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THE RADIOLOGICAL IMPACT OF HIGH-ENERGY ACCELERATORS ON THE ENVIRONMENT

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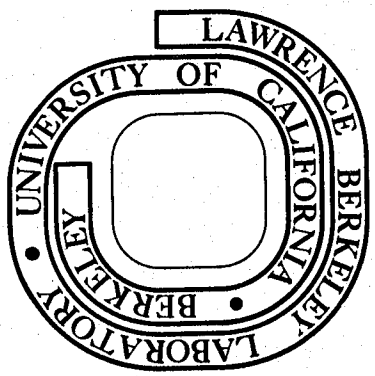

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The Radiological Impact of High-Energy Accelerators on the Environment

Lecture given at Kō Enerugi-Ken
(National Laboratory for High-Energy Physics)
Oho-Machi, Japan

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"Rumors have flown and spread. I may no more entice them back than I might a flock of birds."

Anonymous

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ABSTRACT

The potential radiological impact of high-energy, high-intensity accelerators in the environment is discussed. It is shown that there are three sources of radiation exposure to the general public resulting from the operation of high-energy accelerators. In order of importance these are (a) the prompt radiation field, produced when the accelerator is operating; (b) the release of radionuclides and aerosols into the atmosphere; and (c) the production of radionuclides in the groundwater system around the accelerator. Of these three sources, (a) is dominant and typically exceeds (b) by about an order of magnitude. To date, experience at many accelerator laboratories has shown that the quantity of accelerator-produced radionuclides released to nearby groundwater systems (c) is either extremely small or immeasurable.

The population dose equivalent resulting from the operation of several large high-energy facilities is compared.

1. INTRODUCTION

There are several reasons for discussing the radiological impact of high-energy accelerators on the environment, but I have chosen the subject primarily because it is topical.

In Japan and other countries around the world, there is increasing interest in man's impact on his environment. This interest tends to be converted to fear when the layman focuses attention on nuclear facilities.

It is often difficult for the layman to differentiate between a nuclear power station and a high-energy particle accelerator. Those of us working with particle accelerators are often subjected to the same irrational criticism that falls on the shoulders of those working in the nuclear power industry. We have a responsibility to understand the radiological phenomena associated with accelerators so that we may show our neighbors that we are in fact not subjecting them to any significant risks from ionizing radiation. Unfortunately as the poet says, "the rumors have flown and spread"[1]; although we cannot bring them back, perhaps we can hope that the truth will also fly and spread just as widely.

It is good health physics practice to study the possible radiological impact of any new accelerator. Such a study is greatly assisted by the considerable experience with a variety of large accelerators over the past 30 years. It is most important to place the radiological impact of accelerators in perspective, since it is usually minor when compared with changes in land use, inconvenience during the construction period, water and electrical consumption, and visual impact. This is made evident by Figure 1, which shows the 2-mile long Stanford linear accelerator. As a civil engineering structure the accelerator is comparable in magnitude with the freeway that crosses it. It is important when preparing environmental impact assessments not to overplay the production of ionizing radiation. A neighbor will in fact be much more interested in the other factors as mentioned above.[2]

In fact, although accelerators can be potent sources of ionizing radiation, they do not generally have a large radiological impact

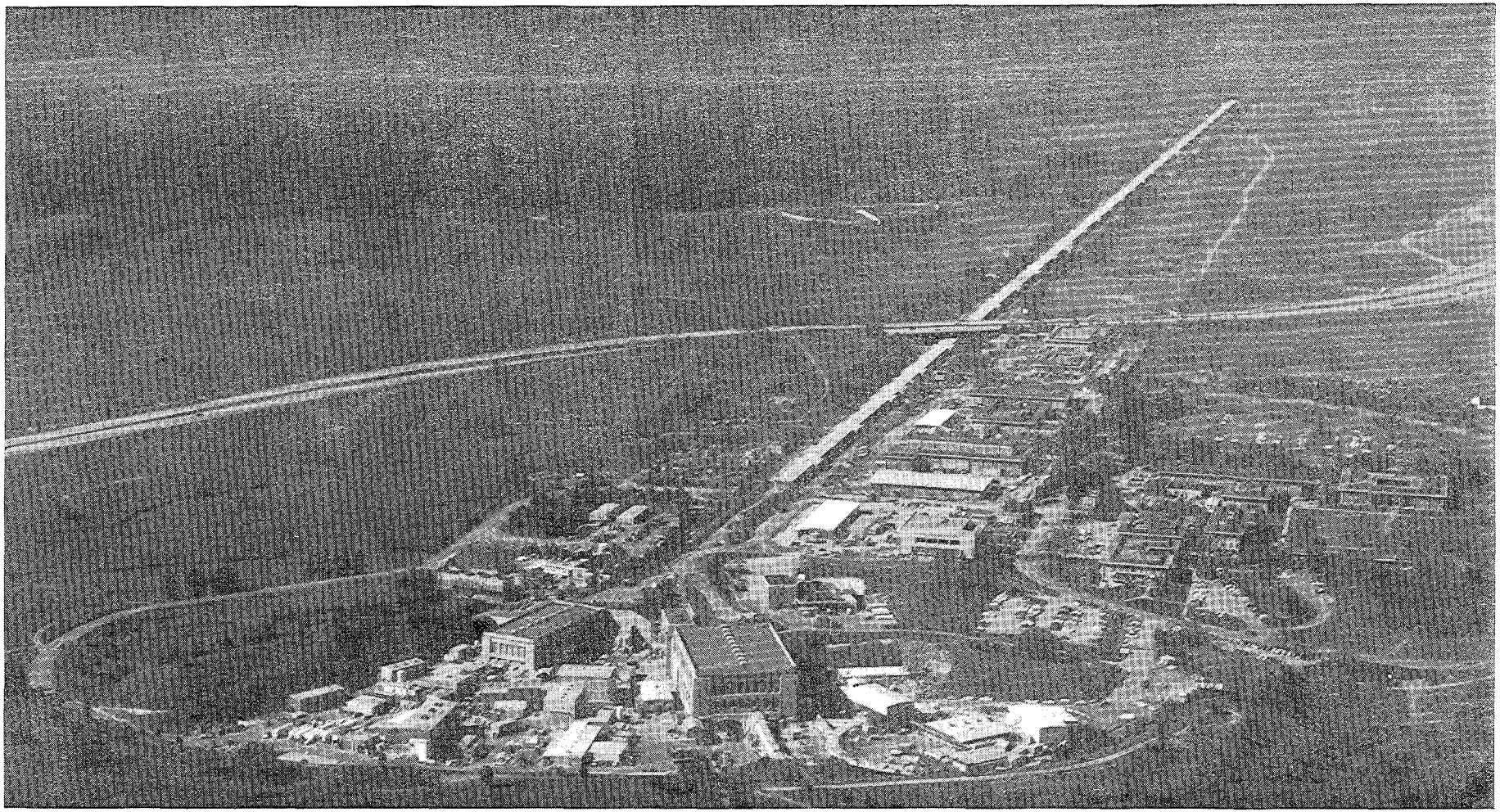

Figure 1. The 20 GeV electron linear accelerator at Stanford. The accelerator was built using cut and cover earth-moving techniques. The klystron gallery can be seen at the present grade level. The accelerator structure is buried 30 ft below the ground surface. (California State Highway #280 may be seen crossing the accelerator from left to right.) Photo courtesy of SLAC. XBB 788-9876

on the environment because they are well shielded. Indeed, it is often the case that the particle detectors used in high-energy physics experiments are much more sensitive to radiation than the scientists using them. In the past few years considerable information has been published describing the environmental surveillance programs of laboratories operating large accelerators, and these reports enable us to compare the radiological impact of different types of nuclear facilities on the environment.[3]

Study of the environmental monitoring reports of multidisciplinary research laboratories with many diverse potential sources of radiological impact on the environment, such as Argonne National Laboratory, Brookhaven National Laboratory, or the Los Alamos Scientific Laboratory, shows that accelerators have in general a relatively small impact. For example, during 1974 the population dose equivalent resulting from the operation of the Brookhaven alternating gradient synchrotron was only 0.7% of the total due to all operations[4] (see Table 1).

Table 1. Population dose equivalent, Brookhaven National Laboratory, 1974.

Airborne effluents	6.70 man rem (91.2%)
Liquid effluents	0.49 man rem (6.7%)
Forest γ-source	0.10 man rem (1.3%)
AGS skyshine	0.05 man rem (0.7%)
Total	7.34 man rem (100%)

At a high-energy laboratory such as KEK, however, accelerators are the only significant source of radiation exposure to the general public, and that is the topic that concerns us here.

In this talk the three principal radiological impacts due to high-energy accelerators will be discussed. In order of importance these are:

(a) The production of "prompt" radiation fields during accelerator operation.

(b) The production of radionuclides in the air in the accelerator vault and their subsequent release.

(c) The production of radionuclides in the soil and groundwater near the accelerator, with the possibility of migration into groundwater systems and consequent migration from the accelerator site.

Anticipating the conclusions of this talk, we shall see later the population exposure resulting from these impacts is in the following ratios:

Prompt radiation	:	radioactive gases	:	radionuclides in water
100	:	10	:	< 1

2. POPULATION DOSE EQUIVALENT - AN INDEX OF RADIOLOGICAL ENVIRONMENTAL IMPACT

The initial problem is how to determine an "index of harm" arising from the extremely small radiation exposures typically resulting from the operation of high-energy nuclear facilities. The International Commission on Radiological Protection (ICRP) has recently discussed some of the fundamental issues involved.[5]

In the absence of demonstrated harmful effects due to exposure to ionizing radiation at absorbed dose rates of a few rads per year, or less, regulatory bodies are forced to make some simplifying assumptions. These assumptions have been eloquently discussed in Evans, et al.[6]

"A linear non-threshold model was specifically chosen on a basis of mathematical simplicity and prudence to represent the upper limit of risk in low-dose domain, for somatic radiobiological effects which had been observed only in a higher-dose domain. The linear non-threshold model was not based on radiobiological data for somatic effects in the low-dose domain.

"As originally introduced, care was always taken in protection committee reports to point out that the true risk in the low-dose domain would be expected to lie between zero and the upper limit given by the linear non-threshold approximation."

Under the assumption that the probability of deleterious effects of exposure to ionizing radiation is linearly related to the dose equivalent, without threshold, an index of possible harm may be defined, termed the population dose equivalent.

The ICRP has defined the population dose equivalent resulting from operation of a nuclear facility, M, by the equation:[7]

$$M = \int_{H_{Min}}^{H_{Max}} HN(H)dH \ , \qquad (1)$$

where N(H) is the number of people receiving a dose equivalent H.

The models developed to calculate the population dose equivalent at high-energy nuclear laboratories are described in the following sections.

3. THE PROMPT-RADIATION ENVIRONMENT OF HIGH-ENERGY ACCELERATORS

3.1 Introduction

High-energy accelerators are potent radiation sources and need to be heavily shielded for radiation safety and to facilitate their experimental use. Whether the accelerator shielding consists of a concrete vault built around an accelerator constructed at ground level (e.g., the Bevatron) or whether the accelerator is buried below an earth shield (e.g., the KEK proton synchrotron), the prompt radiation field dominates the radiological impact of high-energy accelerators.

Extensive experience at both high-energy electron and proton accelerators has shown that, outside of the thick shielding, neutrons are usally the dominant source of dose equivalent.[8,9] The basic theoretical reasons for this observation have been given by DeStaebler[10] and later by Nelson and Jenkins.[9] Figure 2 shows the variation with time of photon and neutron dose equivalent rates at boundary of the Stanford Linear Accelerator Center (SLAC).[11] Operation of the SLAC 20 GeV electron linear accelerator is seen to result in a radiation environment dominated by neutrons. Measurements of the neutron spectra outside thick shielding at the SLAC 20 GeV electron linac and the Bevatron (a 6 GeV proton synchrotron) have demonstrated similarities between the environmental neutron spectra at both accelerators.[12]

3.2 Neutrons

A high-energy accelerator is a neutron source of considerable intensity. (For example, 10^9 neutrons per second leak from the roof shielding of the Bevatron when it accelerates 10^{12} proton sec^{-1}. A source strength of this magnitude will produce at a distance of 1 km from the accelerator a neutron flux density equal to the cosmic-ray-produced neutron background.[13]) Environmental surveillance programs at high-energy laboratories are usually therefore largely devoted to neutron monitoring. Thomas[14] and his co-workers have described the environmental radiological

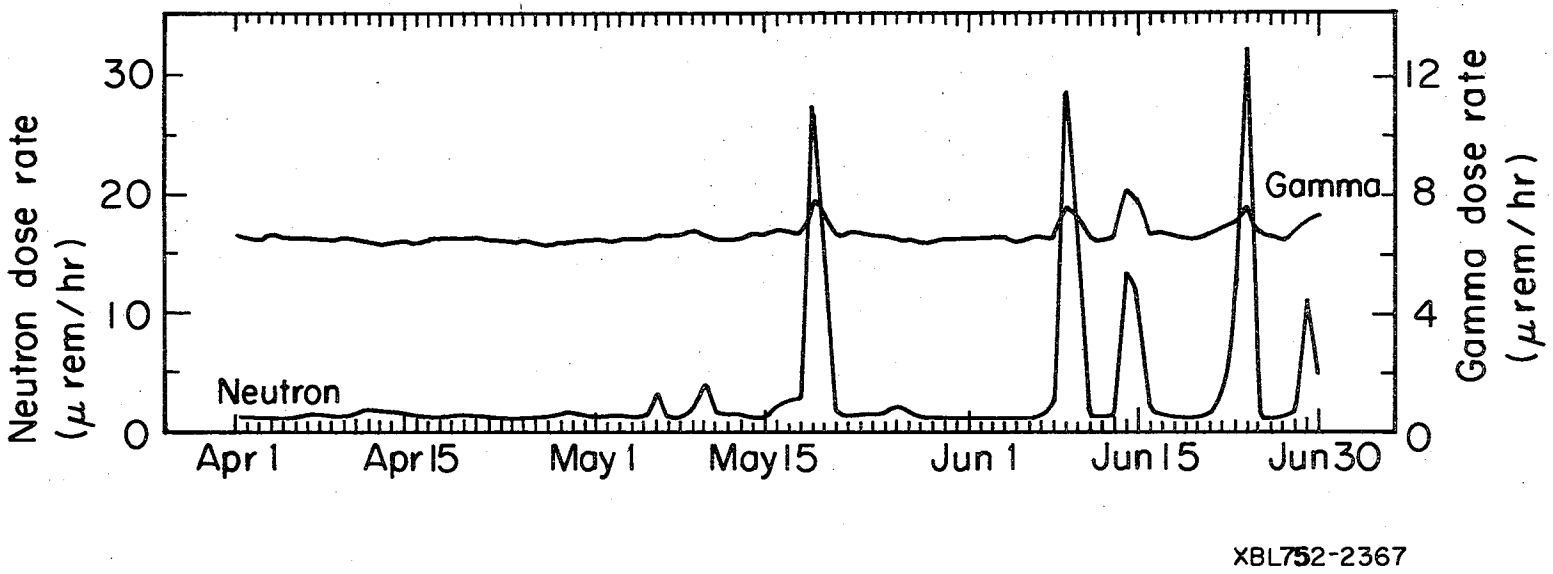

Figure 2. The variation in dose rate due to photons and neutrons observed at the boundary of the Stanford Linear Accelerator Center.

monitoring program of the Lawrence Berkeley Laboratory, which is typical of systems used at high-energy facilities.

At laboratories where more than one accelerator is in operation, it is sometimes necessary to identify the major radiation sources. Changes in mode of operation or additional shielding may the be utilized in the most effective manner if environmental radiation levels must be reduced. Bonifas et al.[15] have described a convenient technique for measuring both the gamma-ray and neutron radiation levels around accelerators using thermoluminescent dosimeters. This method of measurement has the convenience of being relatively simple and inexpensive. Figure 3 shows contours of equal neutron dose equivalent around the CERN laboratory site obtained by this technique. Three principal sources of radiation may be seen— the proton synchrotron (PS), synchrocyclotron (SC) and the intersecting storage rings (ISR). Radiation levels of 10 mrem/yr due to neutrons may be seen to exist at about 500 m from the proton synchrotron. The photon dose equivalent is roughly a factor of 10 lower than that due to neutrons.

As we have seen, accelerators can produce detectable neutron flux densities at distances of several hundred meters. In order to be able to estimate population exposures, it is necessary to understand the transport of accelerator-produced neutrons through the air. As a result of shielding measurements,[16] "skyshine" studies,[17] and some crude measurements of the energy spectrum of neutrons leaking from the accelerator shield,[18] we now have a fair understanding of this matter. The character of the shield-leakage neutron spectrum is controlled by the interaction of neutrons of energy greater than about 100 MeV. The nature of the equilibrium achieved between these high-energy neutrons and their interaction products is determined by the nuclear properties of the shield. Typically, neutrons contribute more than 90% of the total dose equivalent and 50% of the neutron dose equivalent is contributed by neutrons with energies between 0.1 and 20 MeV. For certain leakage spectra, however, neutrons in the keV energy

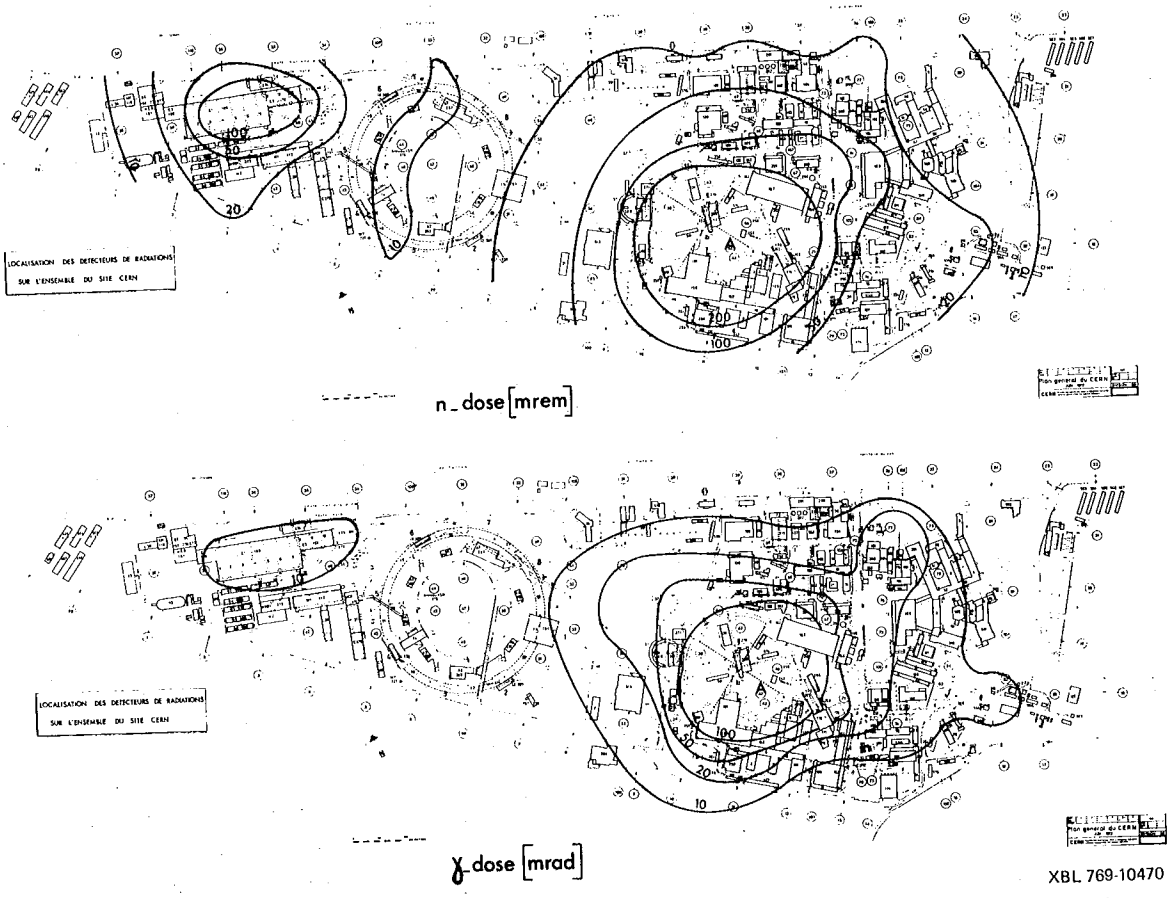

Figure 3. Contours of equal neutron and photon dose equivalent at the CERN Laboratory site for the year 1974. The three principal radiation sources are clearly identified--the synchrocyclotron (SC) to the left, the intersecting storage rings (ISR) in the middle and the proton synchrotron (PS) to the right.

region (for example, steel shields) or neutrons above 20 MeV (wet earth shields) may be more important than this general rule implies.[18] Changes in the leakage spectrum at the air interface are important in the transport of neutrons to large distances. When neutrons leave the shield small changes in the spectrum—most noticeably in the neutron resonance region—are initiated. An equilibrium determined by the nuclear properties of air will be re-established after passage through two to three interaction mean-free paths, corresponding to several hundred meters in air.

Rindi and Thomas[17] have reviewed the published measurements of neutrons at large distances from high-energy accelerators. Figure 4 shows measurements of neutron flux density vs distance made at seven different accelerators. Rindi and Thomas were able to conclude from these and other data that despite difficulties in interpretation, the available experimental data are consistent with our present understanding of electromagnetic and hadron cascade phenomena and that:

(a) The radiation intensity decreases at least as fast as the inverse of the square of the distance from the source.
(b) At large distances from accelerators, neutrons are the dominant component of the radiation field.
(c) For well-shielded accelerators in the GeV region, the neutron spectrum emerging from the shield is in equilibrium. At lower energies or at accelerators with inadequate overhead shielding, hardening of the spectrum with distance is observed.
(d) The empirical relation

$$\phi(r) \approx \frac{aQe^{-r/\lambda}}{4\pi r^2}$$

is a simple but adequate expression for the skyshine intensity around most accelerators. Values of λ reported in the literature discussed by Rindi and Thomas[17] vary between 267 m and 990 m. At large distances (several thousand meters—from our understanding

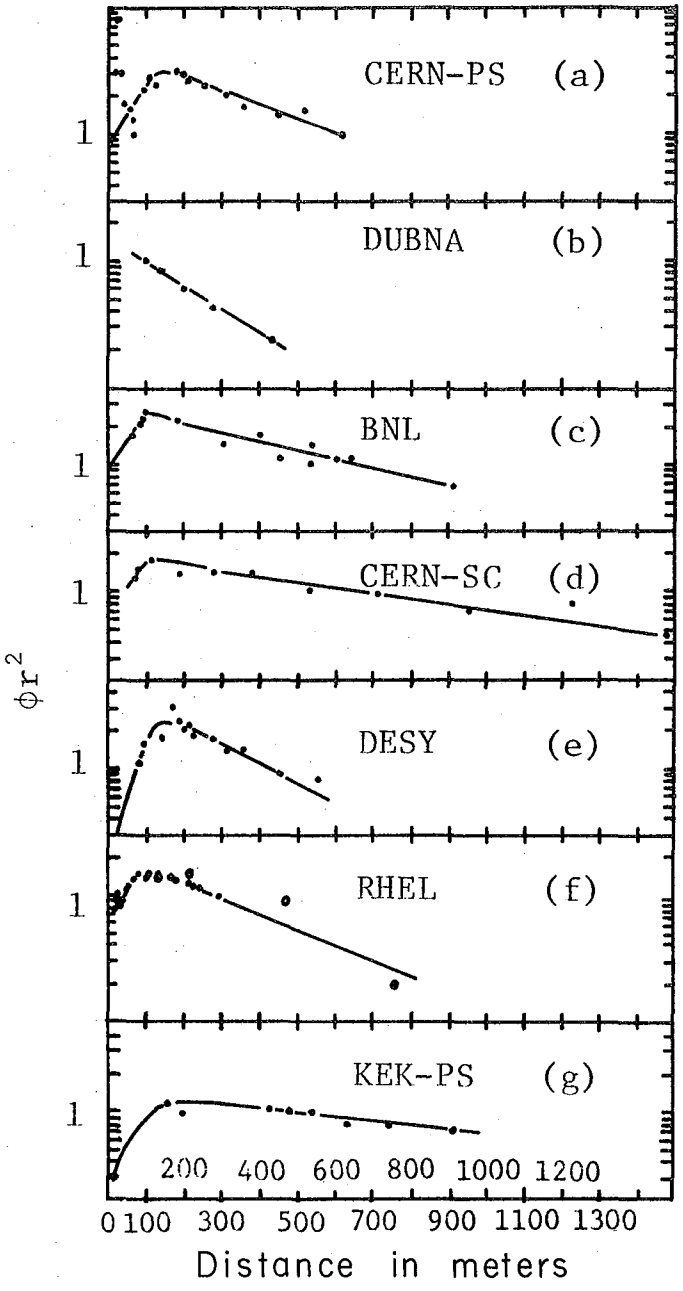

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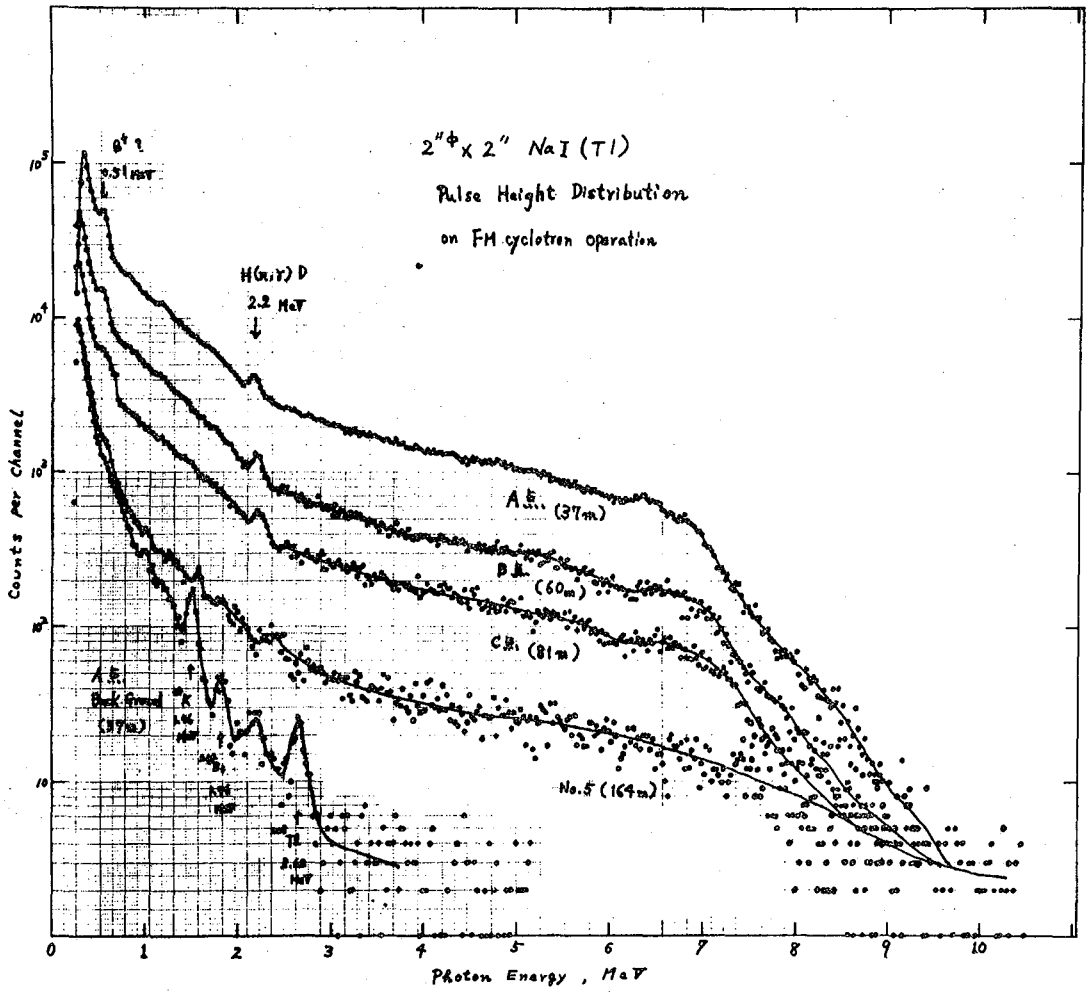
Figure 4. Measurements performed around different accelerators. On the abscissa is the distance from the accelerator in meters, on the ordinate is the product of the measured neutron flux density by the square of the distance. In these coordinates a $1/r^2$ variation shows up as a horizontal line. a) Measurements of fast neutron flux density performed at the CERN 28-GeV Proton Synchrocyclotron.[19] b) Measurements of fast neutron flux density performed at the Dubna 10-GeV Proton Synchrophasotron.[20] c) Measurements of dose-equivalent rate performed at the Brookhaven 30-GeV Proton AGS.[21] d) Measurements of fast neutron flux density performed at the CERN 600-GeV Proton Synchrocyclotron.[22] e) Fast neutron flux density measurements performed at the DESY 7.5 GeV Electron Synchrotron.[23] f) Fast neutron flux density measurements performed at the Rutherford Laboratory Proton Linear Accelerator; the solid dots indicate the measurements taken for a p beam of 30 MeV,[24] and the open dots for a p beam of 50 MeV.[25] g) Measurements made at the 12 GeV Proton Synchrotron at KEK.[26]

of high-energy hadron cascades), we would expect λ to approach the value of 100 g cm^{-2}. (Recently Katoh and his colleagues[26] have reported a value of λ of 1300 m determined from data taken around the KEK proton synchrotron.) In addition to this measurement, a collaboration among the Universities of Kyoto, Tokohu and Tokyo, and the Japan Atomic Energy Research Institute and KEK, is investigating skyshine phenomena from a variety of neutron sources. This collaboration should provide a valuable addition to our knowledge of neutron transport through the atmosphere.[26]

3.3 Photons

Also in Japan, Nakamura and his colleagues have reported some measurements of skyshine neutrons and photons produced by 52 MeV protons of the cyclotron of the Institute of Nuclear Study, Tokyo.[27] The measurements of skyshine photons are particularly interesting because they are the first such measurement reported in the literature. The authors show that skyshine photons are transported in the atmosphere with approximately the same dependence on distance as both thermal and fast neutrons. This observation is consistent with our understanding of nuclear cascade processes because high-energy neutrons eventually control the production of photons in the atmosphere. These measurements support the hypothesis that the relative importance of photons will not increase with distance from high-energy accelerators. In fact at accelerators where protons are initially dominant, the fraction of total dose equivalent contributed by neutrons will increase because photons will be absorbed more rapidly in the atmosphere. Ultimately, an equilibrium will be achieved with high-energy neutrons controlling the photon production. Measurements of the energy spectrum of scattered photons show a prominent 2.2 MeV peak due to the capture of thermal neutrons in hydrogen. At higher energies the photon spectrum falls monotonically with a "knee" at about 7 MeV (Figure 5). More measurements of photon spectra around high-energy particle accelerators are needed to understand transport phenomena better.



Spectra of skyshine photons.

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Figure 5. The pulse height distribution obtained with a NaI detector at distances from a 52 MeV FM cyclotron.[27]

Photons are, however, of little importance in contributing to the population dose equivalent at well-shielded accelerators.

3.4 Muons

Under certain shielding conditions muons may be observed as a major component of the stray radiation field at the Brookhaven AGS[28] or the CERN PS.[29] Baarli and Höfert[29] have described the use of a counter telescope to locate the source of muons and leakage in the shielding at the CERN PS.

At the 500 GeV proton synchrotron of the Fermi National Accelerator Laboratory, muons are the dominant component of the radiation level at the site boundary.[30] This is in part due to the large distances from the accelerator to the site boundary. Neutrons produced at the accelerator are much reduced in intensity by inverse square law and air attenuation at the laboratory perimeter. Muons, on the other hand, are produced in a highly collimated beam directed towards the site boundary. Because of their weak interaction with matter they survive thick shielding and emerge into the air, still well collimated. At the site boundary a well-defined "beam" $\approx$50 m wide can be identified.[30] The maximum dose equivalent at the site boundary was 2 mrem during 1974. Radiation levels outside this "beam" were at least a factor of 10 lower.

3.5 Population Dose Equivalent from Prompt Radiation

The calculation of population dose equivalent due to prompt radiation is a complex problem and, indeed, the form of the variation of dose equivalent with distance from the accelerator is not yet precisely known.[11,17] Thus there is no generally accepted method of calculating the population dose equivalent. Stephens et al.[31,32] have suggested a simple model which assumes:

(a) A population distribution around the accelerator which is constant with time, and

(b) A variation of dose equivalent H(r), with distance, r, given by:

$$H(r) = \frac{ae^{-r/\lambda}}{r^2},$$

where λ is the effective attenuation length of the accelerator-produced neutrons and a is a constant.

Using this model Stephens et al. show that the annual population dose equivalent due to prompt radiation, M, is given by:

$$M = \frac{2\pi r_o^2 H_o e^{r_o/\lambda}}{S_1 S_2} \sum_{i=1}^{i=n} \frac{N_i}{\pi(r_i^2 - r_{i-1}^2)} \int_{r_i-1}^{r_i} \frac{e^{-r/\lambda}}{r} \, dr \, , \quad (2)$$

where H_o is the annual dose equivalent at the laboratory boundary,

N_i is the average number of people who may be considered permanently resident between distance r_{i-1} and r_i from the accelerator,

S_1, S_2 are shielding factors for surrounding hills and buildings,

r_o, r_n are the closest and furthest distances which members of the general public approach the accelerator.

Equation (2) may be numerically evaluated.

Typically, the population dose equivalent converges towards its ultimate value within a few kilometers from the accelerator. Thus, for example, at the Lawrence Berkeley Laboratory (LBL) the population dose equivalent reaches its ultimate value at about 5 km from the Laboratory (Figure 6). Thus, although it is conventional to quote the 80 km (50 mile) population dose equivalent, the value of r_n in equation (2) will, in general, be much smaller.

With the simplifying assumption of a uniform population density distribution, σ, around the accelerator, Stephens et al.[31] show that over a restricted range M is given by

$$M = k \sigma H_o r_o^n \lambda^m \, , \quad (3)$$

where k, n and m are constants.

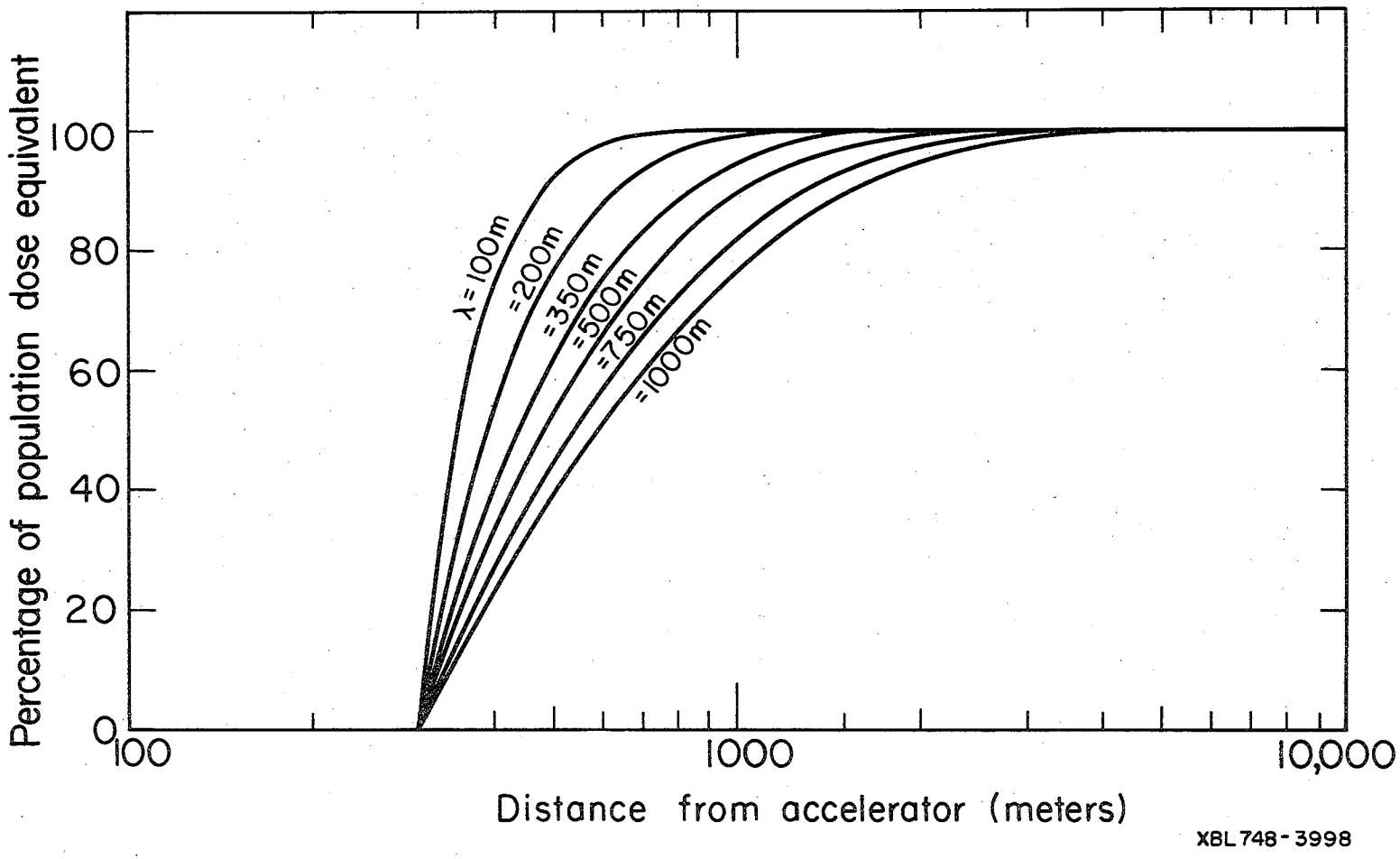

Figure 6. The convergence of the population dose equivalent as a function of distance from the accelerator for different neutron attenuation lengths in air.[31]

Values of m are between 0.6 and 0.8, while n takes values between 1.1 and 1.6. It is usually sufficiently accurate to write

$$M \propto \sigma H_0 r_0^{4/3} \lambda^{2/3} . \qquad (4)$$

Furthermore, since

$$H_0 \propto \frac{1}{r^2}$$

for a fixed radiation source strength we have:

$$M \propto \sigma \left(\frac{\lambda}{r_0}\right)^{2/3} . \qquad (5)$$

For accelerator installations σ and r_0 are fixed and thus the accuracy of the estimate of M is determined by the accuracy of our knowledge of λ. As we have seen λ may take values in the range from about 225 m (corresponding to a fission spectrum) to 850 m (corresponding to neutrons with energy greater than 100 MeV). Thus, if a conservative value of λ is assumed (850 m) the population dose equivalent could be overestimated by as much as a factor of 2.5 if the leakage spectrum from the accelerator shield were rich in low-energy neutrons. More precise experimental studies of the transport of high-energy neutrons, particularly at distances beyond 2500 m from accelerators, and neutron spectrum measurements will be helpful in improving estimates of population dose equivalent. Table 2 summarizes values of population dose equivalent estimated for several high-energy accelerators.

Table 2. Summary of population dose equivalent estimates for several high-energy accelerator laboratories.

Laboratory	M/H_0 Man rem/ fence post rem	Comments
Lawrence Berkeley Laboratory[32]	1023	≈170,000 people living within 5 km from the laboratory. Average population density 2-3 x 10^3 persons/km^2.
Fermi National Accelerator[33]	1000	Dose due to collimated muon beam 50 m wide at site boundary. Approximately 100,000 people in irradiated zone.
Stanford Linear Accelerator Center[34,35]	460	Population dose equivalent calculated out to 1 km from the laboratory. 2,000 people living within 2 km from the laboratory.
KEK[36]	210	Population density 102 persons/km^2 ⩽ 1.0 km, 229 persons/km^2, 1.0 ~ 1.5 km, 340 persons/km^2 > 1.5 km

Table 2. Summary of population dose equivalent estimates for several high-energy accelerator laboratories. (Continued)

Laboratory	M/H_0 Man rem/ fence post rem	Comments
CERN European Organization for Nuclear Research [37,38]	160	30,000 people living within 4 km from the laboratory. Population density 35 persons/ $km^2 \leq 1$ km, 640 persons/km^2 ≥ 1 km from laboratory
Brookhaven National Laboratory[39]	54	5.2×10^6 people living within 80 km from laboratory. Average population density of ≈ 260 persons/km^2.

4. RADIOACTIVITY PRODUCED IN THE ATMOSPHERE BY HIGH-ENERGY ACCELERATORS

4.1 Introduction

The second most important source of exposure of the general population to accelerator-produced radiation is from radioactivity in the air.

Typically, however, the magnitude of this exposure is many times smaller than that from "prompt" radiation. The magnitude of the exposure is in fact often so small that no estimates of population dose equivalent have been made at some accelerator laboratories.

4.2 Radionuclides Produced in Air by Accelerator Operation

The main source of radioactivity in air is the interaction of primary and secondary particles directly with constituent target nuclei of the air. A second source of airborne radioactivity is dust, formed by natural erosion and wear, or by maintenance on radioactive accelerator components. The third and final source is due to the emission of gaseous radioactivity from liquids irradiated in the accelerator-produced radiation environment.

4.2 (a). Radionuclides Produced Directly in Air. During accelerator operation, radioactive nuclides are produced by the interaction of primary and secondary particles with the air in the accelerator halls. Spallation reactions in solid machine parts may also contribute to the formation of radioactive gases. If the air is confined in the accelerator hall there will be no release of radioactivity during the operation of the machine. In such a case, however, a rather high concentration of radioactive gases may accumulate because the specific activity of several of the possible radioisotopes will reach saturation. Most of the present high-energy or high-intensity accelerators provide air circulation, mainly for cooling reasons. The residence time of air inside the hall and consequently the irradiation time of the air, is usually less than 30 minutes so that the production of high concentrations of radioactive gases with a long half-life is minimal.

Table 3 summarizes the target nuclei most abundant in air. A summary of the radionuclides with half-life greater than 1 sec that may be produced from these by thermal neutron capture (γ,n), and spallation reactions with these target nuclei is given in Table 4.

With the exception of ^{3}H and ^{7}Be, the half-lives of most radionuclides are short, so that there will be some decay in the short time before they reach inhabited regions around the accelerator.

From the environmental point of view, the only significant radionuclides are ^{3}H, ^{7}Be, and perhaps ^{11}C, ^{13}N, and ^{15}O. However, the ^{3}H half-life is so long that its rate of production will be rather small.

Among the first reported measurements of radioactive gases produced at accelerators are those of Russel and Ryan[41] in 1965 who detected ^{15}O and ^{13}N in the air around a 70 MeV electron linac produced by (γ,n) reactions with ^{16}O and ^{14}N in the air. By calculating the maximum possible concentration for these radionuclides (which were not at that time available in the literature) and by estimating the diffusion of these radionuclides from the source, these authors concluded the concentrations around the accelerator to be much lower than MPC. Similar conclusions were reached by George et al.[42]

Over the years considerable effort has gone into identifying airborne radionuclides at accelerators. Table 5 summarizes some of the published data.

4.2 (b) The Magnitude of Radionuclide Production in Air. Patterson and Thomas[49] have summarized estimates of the total specific activity, S, of an enclosed volume of radioactive air near an accelerator and show it to be given by:

$$S = C \sum_i \left[\sum_j \Phi_\gamma N_j \bar{\sigma}_{ij_\gamma} + \sum_j \Phi_{th} N_j \bar{\sigma}_{ij_{th}} + \sum_j \Phi_{HE} N_j \bar{\sigma}_{ij_{HE}} \times (1-e^{-\lambda_i T}) e^{-\lambda_i t} \right], \quad (6)$$

where Φ_γ, Φ_{th}, and Φ_{HE} are the average photon, thermal neutron, and high-energy particle flux densities,

and $\bar{\sigma}_{ij_\gamma}$, $\bar{\sigma}_{ij_{th}}$ and $\bar{\sigma}_{ij_{HE}}$ are the corresponding average cross sections.

Equation (6) gives results which are in agreement with observed values within a factor of two or better. Figure 7 compares calculated and measured values of activity around the accelerators at CERN.[44]

Quite accurate estimates of the concentration of radioactivity in air may be made if the influence of air changes is considered. This may be done by substituting an effective decay constant λ'_i for λ_i in equation (6). λ'_i is given by:

$$\lambda'_i = \lambda_i + D/V \quad , \quad (7)$$

where D is the ventilation rate and V is the volume of the accelerator room.

Table 6 gives an example of the good agreement obtained by Peetermans and Baarli[50] between calculated and measured values of specific activity for four radionuclides, at the CERN 600 MeV synchrotron.

4.2 (c) Radionuclides Produced in Dust. The most direct method of determining if any potentially serious internal contamination problem exists is to determine the body burdens of accelerator-produced radionuclides in accelerator workers and to study possible contamination pathways.

Table 3. Most abudant isotopes in the atmosphere.

Isotope	Percentage by volume in the atmosphere
^{14}N	78.1
^{16}O	21.2
^{40}Ar	0.46
^{15}N	0.28
^{18}O	0.04

Table 4. Radionuclides with half-life > 1 min which can be produced in air at accelerators (after Rindi[40]).

Radionuclide	Half-life	Emission	Parent element	Production reaction	Cross section (mb)
$^{3}_{1}H$	12.2 yr	β^-	N	spallation	30
			O	spallation	30
$^{7}_{4}Be$	53 days	γ,EC	N	spallation	10
			O	spallation	5
			Ar	spallation	0.6
$^{11}_{6}C$	20.5 min	β^+	N	spallation	10
			O	spallation	5
			Ar	spallation	0.7
$^{13}_{7}N$	10 min	β^+	N	spallation	10
			N	(γ,n)	10
			O	spallation	9
			Ar	spallation	0.8
$^{14}_{8}O$	74 sec	β^+,γ	O	spallation	1
			Ar	spallation	0.06
$^{15}_{8}O$	2.1 min	β^+	O	spallation	40
			O	(γ,n)	10
			Ar	spallation	
$^{18}_{9}F$	1.85 h	β^+,EC	Ar	spallation	6

Table 4. Radionuclides with half-life > 1 min which can be produced in air at accelerators (after Rindi[40]). (Cont.)

Radionuclide	Half-life	Emission	Parent element	Production reaction	Cross section (mb)
$^{24}_{10}Ne$	3.4 min	β^-,γ	Ar	spallation	0.12
$^{22}_{11}Na$	2.6 yr	β^+,γ	Ar	spallation	10
$^{24}_{11}Na$	15 h	β^-	Ar	spallation	7
$^{27}_{12}Mg$	9.5 min	β^-,γ	Ar	spallation	2.5
$^{28}_{12}Mg$	21.3 h	β^-,γ	Ar	spallation	0.4
$^{28}_{13}Al$	2.3 h	β^-,γ	Ar	spallation	13
$^{29}_{13}Al$	6.6 min	β^-,γ	Ar	spallation	4
$^{31}_{14}Si$	2.6 h	β^-,γ	Ar	spallation	6
$^{30}_{15}P$	2.5 min	β^+,γ	Ar	spallation	4.4

Table 4. Radionuclides with half-life > 1 min which can be produced in air at accelerators (after Rindi[40]). (Cont.)

Radionuclide	Half-life	Emission	Parent element	Production reaction	Cross section (mb)
$^{32}_{25}P$	14.3 days	β^-	Ar	spallation	25
$^{33}_{15}P$	25 days	β^-	Ar	spallation	9
$^{35}_{16}S$	87 days	β^-	Ar	spallation	23
$^{34}_{17}Cl$	32.4 min	β^-,γ	Ar	spallation	
$^{38}_{17}Cl$	37.3 min	β^-,γ	Ar	(γ,pn)	4
$^{39}_{17}Cl$	55 min	β^-,γ	Ar	(γ,p)	7
$^{41}_{18}Ar$	1.8 h	β^-,γ	Ar	(n,γ)	610

Table 5. Radionuclides identified in the air around several accelerators.

Laboratory	Accelerator	Radionuclides identified	(Ref)
RPI	50-MeV electron linac	^{15}O, ^{13}N	(41)
Saclay	330- to 560-MeV, electron linac	^{13}N, ^{15}O, ^{11}C, ^{41}Ar, ^{38}Cl, ^{7}Be	(43)
CERN	600-MeV proton synchrotron	^{11}C, ^{13}N, ^{41}Ar	(44)
PPA	3-GeV proton synchrotron	^{14}O, ^{15}O, ^{13}N, ^{11}C	(45)
RHEL	7-GeV proton synchrotron	^{16}N, ^{15}O, ^{13}N, ^{11}C	(46)
CERN	25-GeV proton synchrotron	^{13}N, ^{11}C, ^{41}Ar	(47)
BNL	30-GeV proton synchrotron	^{13}N, ^{11}C, ^{41}Ar	(48)

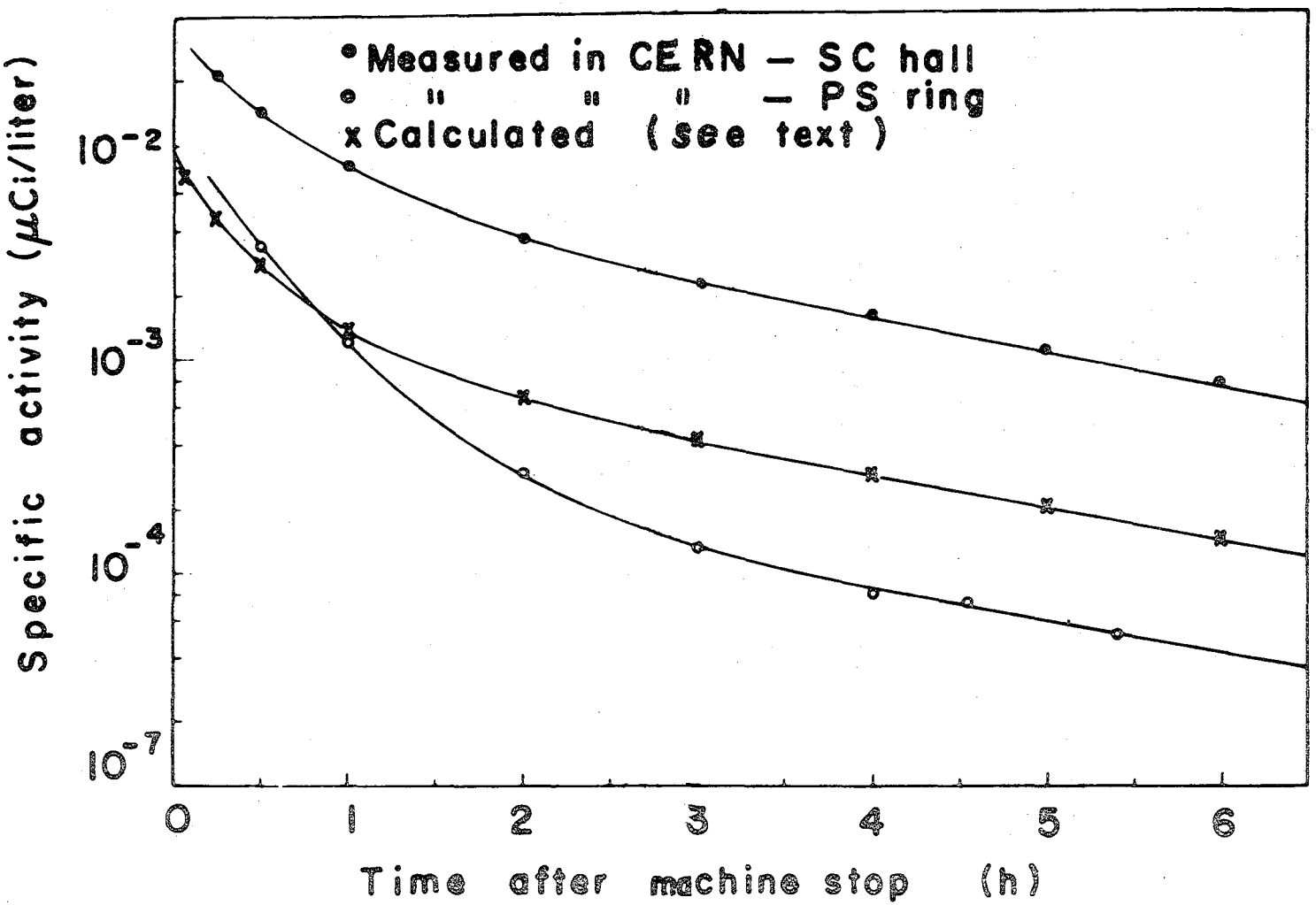

Figure 7. Typical decay curves of radioactive air as measured at the two CERN accelerators and computed decay calculated assuming that the contribution of radioisotopes is as follows: ^{41}Ar (14%), ^{11}C (31%), ^{13}N (47%), ^{15}O(8%), ^{14}O (44%). XBL 719-4361

Table 6. Measured and calculated specific activity in air of some long-lived isotopes leaving the CERN 600 MeV synchrocyclotron ISOLDE area[50] (in pCi/cm^3).

Isotope	Half-life	Measured	Calculated
^{3}H	12.26 yr	-	$1.6\ 10^{-5}$
^{7}Be	53.6 days	$2.7\ 10^{-4}$	$3.2\ 10^{-4}$
^{24}Na	15 days	$3.6\ 10^{-5}$	$4.8\ 10^{-5}$
^{32}P	14.5 days	$6.0\ 10^{-6}$	$8.4\ 10^{-6}$
^{33}P	25 days	$2.0\ 10^{-6}$	$4.5\ 10^{-6}$
^{41}Ar	1.83 h	-	$2.5\ 10^{-2}$

Experience at several large accelerators tends to show that the potential exposure to radioactive dust for maintenance crews working in the accelerator vault is negligible. This problem has been discussed at some length by Thomas and Rindi[51] and so will not be discussed here.

It is of interest to note that Busick and Warren[52] have concluded that chemical toxicity and external radiation exposure are the factors that limit the machining of radioactive accelerator components rather than the inhalation of radioactive dust.

4.2 (d) Gaseous Radionuclides Released from Water. In certain circumstances radionuclides produced in liquids irradiated at accelerators may be released to the environment. For example, tritium produced by spallation reactions in magnet cooling water may be released by the evaporation of water spills and losses during magnet maintenance or replacement.

Warren et al.[53] have studied the production of CO_2 which acts as a carrier for ^{11}C and ^{15}O produced in water beam dumps at the Stanford Linear Accelerator Center.

4.3 Environmental Impact

In 1972, Prantl and Baarli[54] reported finding some small amounts of ^{7}Be in a creek at the CERN accelerators. However, due to the lack of systematic background measurements of ^{7}Be produced by cosmic rays, no final conclusions as to its origin could be drawn at that time. The same year, Rindi[40] made a detailed theoretical study of the production of radioisotopes in the air of a 300 GeV proton accelerator and of the dispersion of these isotopes in the environment. He concluded that, under some adverse atmospheric conditions coupled with some particular machine use, concentration of ^{7}Be and perhaps also of ^{3}H of the order of the MPC for the population at large could be detected at some 5 km from the air rejection shaft of the accelerator. In 1974 Peetermans and Baarli[50] reported some new calculations and measurements performed at the CERN 600 MeV proton synchrocyclotron to estimate

the concentration of radioactive gases in surrounding region when the intensity of that accelerator was increased. ^{7}Be, ^{24}Na, ^{28}Mg, ^{31}Si, ^{32}P, and ^{33}P were identified in the ventilation air around an extracted beam, by flow ionization chambers and γ-spectroscopy on filters. The measured concentrations were in good agreement with those calculated (Table 6). These authors concluded that the annual contribution to the dose equivalent at the CERN boundaries due to radioactive isotopes in air would be about 6 mrem, less than 10% of the dose equivalent due to stray radiation, and that the eventual ^{7}Be surface pollution at distances up to 1 km from the boundaries would be barely detectable. In fact measurements of the ^{7}Be content of rainwater around CERN show much larger fluctuations than at control sites (see Figure 8). Air-sampler filters subjected to γ-spectrometry show that, while the activity of γ-emitting nuclides from nuclear weapons fallout shows similar trends at both laboratories, fluctuations in ^{7}Be activity in air at CERN are much larger than those found at Le Vesinet.* These data tend to suggest that accelerator operation at CERN produces detectable quantities of ^{7}Be close to the high-energy accelerators, corresponding to periods of high-intensity operation. At the Lawrence Berkeley Laboratory where several proton and heavy-ion accelerators are operated, a search for possible contamination from ^{7}Be was made by measuring the radioactivity of bracken ferns collected around the center and at different locations several miles away. These studies show that the ^{7}Be contamination in air due to the presence of the accelerator at the Berkeley laboratory, if any, is within the variation of the background produced by cosmic rays.[55]

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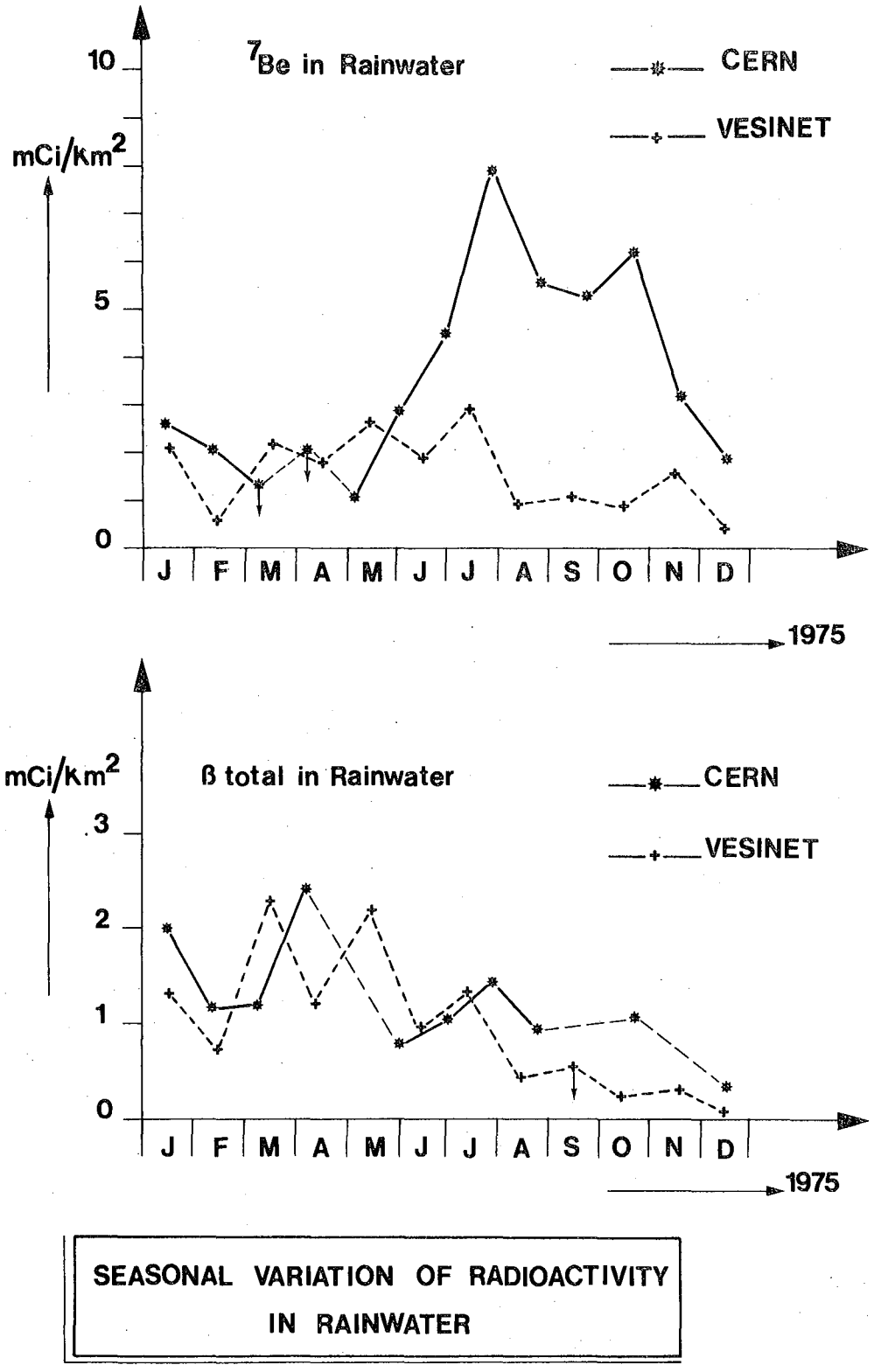
*Le Vesinet is situated in the suburbs of Paris—more than 400 km from Geneva. The value of this station as a control for Geneva CERN is therefore somewhat suspect. However, the gross β-activity and γ-activity from known nuclear weapons fallout nuclides shows similar seasonal trends at many West-European stations.



SEASONAL VARIATION OF RADIOACTIVITY IN RAINWATER

XBL 769-10468

Figure 8. (a) Monthly variation of radioactivity in rainwater at CERN and (b) at Le Vesinet (Paris).[37]

The measurement of the concentration of ^{7}Be in air or in dust around accelerators may prove to be a valuable indicator of radioactive contamination. Further studies are needed, including measurements of the natural background fluctuation of ^{7}Be in rainwater.

4.4 Population Dose Equivalent from Radioactivity in the Air

As we have seen, four short-lived radionuclides — ^{10}C, ^{11}C, ^{12}N and ^{15}O — typically account for the major portion of the total activity released. Of the longer lived radionuclides only ^{41}Ar, ^{7}Be, and ^{3}H are released in significant quantities.

The calculation of population exposure resulting from the release of these gaseous radionuclides is normally performed according to the following steps:

1) Measurement or calculation of the concentration of the different radioisotopes released at the ventilation stacks of the accelerators.

2) Calculation of the transport of the gases outside the boundaries of the Laboratory. This is generally done by applying the Sutton equation for some average meteorological conditions.

3) Conversion of the concentrations of radioactive gases to dose equivalent to an exposed individual by using the MPC values.

4) Summation of the total number of exposed individuals and their dose equivalent to give the total population dose.

Steps 2, 3 and 4 can be summarized in a general equation. Thomas[14] has described the use of the following expression to calculate the population dose equivalent resulting from the release of a given radionuclide that applies for the Lawrence Berkeley Laboratory, but can be easily generalized.

$$M = \sum_{i=1}^{n} \left[(2/\pi)^{1/2} \, (n/2\pi) \, \frac{f_i Q R}{\bar{\mu}} \int_{r_o}^{\infty} \frac{N_i(r)}{r\sigma_z(r)} \, dr \right] , \quad (8)$$

where n is the number of sectors into which the region surrounding the source is divided (this division is decided by the wind direction (frequencies);

f_i is the fraction of time in which the wind blows toward a given sector;

Q is the total quantity of radionuclide released (in Ci);

R is the dose equivalent conversion factor for the given radionuclide (man-rem per curie second per cubic meter);

$\bar{u}$ is the average wind velocity (in ms^{-1});

r is the distance from the rejection point;

$N_i(r)dr$ is the total number of people in the i^{th} sector in the region between r and $r + dr$;

r_o is the distance of closest approach to the Laboratory; and

$\sigma_z(r)$ is the vertical dispersion coefficient (in m).

The integral can be simplified into a summation by subdividing each sector into m regions in which the population density may be assumed to be constant.

The number of residents at a distance r from the rejection point in the j^{th} region of the i^{th} sector is then given by

$$ {}_jN_i(r) = \frac{2\pi r}{n} \sigma_{ij} \, dr \quad , \qquad (9) $$

where σ_{ij} is the population density.

The expression between square brackets in equation 8 can be expressed by

$$ {}_jM_i = (2/\pi)^{1/2} \frac{f_i Q R \sigma_{ij}}{\bar{u}} \int_{r_{j-1}}^{r_j} \frac{dr}{\sigma_z(r)} \qquad (10) $$

and the population dose equivalent is then expressed by

$$ M = \sum_{i=1}^{n} \sum_{j=1}^{m} {}_jM_i . \qquad (11) $$

From the meterological statistics for the region under study, $\sigma_z(r)$ can be given an analytical expression as a function of r such that the integral in (10) can be evaluated.

It is customary at accelerator centers to report yearly on environmental monitoring results. Several centers perform continuous measurements of gaseous radioactivity in air, in addition to radioactivity deposited on filters, and also periodically investigate the radioactivity on ground, water and plants around the boundaries of the center.

At the Fermi Laboratory in Batavia, where a 300 GeV proton synchrotron is located, the annual off-site exposure from airborne release was estimated to be about 9.1 manrem, 1/10 of the exposure from external radiation during 1975.[56]

At the Lawrence Berkeley Laboratory, where several proton and heavy-ion accelerators are located, the air monitoring systems detect releases of radioactive gas only from chemistry laboratories and not from accelerator operation. Thus, for example, during 1975 the estimated population dose from radionuclide release was 0.3 manrem, due to ^{3}H and ^{14}C, equivalent to 8% of the dose equivalent from prompt radiation.[57]

5. RADIOACTIVITY PRODUCED IN ACCELERATOR SHIELDS AND WATER IN THE GROUND

5.1 Introduction

Accelerators buried in the ground may induce radioactivity in surrounding earth or groundwater. Most of the induced radioactivity will be produced in the accelerator structure and shield and is therefore tightly bound in the constituent materials and is not likely to migrate into the environment. However, it is possible that the radionuclides produced in the earth and groundwater may migrate to the water table. We shall show in this section that the probable levels in radioactive contamination due to these processes are very small.

5.2 Magnitude of the Radioactivity Produced

Particle accelerators are potential sources of large quantities of radioactivity. A rough estimate of the total quantity of radioactivity in Ci produced by a proton accelerator at equilibrium Q_{SAT} may be obtained from the equation:

$$Q_{SAT} \approx \frac{B_i}{3.7 \times 10^{10}} \text{ Ci} \quad , \tag{12}$$

where i is the proton intensity (protons sec^{-1}) and B is a multiplication factor. For the Alternating Gradient Synchrotron at the Brookhaven National Laboratory, B has a value of ~ 24,[58] and thus

$$Q_{SAT} \approx 6.5 \times 10^{-10} \text{ Ci p}^{-1} \text{ s (at 30 GeV)} \; . \tag{13}$$

At an intensity of 5×10^{12} p sec^{-1}, this corresponds to a saturation activity of 3200 Ci. It is reasonable to assume that the total activity will be directly proportional to the beam power and thus we finally obtain for high-energy proton accelerators (E >1 GEV).

$$Q \simeq 145 \text{ Ci kW}^{-1} \; . \tag{14}$$

By contrast high-energy electron accelerators produce less radioactivity by about a factor of 100. Swanson[59] has estimated that at the Stanford 20 GeV electron linac the total inventory of radioactivity in the accelerator structure is ~5 Ci. This quantity of radioactivity is produced by 2% of the total beam energy, which averages 200 kW. The quantity of radioactivity produced per kW of beam power is then

$$Q \simeq 1.25 \text{ Ci kW}^{-1} .$$

In order to evaluate the probable contamination of the environment by this radioactivity, it is necessary to understand not only the magnitude but also the distribution of this radioactivity. Our knowledge in this respect is largely empirical and depends upon experience at several high-energy laboratories.

5.2 (a) Magnitude of the Radioactivity Produced in Groundwater. Stapleton and Thomas[60] have estimated the total quantity of radionuclides at saturation, Q_W, produced in the groundwater in the shield of an accelerator buried underground. Using experimental data[61] they obtain the value:

$$Q_W = 5.59 \times 10^{-16} \text{ Ci mb}^{-1} \text{ GeV}^{-1} \text{ sec}^{-1} \qquad (15)$$

Using the value of 290 mb for the total inelastic cross section of oxygen, we can calculate that the total activity produced in groundwater is $\sim 1.7 \times 10^{-13}$ Ci GeV^{-1} sec^{-1}, which is less than 1% of the total activity produced (Eq. 13).

5.2 (b) Magnitude of the Radioactivity Produced in the Earth. The total activity in the earth shield, Q_E, may be estimated from the equation

$$\frac{Q_E}{Q_W} = \left(\frac{1-f}{f}\right) \frac{\rho_E \sigma_E}{\rho_W \sigma_W} \frac{M_W}{M_E} , \qquad (16)$$

where the subscripts E and W represent earth and water, respectively.

Q_W may be calculated from equation (15). In the case of an accelerator buried in chalk, considered by Stapleton and Thomas,[60] assuming the earth contains 20% water by weight, the values to be substituted into Eq. (16) are:

$$f = 0.2;\ \frac{\rho_E}{\rho_W} \simeq 2.0;\ \frac{M_W}{M_E} \simeq \frac{1}{5.5};\ \frac{\rho_E}{\rho_W} = 5.6$$

whence

$$\frac{Q_E}{Q_W} \simeq 8.1 \quad .$$

Conditions will vary somewhat for other accelerators, but we see that typically the total radioactivity in the earth will be 5 - 10 times higher than the radioactivity in the water.

5.2 (c) Summary. Table 7 summarizes the distribution of radioactivity produced by strong focusing accelerators such as the Brookhaven National Laboratory AGS or the CPS.

5.3 Environmental Impact

The radioactive nuclides induced in the accelerator structure or concrete of the accelerator room are relatively immobile. However, the radionuclides produced in the earth shield or groundwater are free to move.

The radioactive nuclides produced in the groundwater might pass into the general groundwater system and therefore potentially into the public water supplies extracted from the area. In addition, the possibility that activity induced in the earth may be leached into the groundwater system must also be considered.

It is important at the outset to remark that up to the present time no significant groundwater system contamination due to accelerator operation has been observed. However, it is good health physics practice that such a possibility be investigated.

An assessment of a potential contamination of drinking-water supplies falls into three stages:

Table 7. Distribution of radioactivity produced by the CPS or AGS.

Total radioactivity produced	~649 Ci — (100%)
Total radioactivity produced in magnet, accelerator components, concrete room	~603 Ci — (93%)
Total radioactivity produced in earth	~42 Ci — (6%)
Total radioactivity produced in groundwater	~5.2 Ci — (1%)

(i) Consideration of the possible radionuclides which could be produced from a knowledge of the chemical composition of rock and water impurities.

(ii) Estimation of the yield of these radionuclides from the known production cross-section, radioactive half-life, particle flux densities and energy spectra.

Should the yield of radionuclides estimated at stage (ii) be sufficiently high then a third stage follows:

(iii) Estimation of the final specific concentration of radionuclides in local water supplies must be made, taking into account site hydrology, dilution, radioactive decay and chemical sorption.

Several authors have reported the observation of radionuclide production in earth and water, either in laboratory simulations or directly in the accelerator shield. None of them is complete, but together they give a fairly comprehensive picture of the most important radionuclides of concern.

5.3 (a) Radionuclide Production in Water. The most obvious potential source of radioactive contamination of groundwater systems arises from the production of radionuclides directly in the water. Thermal neutron capture in hydrogen and spallation reactions in oxygen and dissolved substances in water may result in a large number of radionuclides. Because these radionuclides are produced directly in water which is mobile, there might be a possibility of their transfer from the site of activation (around the accelerator) and entry into local groundwater systems.

A review of studies of the production of radionuclides in water around accelerators prior to 1972 has been published by Stapleton and Thomas[60,62] and more recent work discussed by Patterson and Thomas.[49]

Table 8 lists the possible spallation products from ^{16}O with half-life longer than 10 sec which will be produced directly in water (listed in order of increasing half-life). The production of these radionuclides has been confirmed in several experimental studies.[48,49,51,53,63,64] Nelson[65] reported the first studies of possible groundwater contamination due to a high-energy accelerator, in estimating radionuclide production near beam dumps of the SLAC 20 GeV electron linac. Middlekoop,[66] in a design study for a 300 GeV proton synchrotron, showed that spallation reactions in water could produce concentrations of ^{3}H and ^{7}Be many times greater than the maximum permissible concentrations recommended by the ICRP. Rindi[67] made detailed calculations of the radioisotopes produced in the cooling water of the CERN 300 GeV proton synchrotron and showed that the presence of large amounts of ^{7}Be can also cause external irradiation exposure from pipes and heat-exchangers.

Measurements of the production of radionuclides in water irradiated in accelerator environments have identified ^{11}C as the dominant short-lived radionuclide 1 to 5 hours after irradiation. ^{7}Be was the only γ-emitter with half-life greater than 10 hours produced in measurable quantities, and was produced with a cross section of about 10 mb in a variety of experimental conditions. The production of tritium under these conditions was consistent with a production cross section of 30-35 mb.[60,64,68] From this information and the data of Gilbert et al.,[61] Stapleton and Thomas estimate that at saturation there are about 6 Ci of ^{7}Be and 18 Ci of tritium in the groundwater of the earth shield around a 300 GeV proton accelerator operating at an intensity of 3×10^{13} protons sec^{-1}.

5.3 (b) Radionuclides Resulting from Dissolved Solids in Water.

Middlekoop[66] was the first investigator to estimate the specific activity due to irradiation of the impurities dissolved in groundwater. His investigation was limited to those radionuclides produced by thermal neutron capture and to only a single example, the groundwater of a chalk region in the United Kingdom.

Table 8. Spallation products from $^{16}_{8}O$.

Nuclide	Half-life
^{10}C	19 sec
^{14}O	71 sec
^{15}O	124 sec
^{13}N	10 min
^{11}C	20.5 min
^{7}Be	53 days
^{3}H	12.2 yr

Middlekoop showed that no nuclides are produced in concentrations at saturation comparable to MPC, with the exception of ^{36}Cl. This radionuclide will be of no consequence, however, because its half-life of 3×10^5 yr will result in an extremely low production rate.

In the experiments of Stapleton and Thomas,[60,64,68] the presence of ^{32}P in the dissolved solids of irradiated groundwater taken from the chalk site was detected. However, ^{32}P is fixed in chalk in the form of calcium phosphate and would not be expected to be mobile in groundwater in chalk soil.[69]

Komochov et al.[70] and Borak[71] reported γ-spectroscopy measurements performed in the cooling waters of the Dubna 700 MeV synchrocyclotron and CERN 20 GeV proton synchrotron showing the presence of radioisotopes which were produced by corrosion of machine parts. ^{60}Co, ^{58}Co, ^{57}Co, ^{56}Co, ^{59}Fe, ^{56}Mn, ^{54}Mn, ^{52}Mn, and ^{22}Na were identified. However, these isotopes are absorbed in the ion exchange resins of the secondary cooling circuit and the probability of finding them in water released to the environment is negligible.

5.3 (c) Radionuclides Produced in the Earth. As in the case of groundwater, several studies of the production of radionuclides in earth have been made and the results are summarized in Table 9.

From the chemical composition of the soil at the CERN site and measurements of particle flux densities in the CPS earth shield, Hoyer[72] calculated the specific activity of radionuclides to be expected in the soil. He concluded that only four radionuclides were produced in measurable quantities. To check his calculations, Hoyer compared his estimates with measured values of the specific activity of ^{45}Ca and ^{22}Na found in earth taken from several locations in the earth shield. Measured activities were, in general, lower by a factor of 3 than those calculated, which Hoyer attributed to leaching of these nuclides from the site by rain water. Hoyer takes the rough agreement between his measured and calculated values to confirm his suggestion that only four radionuclides

Table 9. Radionuclides identified in earth or water at accelerator laboratories.

Laboratory	Accelerator	Soil type	Radionuclides identified In soil	In water	Reference
CERN	28 GeV proton synchrotron	Molasse	^{7}Be, ^{45}Ca, ^{54}Mn, ^{22}Na	^{22}Na	Hoyer, F. (1968)[72]
Rutherford Laboratory	300 GeV proton synchrotron	Chalk	^{7}Be, ^{47}Ca, ^{43}K, ^{32}P, ^{47}Sc	^{7}Be, ^{11}C, ^{3}H	Stapleton, G. B. and Thomas, R. H. (1972)[60]
FNAL Batavia	500 GeV proton synchrotron	Glacial till, various clays	^{7}Be, ^{45}Ca, ^{60}Co, ^{51}Cr, ^{55}Fe, ^{59}Fe, ^{3}H, ^{22}Na, ^{46}Sc, ^{48}V	^{45}C, ^{3}H, ^{54}Mn, ^{22}Na	Borak, T. et al.(1972)[73]
Stanford SLAC	1 GeV electron linear accelerator	Sandstone	^{7}Be, ^{58}Co, ^{59}Fe, ^{54}Mn, ^{22}Na, ^{46}Sc	^{54}Mn, ^{22}Na	Thomas, R. H. (1972)[76]

will be of significance. The production of tritium was not considered in this study.

In studying the radiological impact of the 500 GeV proton synchrotron of the Fermi National Accelerator Laboratory, Borak et al.[73] irradiated glacial till and various clay soils in typical high-energy accelerator radiation fields. Table 9 summarizes the radionuclides identified in these soils.

Since operation of the Fermilab 500 GeV accelerator, an extensive program monitoring the radioactivity in earth and groundwaters at critical areas on the accelerator site has been developed.[30,74,75] Water samples are collected from wells and creeks and no accelerator-produced radionuclides have been detected in such samples. However, in some sumps which collect water from the footings around accelerator tunnels and enclosures and from drains under targets and beam dumps, tritium is routinely detected. Occasionally low concentrations of ^{45}Ca and ^{22}Na are also reported. Baker reports that predictions of radionuclide concentrations in the soil outside the accelerator are now generally reliable. Measurements of particle flux density inside the accelerator tunnels and calculations of the hadronic cascade in the earth shielding by Monte Carlo techniques to predict activities in the earth usually result in good agreement with measured values in the soil.[75]

A similar study to that of Borak et al.[73] was carried out for the design of a 300 GeV proton synchrotron (which might be located in a chalk site but was never constructed).[60] The radionuclides identified in chalk are shown in Table 9.

Finally, Thomas[76] has reported measurements of radionuclides produced in an earth beam dump by 1 GeV electrons from the Mark III electron linear accelerator of Stanford University. The significant production of radioactivity in the earth beam dump was dominated by photo-nuclear reactions with the constituent elements of the soil. The data obtained in this series of measurements were in good agreement with electromagnetic cascade theory.

5.3 (d). The Migration of Radionuclides Through the Ground. A large number of radionuclides may be produced in the earth and groundwater by high-energy accelerator production. However, the ultimate concentration of radionuclides in the groundwater will then be controlled by the half-life of the radioactive species, solubility of the radionuclide, its possible dilution and the site hydrology. Radionuclides with too short a half-life will decay so rapidly as to be of no potential hazard when they reach a public water supply. Conversely, if the half-life is too long saturation activities will not be approached.

Knowledge of the hydrology of the accelerator site being studied will indicate the range of radioactive half-lives that are of interest. It is reasonable to study radionuclides with half-lives in the range:

$$10 \text{ hr} < T < 100 \text{ yr} .$$

It should be noted that this general consideration limits the number of radionuclides that can appear in maximal quantities in groundwater systems. In practice, therefore, only a small number of radionuclides usually need to be studied in detail. Detailed investigation of site conditions will identify the appropriate range to be investigated. In particular, the production of ^{3}H, ^{7}Be, ^{22}Na, ^{54}Mn, and ^{45}Ca merit study.

Not all the radioactivity produced will be in soluble chemical form. Thus, for example, less than 0.7% of the γ activity (principally ^{54}Mn and ^{22}Na) produced in the earth dump of the Mark III electron linac was soluble.[76] Borak et al.[73] have made detailed studies of the solubility of the radionuclides produced in glacial till (Table 9). Of these radionuclides only ^{45}Ca, ^{54}Mn, ^{22}Na and ^{3}H were observed in leach waters. Of ^{45}Ca and ^{54}Mn, only a fraction of the activity (5% and 2%, respectively) was readily soluble.

However, even the production of radionuclides in soluble form in the earth shield and the appearance of radioactive species in the groundwater is not necessarily indicative of environmental harm—the radionuclides may be immobile. In a review on the movement of radioactive wastes buried in the ground, Mawson[77] reports that "with few exceptions, absorption and exchange processes occur between the radionuclides and constituents of the soil... If the site is selected with care any radionuclides that enter the soil will progress quite slowly down the water table. Once in the groundwater they will move faster, but still at a rate one to several orders of magnitude less than the rate of movement of the groundwater. These statements apply to most cations--many anions move at about the same speed as the groundwater."

The greater part of radioactivity induced in the soil is confined to regions of high radiation intensity and is typically confined to a few locations close to the accelerator. Stapleton and Thomas[60] estimate that 95% of the activity induced in earth is produced within 2 m from the outer wall of the accelerator tunnel of a proton synchrotron. In the absence of migration of any radionuclides from the activation zone, the radioactivity would increase to a final saturation determined by the accelerator intensity, the nuclear cascade process in the shield and the macroscopic cross sections for radionuclide production in rock and groundwater.

Migration of the radionuclides will be determined by the distribution coefficient, K, which is a measure of the relative affinity of the radionuclide for earth compared with water. It is defined as the ratio of the concentration of ions absorbed by the soil to those remaining in solution at equilibrium. A large value of K indicates that ions preferentially adhere to soil and their migration is retarded.

The average velocity of ions through the earth, v_i, is related to the velocity of the water, v_w, by the equation

$$\frac{v_i}{v_w} = \frac{1}{1+k(\rho/p)} \quad , \tag{17}$$

where k is the distribution coefficient (ml g^{-1}),

ρ is the bulk density of the rock, and

p is the porosity of the rock (fraction of total volume of soil occupied by voids).

For large distribution coefficients equation (17) reduces to:

$$\frac{v_i}{v_w} = \frac{p}{k\rho} \quad . \tag{18}$$

Table 10 summarizes measurements of the distribution coefficient, K, for some radionuclides in various soils.

If the distribution coefficient is high, the potential for contamination of public water supplies is much reduced. Thus Stapleton and Thomas point out that for ^{7}Be in chalk the beryllium ions move more slowly than the groundwater by a factor of about 15,000. Under these conditions migration rates would be so slow that radioactive decay will reduce the original activity to negligible levels.[78]

In the case of ^{22}Na there seems to be some significant difference between the observations of Hoyer, who determined a distribution coefficient of 10^2 at CERN, and those of Borak et al. at FNAL. The latter workers found in the case of ^{22}Na, 10-20% of the activity is produced in a chemical form which is extremely soluble in water. In equilibrium the distribution coefficient was determined to be 0.20 and the ion velocity about 40% of that of the groundwater.

Of all the radionuclides produced in the earth and groundwater, it is likely that only tritium will move freely in groundwater, without significant holdup due to absorption on rock surfaces. In static measurements of distribution coefficient, Borak et al. report that the quantity of tritium found in leach waters

Table 10. Values of the distribution coefficient, K.

Radionuclide	Soil type	Distribution coefficient	Reference
^{7}Be	Chalk	~ 4500	78
^{22}Na	Molasse*	100	72
^{22}Na	Glacial till	0.2	73
^{32}p	Molasse*	100	72
	chalk	Very high	69
^{45}Ca	Molasse*	1000	72
^{54}Mn	Molasse*	100	72

*Arenaceous rocks, typical of alpine orogeny, related to the Flysch formation.

was consistent with the quantity produced in the free water in the soil. The distribution coefficient measured was very small and thus the tritium will migrate with the same velocity as water through the aquifier.

It is therefore probable that tritium may be the radionuclide that should be most carefully studied in groundwater around accelerators.

5.3 (e) Potential Contamination of Drinking Water Supplies. From what has gone before we can speculate that the possibility of significant contamination of drinking water supplies is extremely remote.

A detailed study of these phenomena would be extremely complex, but Thomas has proposed a simple model that may be used to understand their magnitude.[62] This model is schematically shown in Figure 9, where an accelerator is buried underground. Radioactivity is induced in an "activation zone" close to the accelerator building. It is postulated that the radionuclides may be washed downward to the water table by rainfall. When they reach the water table they are transported to the boundaries of the accelerator laboratory. In the movement they are mixed with and diluted in the groundwater. This water might be available for public use.

The specific activity, S, of this water available to the public is then given by

$$S = D \sum_i \frac{\varepsilon_i Q_i \left(1-e^{-T_i/\tau_i}\right) e^{-t_i/\tau_i}}{M_i} \quad , \qquad (19)$$

where there are i radionuclide produced and

D is a dilution factor,

ε_i is the fraction of activity produced that migrates from the site of its production,

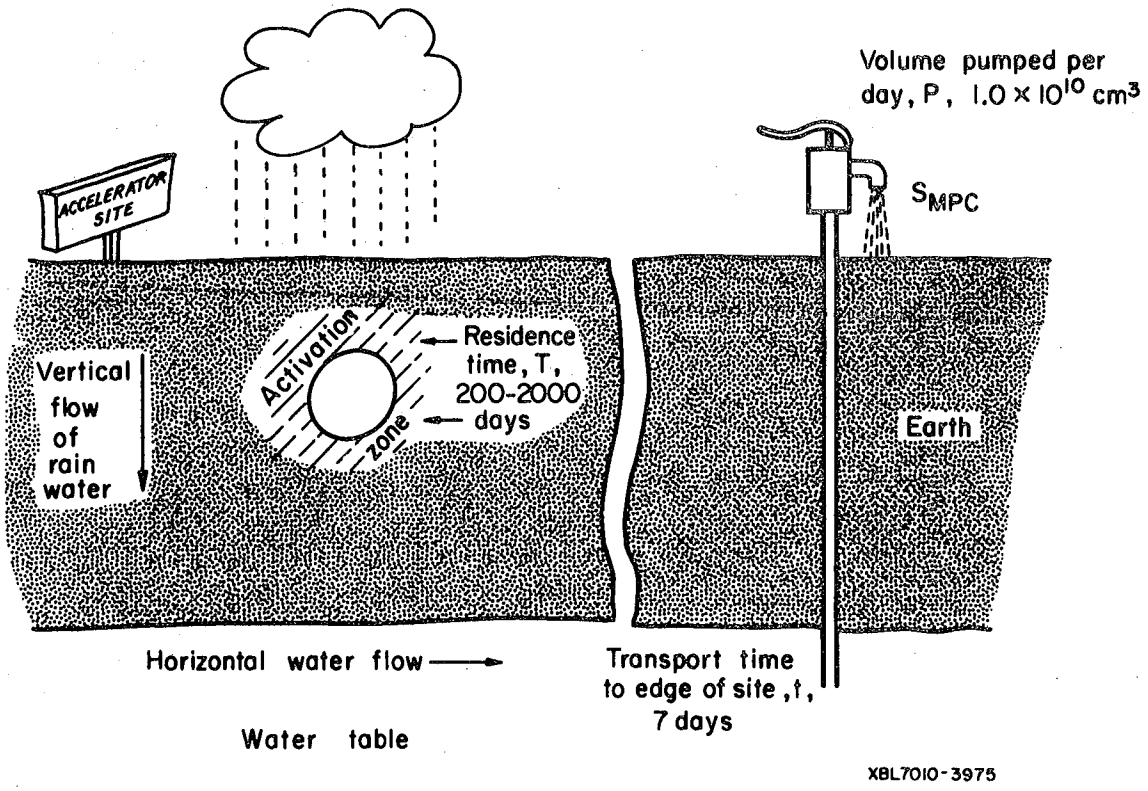


Figure 9. Model illustrating the mechanism by which accelerator-produced radioactivity may appear in groundwater.

Q_i is the total quantity of the ith radionuclide produced at saturation,

T_i is the residence time in the activation zone,

τ_i is the mean life of the ith radionuclide,

t_i is the transport time from leaving the activation zone to reaching the laboratory perimeter,

M_i is the MPC of the ith radionuclide, and

S is measured in units of maximum permissible concentration (MPC).

Equation (19) permits a crude assessment of the magnitude of the problem. The maximum rate of release of activity occurs at small residence times (T = 0), when all the activity produced migrates ($\varepsilon = 1$) and the transport time to the site boundary is very short ($e^{-t_i/\tau_i} = 1$) when:

$$S_{Max} = D \sum_i \frac{Q_i}{M_i \tau_i} \quad . \qquad (20)$$

At an accelerator site where the water table is not disturbed by pumping, the outflow of water would equal the inflow from rainfall and the radioactivity released would be diluted in a volume of water equivalent to the rainfall on the site.

If, as we have seen, it is likely that tritium is the only radionuclide of significance, the value of S_{Max} reduces to:

$$S_{Max} = D \frac{Q_H}{M_H \tau_H} ,$$

where the subscript H refers to tritium.

If we are considering a large accelerator site such as CERN or the Fermi lab the factor D can be quite small — typically 10^{-10} ml^{-1} day—and substituting other typical values:

$1/D = 10^{10}$ ml day^{-1}

$Q_H = 20$ Ci

$\tau_H = 17.6$ yr

$M_H = 3 \times 10^{-3}$ μC ml^{-1} (general population),

we obtain:

$$S_{Max} \approx 1 \times 10^{-4} \text{ MPC} .$$

Thomas[62] has reported calculations for a 500 GeV proton synchrotron similar to the CERN 28 GeV proton synchrotron, losing 10^{12} protons per second to the shield. It was assumed that all the radionuclides produced in the earth and water in the shield were released directly to the groundwater, rapidly transported to the site boundary and diluted with a quantity of water equivalent to the net rainfall on the accelerator site (10^{10} ml/day). Figure 10, which summarizes these calculations, shows that even under these extremely conservative assumptions the specific activity of the water would never exceed 0.03 MPC and that this value is rather insensitive to residence time up to periods of 1000 days. This crude treatment shows that the problem is not likely to be a serious one, and in actual practice the magnitude of radioactive contamination of drinking water supplies will be extremely small.

5.4 Population Dose Equivalent from Accelerator-Produced Nuclides in Ground Water

The arguments presented in Section 3.6 show that the concentration of the accelerator-produced nuclides will be insignificant at existing accelerator intensities.

The radioactivity from natural sources will in fact be higher by orders of magnitude than from accelerator-produced nuclides.

There have been no reported observations of accelerator-produced radionuclides in public water supplies and the consequent population dose equivalent is therefore effectively zero.

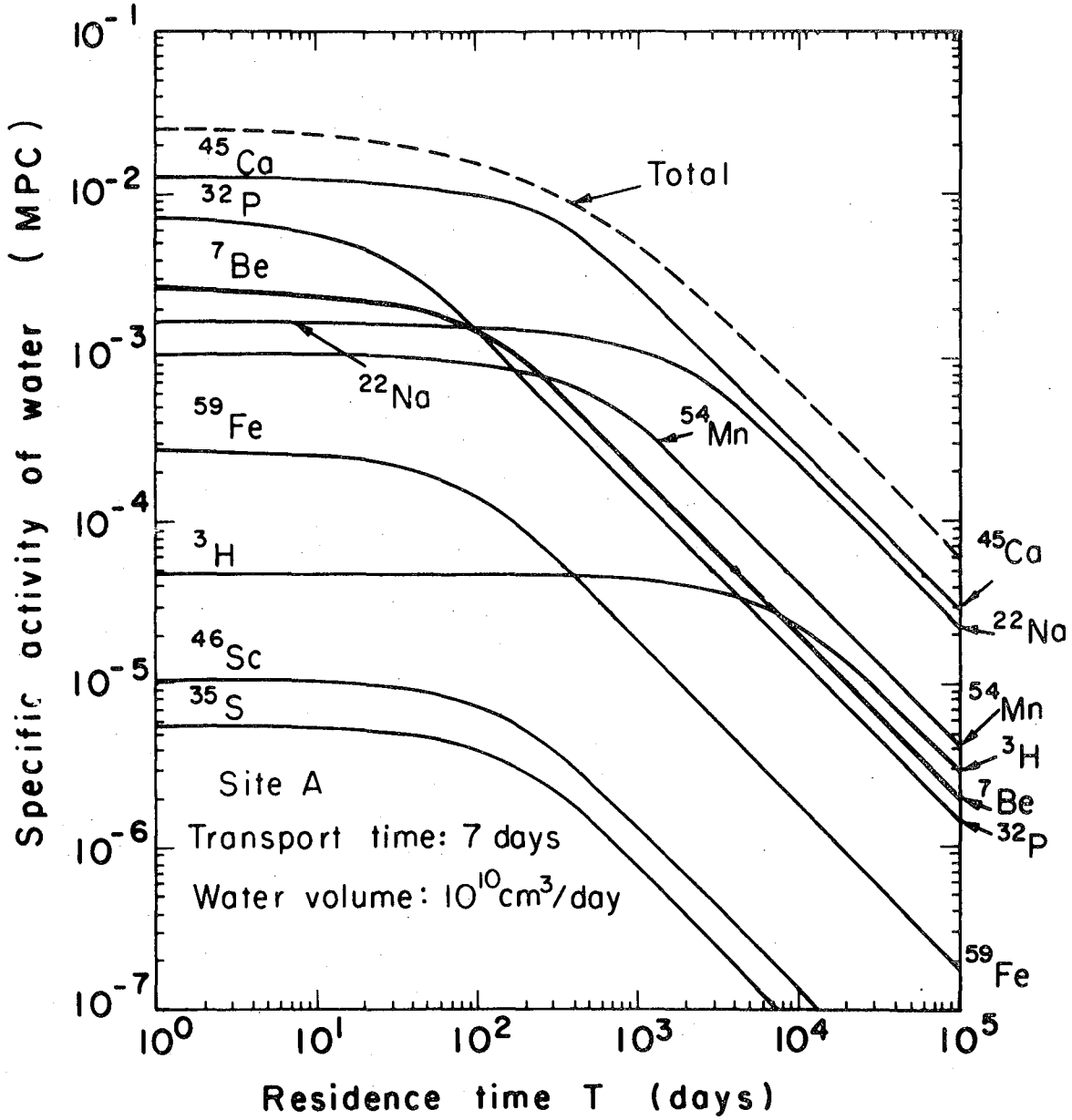


Figure 10. The specific activity of accelerator-produced radionuclides in the groundwater at a hypothetical 500 GeV accelerator site, as a function of water residence time in the activation zone.

6. CONCLUSIONS

The radiological environmental impact of high-energy accelerators is qualitatively different from most other nuclear facilities. Prompt radiation produced when the accelerator is in operation is the principal source of population exposure.

Typically the population exposure due to prompt radiation, radioactive gases and the radioactive contamination of water supplies are in the proportion $100 : <10 : \ll 1$.

For high-energy, high-intensity facilities, neutrons are the principal component of this prompt radiation field. At some electron facilities, or thinly shielded proton accelerators, photons may be a significant source of dose equivalent, while at the highest energies, muons have been detected. The magnitude of population dose equivalent due to high-energy accelerator operation is estimated to be in the range 50-1000 man rem per rem at the site boundary. Typical annual dose equivalents at accelerator boundaries are $\sim$10-50 mrem, with resultant population dose equivalents of the order of $\sim$10-50 man rem.

The production and release of radionuclides to the atmosphere is the other potentially significant source of population exposure. Relatively long-lived radionuclides such as ^{41}Ar, ^{7}Be, and ^{3}H are of potential concern, but the population dose equivalent from the source is at least an order of magnitude lower than that due to prompt radiation. Observations of the concentration of ^{7}Be in rainwater, on the ground surface and on vegetation, provide a convenient monitor of potential environmental contamination.

No significant concentration of accelerator-produced radionuclides has been reported in groundwater systems near high-energy accelerators—but calculation shows that none is to be expected. Tritium is the only likely radionuclide to move freely in the groundwater. No significant population exposure from this source is expected at present energies and intensities.

It is of interest to compare the radiological environmental impact of typical high-energy accelerators with the recommendations of government agencies that site boundary levels at nuclear facilities should be restricted

to 25 mrem/yr or that the population dose equivalent should not exceed 100 man rem/yr. Experience shows that, typically, high-energy accelerators are well below both design goals.

Finally, since this lecture is being given in Japan, I would like to mention the important work which is being done here that will improve our understanding of the radiological impact of high-energy accelerators on the environment.

The "skyshine collaboration" between the Universities of Kyoto, Tokohu, and Tokyo, and the Japan Atomic Energy Research Institute[26] will provide valuable information on the transport of photons and neutrons through the atmosphere. Indeed, work from the Institute for Nuclear Study[27] and KEK[79] has already been published. The work at the Institute for Nuclear Study is of particular interest in that photon spectra are being measured.

Careful radiation surveys around the KEK-PS are improving our knowledge of the quantity, species and distribution of the radionuclides produced[80] and the MPC's for many accelerator-produced radionuclides[81,82] calculated.

In Japan, Government Agencies are very concerned with air-borne radioactivity; as Katoh has pointed out, perhaps over concerned.[83] This concern will, however, result in better measurements and an improved understanding of this aspect of the radiological impact of high-energy accelerators —an impact which this paper attempts to show is very small indeed.

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