part A (Cont.) - Surface and Shallow Ground Water

Month	July	Aug.	Sept.	Oct.	Nov.	Dec.
Batch No.	29	30	31	32	33	34
Location						
S1	None	None	None	None	None	None
S5		None				None
S9			None			
S12	None	None	None	None	None	None
S16				None		
S21	None					
T1	None	None	None	None	None	None

†(Footnotes follow Part B.)

Table 4

Results of Water Sample Analyses†

Part B - Wells

Month	Jan.	Feb.	Mar.	Apr.	May	June
Batch No.	22	23	24	25	26	27
Location						
V						None
W4			None		None	None
W7						None
W21	None	None	None	None	None	None
W29	None				None	
W38		None		None		
W43	None	None	None	None	None	None
W49	None	None	None	None	None	None
W50			None			
W52	None				None	
W55			None			
W59						None
W64				None		
W66		None				None
W68				None		
W74			None			
W75	None				None	

† (Footnotes follow end of Part B.)

Table 4

Results of Water Sample Analyses†

Part B (Cont.) - Wells

Month	July	Aug.	Sept.	Oct.	Nov.	Dec.
Batch No.	29	30	31	32	33	34
Location						
V						None
W4	None	None	None	None	None	None
W7				None		
W16						None
W21	None	None	None	None	None	None
W29						None
W38				None		
W39		None			None	
W43	None	None	None	None	None	None
W49	None	None	None	None	None	None
W50	None					None
W52			None			
W55	None				None	
W59		None		None	None	None
W64		None	None			
W66				None		
W68					None	
W74	None					
W75			None			

† Interpretation of data entries:
A blank indicates no sample was taken. "None" means that none of the five radionuclides tested for was observed. Refer to Table 5 for applicable sensitivities. Well locations are shown on Figure 2.

Example: A sample was collected in July from well W21; no radionuclides were detected.

cooling water systems will increase considerably. Release of these radionuclides from closed loops will be carefully monitored and controlled. In 1972 a total of 2.9 mCi of ^{7}Be was released to the environment at an average concentration of 2×10^{-5} μCi/ml (maximum concentration 3×10^{-5} μCi/ml or 4.5 per cent of the MPC). This activity was released three feet below the surface of the soil inside the ring of the main accelerator. The short half-life (53 days) of ^{7}Be and its very strong chemical affinity with the soil insures that its release will place no burden on the environment. There was no known release of tritium or other radionuclides besides ^{7}Be in 1972.

For the purpose of determining the concentration of accelerator produced radioactivity in the soil, soil samples taken from just outside the Neutrino Area Target Tube were analyzed using the lithium drifted germanium (Ge(Li)) gamma-ray detector in the Nuclear Counting Laboratory. The soil was recovered from the region just above the target approximately five months after the first beam struck the target. Beam intensities during this period were below 10^{11} protons per pulse (less than 0.2 per cent of design intensity), hence the choice of one of the highest radiation areas as the sampling location was made.

The concentration of the 2.6 year half-life ^{22}Na activity, 10 to 20 per cent of which can be leached from the soil,[3] was determined to be 9×10^{-6} μCi/g just outside the 2 m dia. steel Target Tube. Assuming 20 per cent could be leached into a volume of water having 10 times the weight as the soil (the same ratio as was used in the leaching tests), the resulting concentration in the water would be approximately two per cent of the maximum permissible for general consumption (Section 4). The activity of ^{22}Na, which is probably the worst

environmental offender in soil of all the accelerator produced radioisotopes, had become barely detectable ($\approx 4 \times 10^{-7}$ μCi/g) an additional meter away.

3.4 Nonradioactive Pollutants

3.4.1 Purity of Drinking Water

The domestic water supply at NAL is provided essentially by two wells approximately 220 feet deep. One (W1 in Fig. 2) is located in the Central Laboratory Area and the other (V in Fig. 2) is in the Village. In case of low pressure, a third 220 ft. deep well (W3 in Fig. 2) is used to supply the additional water in the Central Laboratory Area. Samples of water from taps at 27 different locations throughout the Central Laboratory Area and its periphery have been analyzed throughout 1972 on a three sample per week basis for coliforms (bacterial contamination) by Aurora Clinical Laboratory, Aurora, Illinois. Samples of water from the domestic water supply in the Village have been analyzed by the State of Illinois twice a month during 1972. The high purity necessary for drinking has been maintained in the NAL domestic water supplies without excessive chlorination throughout this past year.

3.4.2 Tests for Pollutants in Waters Leaving the Site

All three creeks carrying water off the site (Fig. 2) have been analyzed monthly, by means of water samples sent to the State of Illinois, for various dissolved chemicals with good results. Ferry Creek which carries overflow water off site from the Village oxidation pond (H3 in Fig. 2), a holding pond for waste water from the Village, has been analyzed every two weeks since June 1972 by Tenco Hydro/Aerosciences, Inc. of St. Charles, Illinois for biological oxygen demand (BOD), suspended solids and pH with results verifying the absence of pollution. One complete analysis of the

dissolved salts was also made on the three creeks. The surface waters leaving the site have been clear of any harmful waste products during 1972.

3.4.3 Storage and Treatment of Waters Kept on Site

A new reservoir, Casey's Pond, is being filled and a pumping station built to increase our water reserves for times of inadequate rainfall. Because of the abundant rainfall at the present time, it may be possible to fill it without using water diverted from the Fox River which was polluted in 1972 according to results reported for samples we had analyzed.

A new sewage plant in the Central Laboratory Area will be completed in early 1973. The dissolved oxygen level, BOD, pH, settled and suspended solids content of the Village oxidation pond effluent are determined at the present time at NAL by an operator licensed by the State, and results are reported to the Bureau of Water Pollution Control in Springfield, Ill. The same operator will analyze the new plant operation effluent and forward results to the State.

Our policy with regard to chemical treatment of our water system is to use the least possible amount, partly to prevent our site from becoming contaminated by noxious chemicals, and partly to protect wildlife and fish. We employ a biochemist as a consultant to advise us on the proper use of chemicals. The Main Ring cooling ponds have so far not been much of a problem. We have used a little copper sulfate to inhibit algae growth. However, it was necessary to treat the Booster Pond (H1 in Fig. 2) with an algaecide and the Village oxidation pond with a chemical specifically for killing duckweed last year.

4. References

The concentration guides (Maximum Permissible Limits) used in the analyses of the water samples were taken from the Atomic Energy Commission Manual, Chapter 0524, Annex A, Table II, Column 2 (Water in Uncontrolled Areas) and reduced by a factor of three as appropriate for a suitable sample of exposed population. The smaller of the values given for soluble and insoluble forms has been used in each case. The specifications are given in Table 5. The concentration guides for air-borne activity were taken from the same source, Table II, Column 1 (Concentrations in Air in Uncontrolled Areas), and divided by a factor of three for application to populations.

The appropriate standard for penetrating radiation applied to populations was taken from R. R. Wilson's directive to M. Awschalom which states that the radiation contributed to the off-site environment shall be less than ten millirem per year. It must be noted that this directive is seventeen times more stringent than the limits set by the AEC Manual, Chapter 0524, Paragraph II.A, i.e., one hundred and seventy millirem per year.

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Table 5

Specifications for the Analyses of Radionuclides in Water

Radionuclide	Concentration Guide μCi/ml	Specified* Sensitivity μCi/ml	Specified* Precision μCi/ml
^{3}H	1×10^{-3}	3×10^{-6}	3×10^{-6}
^{7}Be	6.7×10^{-4}	5×10^{-7}	5×10^{-7}
^{22}Na	1×10^{-5}	3×10^{-7}	3×10^{-7}
^{45}Ca	3×10^{-6}	3×10^{-7}	3×10^{-7}
^{54}Mn	3.3×10^{-5}	5×10^{-8}	5×10^{-8}
^{60}Co	1×10^{-5}	1×10^{-7}	1×10^{-7}

* The precision and sensitivity are stated for the 68% confidence level (one standard deviation).

5. Acknowledgements

The author wishes to acknowledge the assistance of M. Awschalom, W. Bosworth and P. Gollon of the National Accelerator Laboratory, R. Sasman of the Illinois State Water Survey, K. O'Brien of the Health and Safety Laboratory, M. Lardy and H. Oens of the U.S. Testing Company, in the preparation of this report.

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