national accelerator laboratory
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ENVIRONMENTAL MONITORING REPORT
For Calendar Year 1973

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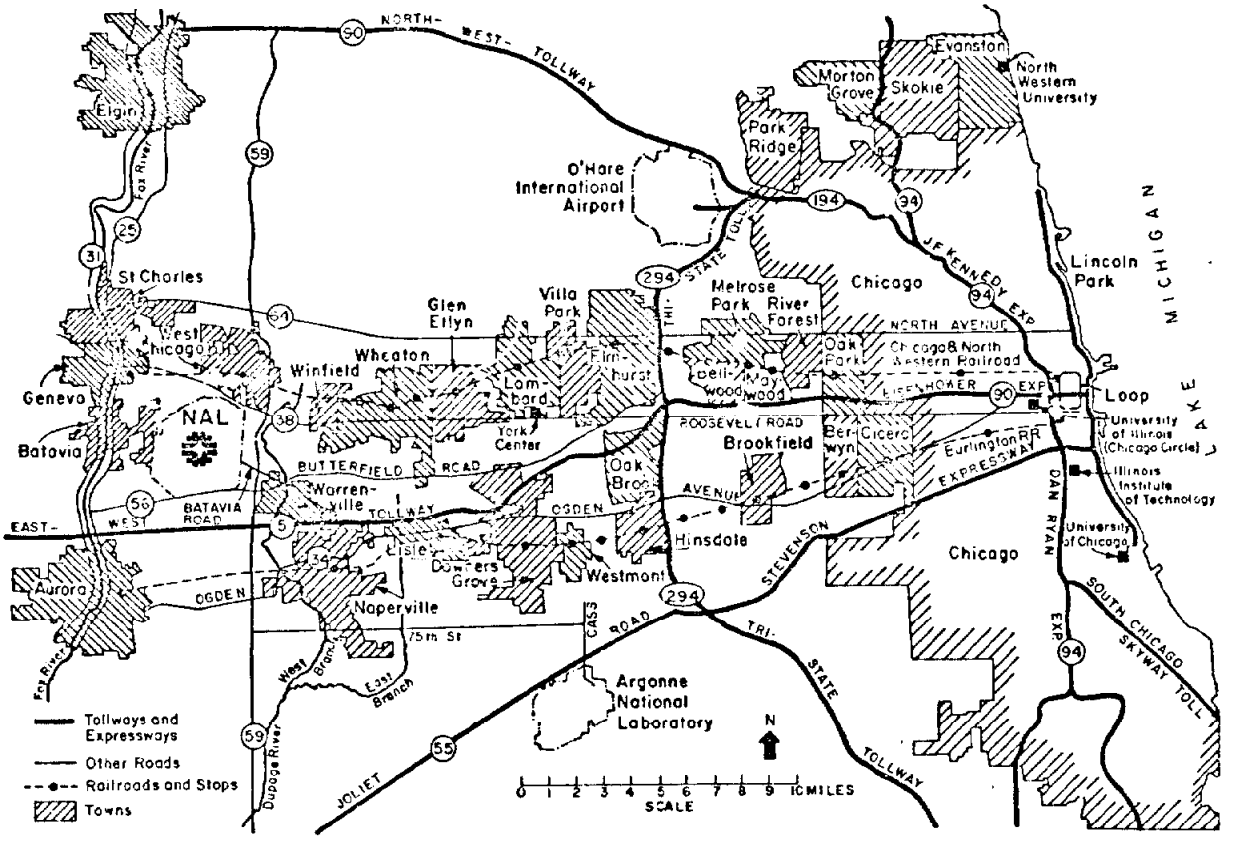
1. Introduction

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 with several weeks at 400 GeV in 1973. 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 rainfall on site during 1973 was 36 inches. 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,

Figure I - Location of National Accelerator Laboratory (NAL)

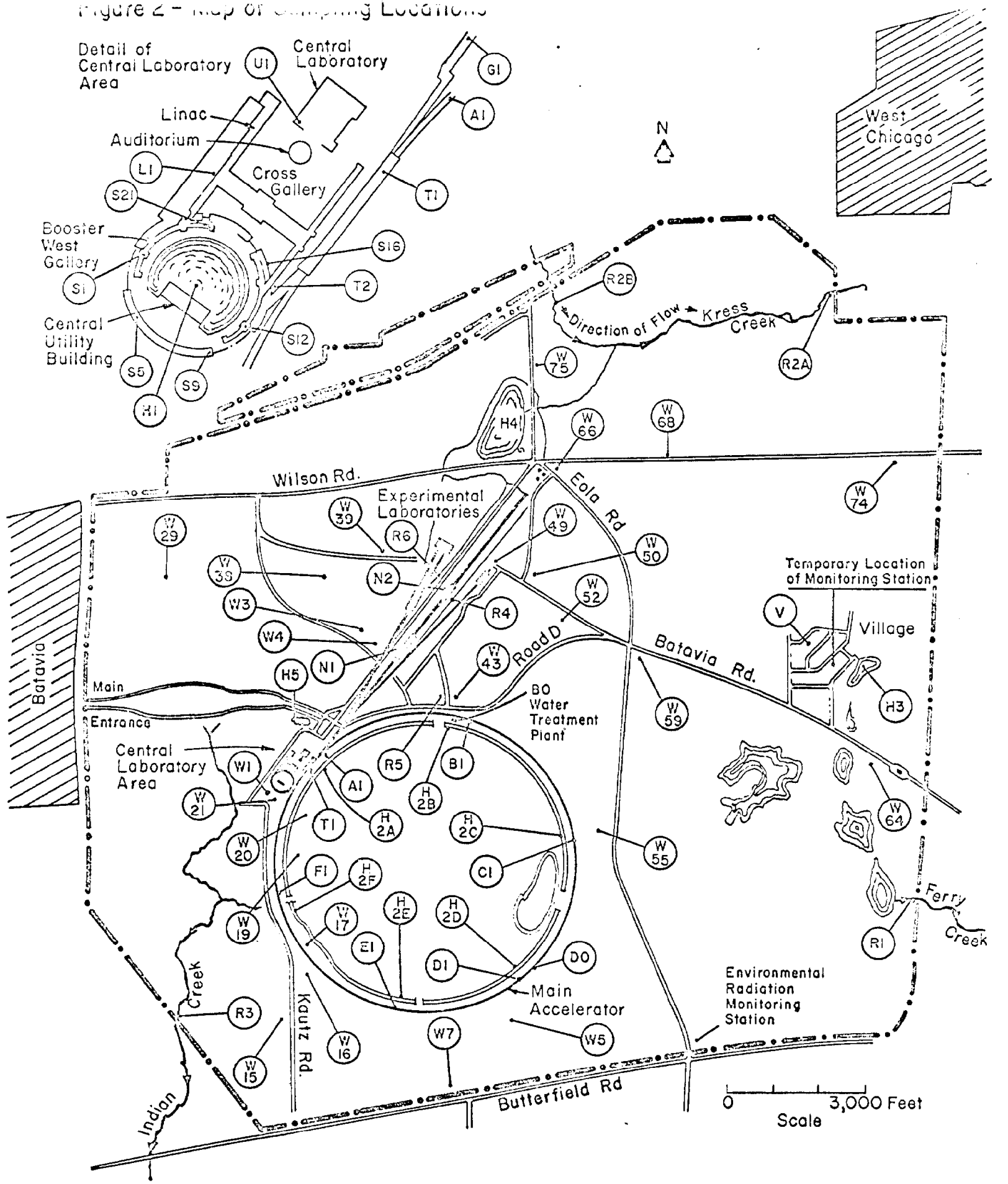


toward the Fox River. The drinking water in many of the surrounding communities comes from deep wells usually drilled 1,200 feet deep into the Cambrian Ordovician aquifer system[1].

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 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 has been in operation throughout 1973 except during an accelerator maintenance and development period in October when it was moved from its temporary quarters in a house in the NAL village to its permanent location on the modified second floor of a barn near the southern boundary of the Laboratory (Fig. 2). The present location has the advantage that the measurements will not be affected by the use of calibration sources in the village as were previous measurements. In addition to monitoring the radiation near the site boundary, the station will provide a reference background level for the Mobile Environmental Radiation Laboratory (MERL), a four-wheel-drive vehicle specially equipped to seek radiation levels

Figure 2 - Map of Sampling Locations



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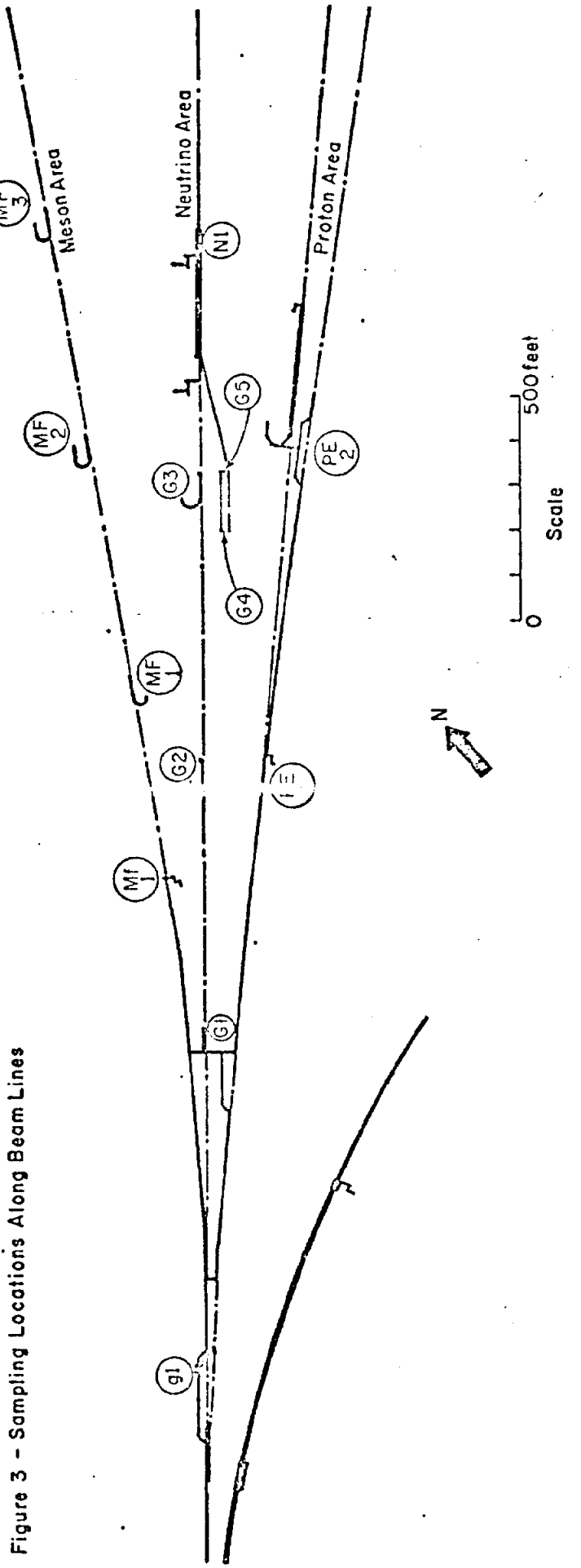

Figure 3 – Sampling Locations Along Beam Lines

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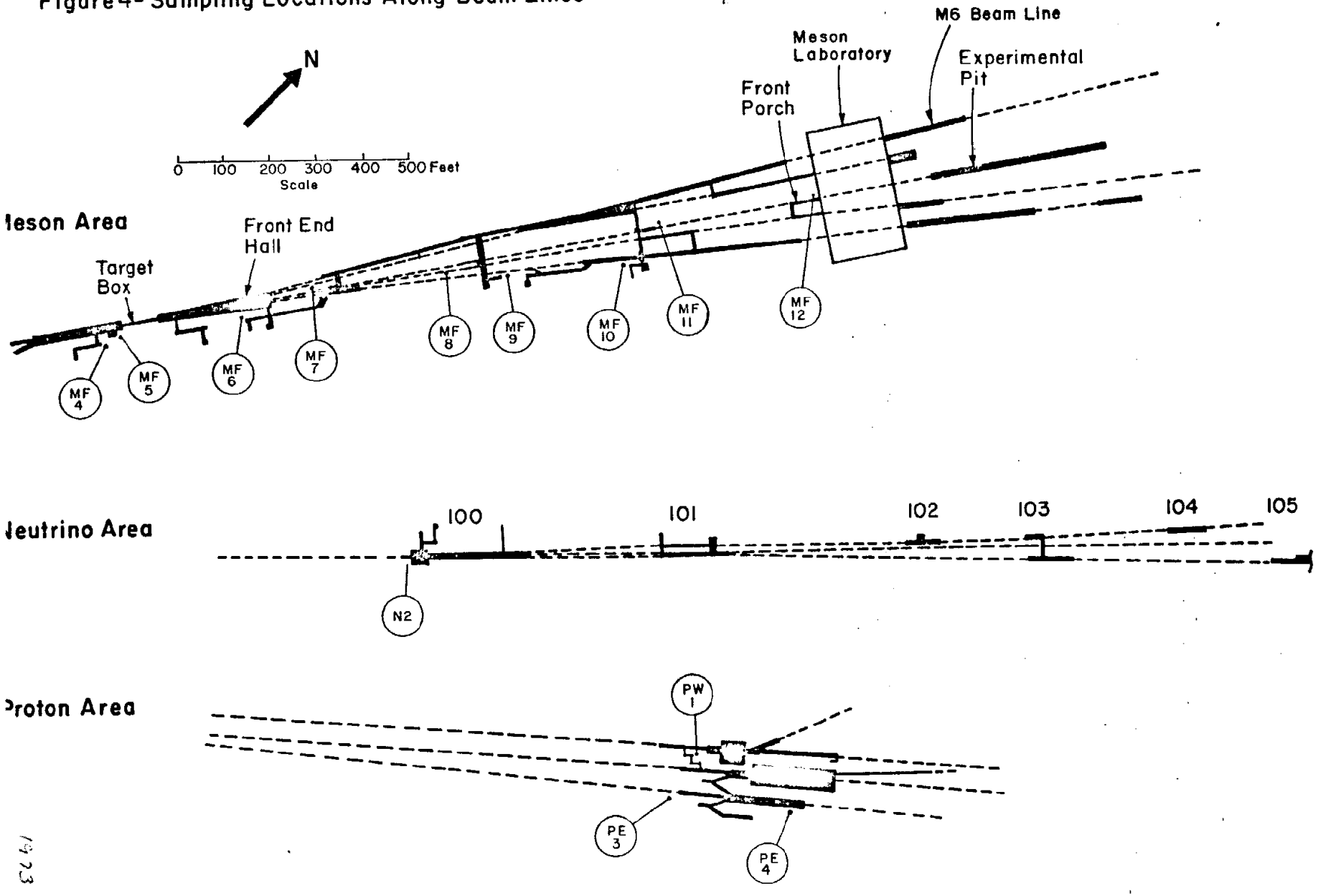
Figure 4- Sampling Locations Along Beam Lines



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above normal background from neutron, gamma-ray and charged particle sources. The mobility of the MERL is very desirable in view of the long distance from one corner of the site to the other, or from the origin of a beam line to its terminus. The MERL is particularly valuable for evaluating a new set of operating conditions, e.g., when a new experiment becomes operational.

To augment the single mobile laboratory, a large network of radiation detectors fixed in place send information back to a central data collecting area. Their primary function is to warn personnel of increases in the radiation levels in occupied areas, but an increasing number are placed strategically for environmental monitoring.

Water samples collected monthly at NAL are analyzed for residual radioactivity by an independent testing laboratory as well as in the NAL Nuclear Chemistry and Counting Laboratories. The analyses are performed specifically for accelerator-produced nuclides. Natural background levels of radium and thorium have been measured both on and off site and, except for a monthly check of those levels in our deep well, that program is being terminated. 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 fallout 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

During 1973 the intensity of the accelerator was steadily increased. Three times as many protons were

accelerated in the second half of the year as in the first half. Beam splitters were placed in operation and allowed the three external areas (Meson, Neutrino and Proton Areas) to receive portions of the same beam pulse concurrently. In addition, using a pulsed bypass system, pictures were taken at both the 30" and 15' bubble chambers. The highest intensity beam, reaching a maximum of about 10 per cent of design intensity, was delivered to the Neutrino Area. The Meson and Proton Areas received considerably less beam than the Neutrino Area in 1973, but efforts are underway to improve their beam handling capabilities and higher intensities can be expected in those areas in 1974. The Internal Target Area samples all of the beam accelerated but remains relatively free of residual radioactivity because its targets are so thin. Targets there are placed in the beam during the acceleration cycle and are not permitted to scatter enough beam to interfere with accelerator operation.

Efforts have been made in the design and construction of the facility to target correctly and to dissipate unwanted beam in properly shielded dumps to protect personnel and the environment. To verify that the induced radioactivity is properly contained, a broad program of water sampling and soil activation studies is being maintained. Wells and creeks on site were sampled routinely for accelerator produced activity. Also, water samples from sumps, closed loop magnet cooling systems, ponds, rivers near the site and other sources are being collected. Water samples were analyzed independently by the U.S. Testing Company, Inc. of Richland, Washington, as well as at NAL in 1973.

Tritium is now found in a few sumps and in the closed loop magnet cooling water systems. Be-7 is concentrated into measurable quantities by capture in the ion exchange resin used to maintain low conductivity in the closed loop 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[2], and the effluent 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.

The program of soil activation studies was increased in 1973. Soil samples were collected from holes drilled at several locations on site. Most samples contained no accelerator produced activity. The most radioactive sample, collected near the Abort Dump System in the Main Ring, was analyzed independently at Argonne National Laboratory. Additional theoretical calculations of percolation rates for radioactivity through NAL soils were made in 1973 and were consistent with the maintenance of negligible radiation levels in the aquifer.

Air-borne radioactivity has been measured at several locations on site where beam losses occur. The highest levels were detected in the Neutrino Area Target Tube (N1 on Fig. 2). Air is exhausted from the Target Tube through a stack almost 100 meters away. The radioactivity in the gas leaving the stack was almost exclusively Carbon-11 as determined from its decay half-life of 20 minutes. The concentration leaving the stack was

measured under a number of different operating conditions and found to present no hazard off site, and a radiation hazard on site only within a few feet of the exhaust outlet. The average concentration at the exhaust fan outlet was 7$\mu Ci/m^3$ and the total activity released in 1973 was 8 curies.

Penetrating radiation is monitored at the Environmental Radiation Monitoring Station near the site boundary and at a number of locations near the beam lines. 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 0.012 millirem/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[3].

Surveys of neutrons and muons penetrating the shields in the Meson and Neutrino Areas were made during operation at the highest energies introduced to those areas to date (300 GeV protons in the Meson Area and 400 GeV in the Neutrino Area). Measurements were made at the points where straight extensions of existing beam lines intersect Wilson Road (Fig. 2), and no levels above background were detected.

There was an unusual occurrence during the reporting period, a fire in the Meson Area (Fig. 4) on 12/26/73. The fire in the M6 beam line enclosure lasted only 30 minutes but released large volumes of dense smoke through

the ignition of "Isofoam" insulation, a polyurethane foam sprayed on the inner surface of the enclosure. The Meson Area was in the middle of a long scheduled shutdown at the time and no one was injured. The smoke created no off-site hazard and soon dissipated. There were no abnormal natural occurrences which could have resulted from or have had some impact upon the facility or its operation. The total exposure to the general population off-site was less than one man-rem for 1973.

3. Monitoring, Data Collection, Analysis and Evaluation

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

3.1.A 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 monitoring station is maintained for detecting penetrating radiation. It was located temporarily in the NAL village until October 23, 1973, when it was moved to permanent quarters on Eola Road (Fig. 2) near the site boundary. The monitoring equipment consists of five major components.

1. 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 hourly integrals of the ionization current. A continuous strip-chart record of ionization current is also made.

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

3. 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[4]. The data is recorded as hourly integrals of the counts and as a strip-chart record of count rate.

4. 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.

5. Precision reproducible (DePangher) long counter[5]. 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 hourly integrals of neutron counts.

During 1973 tests were made of a new "Phoswich"[6,7] detector system for the Bonner spectrometer. This system will be installed in 1974 in place of the present system. Due to interference from gamma-ray interactions, the data from the present system is very difficult to interpret. Thus, data collection was suspended in 1973 and will be resumed when the Phoswich system is installed.

Data from the new location for the Environmental Radiation Monitoring Station is transmitted to a central location near the Accelerator Control Room where it is processed by the "Aardvark" data logger, along with information from radiation detectors elsewhere on site.

The data is stored on magnetic tape and a daily printout with hourly radiation levels is obtained.

No increases in background level which correlate with accelerator operation were detected at the Monitoring Station during 1973. The system operated reliably during the year; however, while it was located in the village it was turned off periodically so radioactive sources being used in the vicinity would not result in false readings. The move to the new location will eliminate these periods. Also, the output from a 7.6 cm diameter x 7.6 cm NaI(Tl) detector located at Site #43 (Fig. 2) was recorded during 1973 and used as a backup and as a second system to detect increased radiation levels from radon daughters in rain clouds.

All increases in background were small and all except one were of the same magnitude on both NaI(Tl) detectors. The one anomalous event was detected by the detector at Site #43 on November 19, 1973, but not by any detector at the Monitoring Station. The event was of short duration and increased the background level to twice its normal level. It may have been correlated with beam tuning in progress in the vicinity at that time.

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 cosmic-ray neutron background was measured using the precision reproducible (DePangher) long counter[5,8], 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[5,8]. 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[3] 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 (approximately the average man), using the neutron flux spectrum and dose equivalent conversion factors of Hajnal, et al.[3].

The calculated average cosmic-ray neutron flux at the Environmental Radiation Monitoring Station in calendar year 1973 was 0.0092 n/cm^2-sec) and the dose equivalent rate was 0.00049 mrem/hr (4.3 mrem/year). Correcting the sea level using the dependence of neutron flux on thickness of the atmosphere reported by K. O'Brien[9],

the result becomes 0.0072 n/(cm^2-sec) in good agreement with 0.0082 obtained by Hajnal, et al.[3], and other values ranging from 0.0065 to 0.018 measured previously[3].

The tissue-equivalent ionization chamber was calibrated using a radium source whose strength was determined by direct comparison with a radium source calibrated by the National Bureau of Standards. The chamber was then taken outside and a measurement made of the natural background. The aluminum-argon ionization chamber and the NaI(Tl) scintillator were standardized to agree with the tissue-equivalent ionization chamber several years ago for measurements made inside the house in the village. The three detectors have been tracking well since that time. The background levels inside the new location for the Environmental Radiation Monitoring Station are quite similar to those in the wooden house in the village, even though the outer wall is of cement block construction.

The natural-background-level outside as determined by detectors inside the station has been 0.012 millirem/hr except for brief periods when radon daughters in rain clouds were detected. This 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[10] and 0.014 to 0.015 mrad/hr reported by Kastner, et al., using a similar chamber[11].

During 1973, measurements were made of the penetrating radiation at many different locations around the accelerator on a 24 hour per day basis. At a location where beam losses were typical of the Main Ring there were 15 days when the radiation levels were greater than 50 per cent

above background. The dose at the site boundary was calculated[12], based on the condition that losses of the same magnitude occurred everywhere else around the Main Ring. The resultant dose at the site boundary was 0.013 mrem for 1973 or less than 0.01 per cent of the AEC criterion of 0.17 rem (maximum) for the general population. There was one location around the Main Ring (above the Transfer Hall where the beam is extracted from the accelerator) which produced radiation above background levels on 160 days. The dose at the nearest site boundary from that source was less than 0.1 per cent of the criterion.

3.2.B 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.

Radioactive gas, primarily Carbon-11, was monitored continuously during all releases from the stack in the Neutrino Area Train Spur until October 15, 1973, and then periodically thereafter the remainder of the year. Measurements were made under the various operating conditions such as with and without beam pipe in place in the Neutrino Target Hall and calculations were made of the expected concentration at the site boundary.

The monitoring of air-borne radioactivity was carried out using a Johnston Laboratories, Inc. Triton Monitor Model 955B calibrated using tritiated methane. The meter reading of the 955B is greater for a given concentration

($\mu Ci/m^3$) of Carbon-11 than for the same concentration of tritium because the beta particle emitted by C-11 has a much higher end-point energy than the tritium beta. According to measurements and calculations made by A. Peetermans[13], the meter reading of the 955B for C-11, N-13 or O-15 is approximately five times that for the same concentration of tritium. Hence, this factor is applied in determining the C-11 concentration.

A table of maximum permissible concentrations (MPC's) for radioisotopes produced in air follows[14].

Table 1

Calculated 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	$\mu Ci/m^3$

Measurements were made at the exhaust fan in the Neutrino Area Train Spur Stack and the highest concentration observed was $15 \mu Ci/m^3$. From the decay half-life it was determined that the activity in the gas at that point was essentially C-11. A calculation based on a Gaussian plume diffusion model[15], was made to determine levels at the nearest site boundary more than 1600 meters away. Under typical wind conditions the expected concentration at the site boundary would be approximately $5 \times 10^{-6} \mu Ci/m^3$ when it was $15 \mu Ci/m^3$ at the stack. Using the MPC for the population at large given

for C-11 in Table 1, one obtains 0.03 mrem/year under around-the-clock operation at 5 x 15^{-6} μCi/m^3. Hence, the quantity of air-borne activity released by the Laboratory to the general population in 1973 was small. One can calculate the total exposure to the population off site using the above radionuclide concentration information and the distribution of population around the Laboratory, obtained from Civil Defense Research Section of the Health Physics Division ORNL, as shown in Table 2. Even using the conservative assumption of continuous operation at the highest observed release rates, one obtains a total exposure for calendar year 1973 of less than one man-rem to the general population.

3.3.C Water-borne Radioactivity

During accelerator operations, some radioactivation of the soil will occur[2,16]. Leaching of these radionuclides into the ground water provides a possible mechanism for transport of NAL produced radionuclides into surface run-off waters and 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 in 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

Table 2

INCREMENTAL POPULATION DATA IN VICINTY OF NAL, 1970

DISTANCE, MILES FROM CENTER OF MAIN RING / DIRECTION	0-2	2-3	3-4	4-5	5-10	10-20	20-30	30-40	40-50	50-60	60-70
N	97	0	0	73	607	72549	59787	48373	28900	29156	2521
NNE	0	0	2306	1338	3728	75631	77070	108837	131661	102076	12908
NE	0	0	2692	4815	10321	63960	306836	123882	0	0	
ENE	23	0	1587	20	44882	211107	864920	649681	0	0	
E	0	0	0	0	11567	185533	1144118	1061396	0	0	1363
ESE	0	0	1998	2403	11764	74815	304494	629984	385309	193880	5960
SE	0	0	1657	0	22797	30689	39084	106622	30481	25141	1066
SSE	0	33	0	0	0	29540	124447	4559	15496	67239	1147
S	0	316	0	0	1338	4201	9058	10815	10829	8939	308
SSW	0	45	1326	7579	44014	5063	1470	13488	6673	26103	1594
SW	0	0	3053	1009	34667	10155	14275	4543	27436	37847	1094
WSW	0	0	1671	3172	1733	4569	6054	5271	10716	6700	1286
W	0	0	3732	0	0	4059	2320	4039	11657	8043	2932
WNW	0	1143	5987	55	184	3109	48301	4087	11213	44197	1180
NW	69	0	890	3585	7003	1232	7547	3978	46860	165281	6808
NNW	0	0	362	3353	14633	20243	7256	23393	10634	28732	1054
TOTAL	189	1537	27261	27402	209288	796455	3017047	2802948	728965	743334	41227
CUMULATIVE TOTAL	189	1726	28987	56389	265677	1062132	4079179	6882127	7611092	8354426	876670

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 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 before shipment to U.S. Testing Company in Richland, Washington, to prevent precipitation of radionuclides. Samples collected from key locations where some radio-activation 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-

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[17]. 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 these 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. At least one sample in each shipment is analyzed twice.

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: 0.060, 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 ^{141}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, Washington, 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, protactinum, 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.

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 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. Ten per cent of all samples counted contain no activity and thus establish background levels. Ten per cent of all samples counted contain known amounts of activity for calibration of the system. 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 Trition 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 Company from the locations shown on Figures 2, 3 and 4. A description of the waters sampled is given in Table 3, and elevations of sump pit bottoms and well water levels are reported in Table 4. The sampling frequencies, the type of analysis, and the results of the analyses are tabulated in Table 5. The concentration guides on which the specifications for the analyses were based are discussed in Section 4.

Table 3

DESCRIPTION OF SAMPLING LOCATIONS

DESIGNATION	DESCRIPTION	WATER SYSTEM SAMPLED
A1,B1,C1,D0, D1,E1,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
G4,G5	Sumps in Target Service Building	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
H4	Casey's Pond	Industrial cooling water
H5	Reflecting Pond	Surface water
L1	Linac Lower Gallery	Shallow ground water from footings
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 Target Hall	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

DESCRIPTION OF SAMPLING LOCATIONS (Cont.)

DESIGNATION	DESCRIPTION	WATER SYSTEM SAMPLED
PE3,PE4,PW1	Sumps in Proton Area	Shallow ground water from footings
R1	Ferry Creek	Surface water
R2A,R2B	Kress Creek	Surface water
R3	Indian Creek	Surface water
R4	Drainage ditch between Neutrino and Proton Areas	Surface water
R5	Drainage ditch by Magnet Factory	Sump water
R6	Drainage ditch east of Meson Area	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
T2	Sump near 8GeV Transfer line	Shallow ground water from footings
U1	Sump in Central Lab	Shallow ground water from footings
V	NAL village water supply	Silurian dolomite aquifer
W1	NAL Central Lab area well	Silurian dolomite aquifer
W4	NAL deep well	Cambrian-Ordovician aquifer
W5,W7,W17,W19, W20,W21,W29,W33, W39,W43,W49,W50, W52,W55,W59,W64, W66,W68,W74,W75	Cased farm wells	Silurian dolomite aquifer

Table 4

WATER SAMPLING ELEVATIONS

A. APPROXIMATE WELL WATER LEVELS

DESIGNATION	ELEVATION ABOVE SEA LEVEL* (FEET)
W5	693
W7	694
W17	693
W19	693
W20	693
W21	693
W29	701
W38	696
W39	701
W43	688
W49	708
W50	704
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 4

WATER SAMPLING ELEVATIONS

B. ELEVATIONS OF SUMP PIT BOTTOMS

DESIGNATION*	ELEVATION ABOVE SEA LEVEL (FEET)
S1,S5,S9,S12,S16,S21	714
A1,B1,C1,D0,D1,E1,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 3 for description.

Table 5

RESULTS OF WATER SAMPLE ANALYSES†

PART A - SURFACE AND SHALLOW GROUND WATER
Tritium Concentrations in pCi/ml

Month / Location	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
A1	N		N		1		N		N		N	
B1		N		N		N				N		N
C1	N		N		N		N		N		N	
D0	N	N	N		N	N	N	2	1	1	N	
D1		N		N		N	N			N		N
E1	N		N		N				N		N	
F1		N		N		1				N		N
g1												
G1	N	N	N	N	N	N	N	N	N	N	N	N
G2												
G3												
G4									3	N	2	N
G5									N			
H1	N	N	N	N	N	N	N	N	N	N	N	N
H2A		N						N				
H2B			N						N			
H2C				N						N		
H2D					N						N	
H2E						N						
H2F	N						N					
H3						N						
H4						N						
H5								1		N		
L1										N	1	
MF4											277**	
MF5	N	N	N	N	N	N	N	N	23*	1	1580**	5
N1	N	N	1	N	N	N	3	4	2	4	N	1
N2	N	2	N	N	N	N		9	2	3	2	N

PART A (Cont.)

Month / Location	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
PE4	N	N	N	N	N	N		2	6	10	9	6
R1			N						N	N		
R2A				N						N		
R2B										N		
R3					N						N	
R4	N	N	N	N	N	N	N	N	1	N	N	N
R5				N								N
S1	N	N	N	N	1	N	N	2	2	1	1	N
S5									N			
S9									N			
S12	N	N	N	N	N	N	N	1	N	1	N	2
S16		N							N	N		
S21			N								N	
T1	N	N	N	N	N	1	N	3	2	3	2	3
T2											1	
U1										N		

Table 5

RESULTS OF WATER SAMPLE ANALYSES†

PART B - WELLS

Month / Location	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
V												N
W1		N										
W4	N	N	N	N	N	N	N	N	N	N	N	N
W5		N				N				N		
W7		N				N				N		
W17										N		
W19					N							
W20			N				N				N	
W21	N	N	N	N	N	N	N	N	N	N	N	N
W29				N				N				
W38		N					N					N
W39			N				N				N	
W43	N	N	N	N	N	N	N	N	N	N	N	N
W49	N	N	N	N	N		N	N	N	N	N	N
W50				N				N				N
W52	N				N				N			
W55				N				N				N
W59	N	N	N	N	N	N	N	N	N	N	N	N
W64			N				N		N			
W66		N				N				N		
W68					N							
W74	N						N					
W75	N				N				N			

† No other radionuclides detected except in four samples Be-7 at concentrations less than 1.5 pCi/ml or 0.2 per cent of MPC for consumption by the general population.

Interpretation of data entries:
A blank indicates no sample was taken. "N" means that none of the six radionuclides tested was observed or that if tritium was observed its concentration was less than one pCi/ml. Refer to Table 7 for applicable sensitivities.

Footnotes (Cont.)

* A release from the closed loop magnet cooling system occurred on the day this sample was collected.

** The samples were recounted by U.S. Testing Company with similar results. MF4 and MF5 samples collected in January, 1974 were counted by Eberline Instrument Corp. of West Chicago, Illinois, with results less than 10 pCi/ml. These sumps collect water from underdrains beneath the Meson Area Target Box (Fig. 4), so the high concentrations could result from activity produced just outside the Target Box. Further investigation is in progress.

Measurable concentrations of accelerator-produced tritium began to appear after the middle of the year in sumps collecting water from areas where significant beam losses or dumps occur. In some areas such as at Neutrino Area enclosure 100, N2 in Figure 2, it is possible for scattered radiation to activate water standing in the sump. This was proved by hanging a polyethylene bottle filled with water in the sump pit and sending it for analysis after a period of accelerator operation.

Concentrations of tritium in the closed loop systems are also increasing. The Be-7 produced at the same time is removed by the resin used to maintain the low conductivity of the water. Concentrations of long-lived tritium would be expected to increase even without an increase in beam intensity. However, beam intensity may increase still one order of magnitude, hence, ^{3}H concentration in the cooling water systems will increase considerably. Release of these radionuclides from closed loops will be carefully monitored and controlled. In 1973 a total of 146 mCi of Be-7 was released at an average concentration of 2.5×10^{-4} μCi/ml or 38 per cent of the MPC. This activity was released into the soil six feet below the surface inside the Main Ring. The short half-life (53 days) of the ^{7}Be and its very strong chemical

affinity with the soil insure that its release will place no burden on the environment.

The total release of tritium, almost all of it remaining on site, for 1973 was 4.3 mCi at an average concentration of 17 pCi/ml or 1.7 per cent of the maximum permissible for drinking purposes. The tritium has a half-life of over 12 years; hence, the build-up of levels in the ponds on site from the releases which occur primarily through losses of water from the closed loop cooling systems is being closely monitored. Water from the ditches in the external experimental areas flowed into Casey's Pond throughout 1973 until December when the pond became full and water had to be diverted around it into Kress Creek. There was an unplanned release (leak) of about 0.1 mCi of tritium from a Meson Area closed loop magnet cooling system into the M4 pit in the front end hall (Fig. 4) on December 10, 1973 at a concentration of 12 pCi/ml. Dilution between the Meson Area and the site boundary is estimated to have reduced that concentration to about 1 pCi/ml or 0.1 per cent on the maximum permissible concentration for continuous consumption by the general population. Arrangements are being made to direct water from the external experimental area ditches at all times into Casey's Pond, thus preventing future releases off site via Kress Creek.

For the purpose of determining the concentration of accelerator-produced radioactivity in the soil and initiating a study of soil activation with time near a beam loss point, soil samples were taken from a hole drilled just outside the concrete wall of the Main Ring tunnel near the DO Abort System Target. All but a few samples were sealed and returned to the hole for future retrieval and leaching tests. The most radioactive

sample, a sand and gravel mixture from a depth equal to the elevation of the target, was counted independently by Jacob Sedlet at Argonne National Laboratory with the results for the principal accelerator-produced gamma-ray emitters plus tritium given in Table 6, corrected to the date of removal from the hole.

The sample was removed at mid-year (June 25) after about 10^{18} protons had been accelerated. 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[2], was 1.65×10^{-5} μCi/g. From the leaching tests the concentration of ^{22}Na activity in the solvent can be estimated.

Table 6

CONCENTRATIONS OF PRINCIPAL GAMMA-RAY EMITTING ISOTOPES AND TRITIUM IN SAND NEAR MAIN RING TARGET.

Isotope	Activity pCi/g	Activity pCi/ml
^{3}H		75
^{7}Be	143	
^{22}Na	16.5	
^{46}Sc	4.3	
^{51}Cr	19	
^{54}Mn	8.9	
^{56}Co	0.4	
^{59}Fe	1.6	
^{60}Co	0.3	

Assuming 20 per cent could be leached into a volume of water having ten times the weight as the sample (the same ratio as was used in the leaching tests)[2], the resulting concentration in the water would be 3.3 per cent of the maximum permissible for general consumption (Section 4). The other isotopes in the sample were smaller fractions of their maximum permissible concentrations than was ^{22}Na. The activity of ^{22}Na, which is probably the worst environmental offender in soil (tritium being the worst in water), was not detectable in the soil taken at an elevation in the hole equal to the top of the Main Ring tunnel.

3.4.D Non radioactive 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. The average use from both wells is 100,000 GPD. Samples of water from taps at 27 different locations throughout the Central Laboratory Area and its periphery have been analyzed throughout 1973 on a three sample per week basis for coliforms (bacterial contamination) by Aurora Clinical Laboratory, Aurora, Illinois. The coliform level was less than one per 100 ml. Samples of water from the domestic water supply in the village have been analyzed by the State of Illinois twice a month during 1973. The coliform tests showed zero per 50 ml. 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, was analyzed once a month in 1973 by Tenco-Hydro/Aerosciences, Inc. of Countryside, Illinois for biological oxygen demand (BOD), suspended solids and pH with results verifying the absence of pollution.

A new sewage plant in the Central Laboratory Area was completed in early 1973. The overflow water from this plant is carried off site by Indian Creek. Since the plant became operational, monthly samples from Indian Creek have been analyzed by Tenco Hydro/Aerosciences with good results. The dissolved oxygen level, BOD, pH, settled and suspended solids content of the Central Laboratory Sewage Plant and of the village oxidation pond effluents 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, Illinois. National Pollutant Discharge Elimination System (NPDES) permits have been obtained for both sewage plants.

3.4.3 Storage and Treatment of Waters Kept on Site

A new reservoir, Casey's Pond, has been filled and a pumping station built to increase our water reserves for times of inadequate rainfall. 130 million gallons of water were diverted from the Fox River in 1973.

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 duckweek and cattails last year.

3.5 Off-Site Monitoring

Although an extensive program of on-site monitoring is in progress, there has been very little off-site monitoring done to date. This is because, until recently, only very low concentrations of radionuclides were detected on site. In addition, the large area of the site itself (10.6 sq. mi.) allows us to make on site the same type of measurements which installations with smaller land areas must make off site. For our facility under current and anticipated future modes of operation measurements on site can be expected to yield positive results well in advance of off-site measurements.

Measurements are planned off site during 1974 of radiation levels along the paths the beams would travel if extended beyond the Laboratory boundaries. Also, a few off-site samples will be taken from the three creeks which leave the site. Some silt, soil, and vegetation samples may also be collected if this seems appropriate. During 1973, samples were collected from the Fox and DuPage Rivers (Fig. 1). These samples were evaporated, and the residues were analyzed for accelerator produced nuclides, but none were found.

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

4. References (Cont.)

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 the AEC Manual, Chapter 0524, Paragraph II.A. The value is 0.17 rem per year, per person. As a working procedure at the National Accelerator Laboratory a much lower limit has been adopted. According to R. R. Wilson's directive the radiation contributed to the off-site environment shall be less than ten millirem per year.

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Table 7

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).

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