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POSSIBLE CONTAMINATION OF GROUND WATER SYSTEMS BY HIGH ENERGY PROTON ACCELERATORS

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ABSTRACT

A general evaluation of the problem of possible ground water contamination due to the release of accelerator produced radionuclides to ground shows it to be an unlikely problem for existing and projected high energy accelerators in the energy region of several hundred GeV.

I. INTRODUCTION

High energy particle accelerators are not often thought of as potential polluters of the environment. Indeed by comparison with nuclear reactors they are rather puny in their ability to produce radioactivity. Thus, for example, it will be shown here that the total inventory of tritium produced in the ground water of earth shields of the new generation of high energy accelerators currently under construction (1) or being designed (2) (3) is about ten curies of tritium at saturation.

High energy accelerators are nevertheless potent radiation sources and need to be heavily shielded both for radiation safety and to permit their experimental utilization. Large strong-focusing accelerators (such as at Brookhaven National Laboratory in the United States, CERN in Western Europe, or Serpukhov in the Soviet Union) are buried underground to provide the necessary shielding. This method of construction brings the surrounding shield in fairly close proximity to the accelerator, and, if substantial particle fluxes may be generated in the earth and ground water, it is possible that radioactivity may ultimately appear in local ground-water systems. Thus, although the total inventory of tritium in the ground water of the shield is only ten curies, it is produced directly in the environment--a release comparable to or in excess of that reported for many power reactors (4). Although the particle fluxes appearing in the earth may be quite small compared with those produced by nuclear reactors, they may nevertheless be sufficient to induce specific activities in excess of those recommended by the International Commission on Radiological Protection (ICRP) (5) as fit for human ingestion. Thus, for example, a high energy flux only $\approx 10^5$ neutrons/cm² sec will produce a maximum permissible concentration (MPC) of tritium by spallation in water.

The first reported study of possible contamination of ground water by accelerator-produced radionuclides is due to Nelson(6) at the Stanford Linear Accelerator Center, who estimated the radioactivity induced in ground water close to beam dumps at the SLAC 20-GeV electron linear accelerator. The expectation that no significant water contamination would arise has been subsequently confirmed by environmental radioassays at SLAC(7) and by measurements at the Stanford Mark III electron linear accelerator(8). Hoyer(9) has reported the observation of measurable quantities of Na-22 in drainage water from the CERN 25 GeV Proton Synchrotron (CPS) but concluded that-- "at working conditions of those at the CERN Proton Synchrotron at present . . . -- no hazard exists from the effluent drainage water or even manipulation of the soil near target regions. But it cannot be ruled out that radioactive isotopes, especially Na-22 and Be-7, near or above maximum permissible concentrations may be washed out from the soil surrounding future high energy accelerators or be directly built* in the ground water . . . "

Thus it is clear that the question of possible ground water contamination by high energy accelerators merits some detailed study, since it could be an important factor in accelerator design or siting, and, in any case, such pre-operational studies are good health physics practice. Furthermore, in a climate of increasing public concern about water pollution it is valuable to have a closely documented study for public information.

This paper presents a simple model which may be used to rapidly assess the magnitude of possible contamination problems at any accelerator site. General considerations show that it is unlikely that high energy accelerators, of the energy and intensity planned for the near future, will produce significant concentrations of radionuclides in water systems at site boundaries. The recommendations of Committee II of the ICRP(5) for continuous occupational exposure, reduced by a factor of 10, have been accepted in this paper as providing acceptable levels for the concentration of radionuclides in drinking water supplies.

Nevertheless, it is recommended that detailed studies at particular sites be made to ensure that no hydrological conditions apply which result in high concentrations of radioactivity in ground water systems. Some typical examples are given.

* produced (author)

2. GENERAL FORMULATION OF THE PROBLEM

In general, when the specific activity of a radionuclide appearing in the ground water system contiguous to a high energy accelerator merits concern, estimates of the residence time in the activation zone, transport time into the public water supply, and time for mixing, dilution, sorption in the aquifer will be needed.

Thus, if the total inventory of a particular nuclide at saturation in the activation zone is Q_0 curies, the quantity of activity reaching the public water supply, Q , can be written

$$Q = \epsilon Q_0 (1 - e^{-T/\tau}) e^{-t/\tau} \quad (1)$$

where

τ is the mean life of the nuclide,

T is the residence time in the activation zone,

t is the time taken to reach the public water supply after leaving the activation zone,

ϵ is the fraction of activity produced that flows freely in the ground water.

Dilution is difficult to estimate, but limits may be set by considering dilution resulting from pumping water from the general area. Thus the specific activity, S , of water reaching public water supplies may be written

$$S = DQ \quad (2)$$

where D is a dilution factor.

The total specific activity reaching the public water supply in units of MPC is given by:

$$S_{MPC} = D \sum_i \frac{\epsilon_i Q_i (1 - e^{-T/\tau_i}) e^{-t/\tau_i}}{M_i} \quad (3)$$

where M_i is the MPC of the i^{th} nuclide.

The logical sequence of this study is an evaluation of the parameters of Eq. (3). It will be seen that the chemical sorption parameter

ϵ plays an extremely important role.

3. MAGNITUDE OF THE PROBLEM - SELECTION OF RADIONUCLIDES TO BE STUDIED

Equation (3) shows that the maximum rate of release of activity occurs at small residence times when:

$$\left[S_{MPC} \right]_{\max} = D \sum_i \frac{\epsilon_i Q_i}{M_i \tau_i} e^{-t/\tau_i} \quad (4)$$

If the conservative assumptions are made that all activity produced is released to the ground water and that transport to the edge of the site occurs very quickly, we may write:

$$\left[S_{MPC} \right]_{\max} \leq D \sum_i \frac{Q_i}{M_i \tau_i} \quad (5)$$

If the activity released can be considered to be associated with the inflow of water due to rainfall in the accelerator site, the average rate of outflow of water--from a site of some 20 km² in area with net average inflow due to rainfall (corrected for evaporation and runoff) of 0.20 m/yr--would be approximately 10¹⁰ cm³/day.

Fairly elementary considerations lead one to conclude that the total radionuclide production of a 500 GeV accelerator losing some 10% of its beam intensity of 10¹³ protons/sec⁻¹* would be about 1500 curies in the accelerator components, accelerator room and surrounding shield. Of this 1/3 or less, depending upon precise details of accelerator construction, can appear in the earth. Because short-lived nuclides will be of no concern, we might expect some 10% of the total activity produced in the shield-- ~50 curies--could in principle contribute to ground water contamination.

Gabriel et al(10, 11) have estimated the mean life of a mixture of radionuclides produced in the earth shield of a high energy accelerator as ~ 1-2 years. Thus substituting the values

*This would be considered an unusually high beam loss.

$$\begin{aligned}\sum Q_i &= 50 \text{ Ci} \\ D &= 10^{-10} / \text{ml} \\ \tau &= 365 \text{ days} \\ M &= 3 \times 10^{-6} \text{ } \mu\text{c/ml for unidentified} \\ &\quad \text{accelerator-produced radio-} \\ &\quad \text{nuclides}\end{aligned}$$

into Eq. (5) we obtain:

$$\left[S_{\text{MPC}} \right]_{\text{max}} \leq 5 \text{ MPC}$$

Although this simple argument does not, ab initio, completely eliminate the possibility of ground water contamination, it does indicate that such an eventuality is unlikely. In general the total inventory of long-lived radionuclides will not be released to the ground water, they will not be produced at maximal quantities and the MPC's will be larger than the restrictive value of $3 \times 10^{-6} \mu\text{c/ml}$.

Table I gives the quantity of some typical accelerator produced radionuclides that would give a concentration of 1 MPC if released at maximum production rate into 10^{10} ml water per day.

Although the number of radionuclides produced in the earth and ground water in an accelerator shield is potentially very large, only a few can be produced in maximal quantities in ground-water systems.

Equation (4) shows that radionuclides of most concern will:

- a. Be produced in large quantities, and/or
- b. have a low MPC,
- c. pass efficiently into the groundwater system,
- d. not decay significantly in being transported to a public water supply.

Nuclides with short half-lives will decay so rapidly as to be of no potential hazard when they reach a public water supply. Conversely, if the half-life is long the production rate will be too small and the nuclide will not appear in significant quantities. Knowledge of the hydro-geology

Table I. Quantity of typical accelerator-produced radionuclides, Q, that would result in a concentration of one MPC if released at maximum rates.

| Nuclide | MPC, M ($\mu\text{c}/\text{cm}^3$) | Mean Life, τ (days) | Q (Ci) |
|---------|---|--------------------------------|-----------|
| P-32 | 2×10^{-5} | 20.63 | 4.1 |
| Fe-59 | 6×10^{-5} | 65 | 39 |
| Be-7 | 2×10^{-3} | 77.04 | 1540 |
| Sc-46 | 4×10^{-5} | 121 | 48 |
| S-35 | 6×10^{-5} | 126 | 76 |
| Ca-45 | 9×10^{-6} | 220 | 20 |
| Mn-54 | 1×10^{-4} | 410 | 410 |
| Na-22 | 4×10^{-5} | 1370 | 550 |
| H-3 | 3×10^{-3} | 6428.4 | 190,000 |

of the accelerator site being studied will indicate the range of radioactive half-lives that are of interest. It is usually reasonable to study radionuclides with half-lives in the range 10 hours $< T < 100$ years, but detailed investigation of site conditions will identify the appropriate range to be investigated. Those nuclides in this range of half-life that also satisfy conditions (a), (b), and (c) are fortunately few in number.

4. PRODUCTION OF RADIOACTIVITY IN ACCELERATOR SHIELDS

4.1. Neutron Activation

Several experimental(12) and theoretical studies(13, 14, 15) have shown that the production of radioactivity in accelerator shields is largely

due to neutrons.

Photon-induced activation is insignificant, other than very close to the accelerator vacuum chamber. By consideration of the equilibrium spectra measured in accelerator shields(12) and the relative population of neutrons and charged particles(13) it can be shown that the greater part of the radionuclide production will arise from neutron interactions between a few MeV and a few hundred MeV. Two mechanisms for the production of radioactivity are therefore important.

- a. Thermal Neutron Capture. Neutrons generated in the nuclear cascade are ultimately thermalized and captured in the surrounding earth and water. This mechanism is clearly more important in earth than in ground water because of the high thermal neutron absorption cross section of some of the constituent elements of the soil and the relatively low concentrations of impurities in the ground water. Hughes and Schwartz(16) have published a compilation of thermal neutron absorption cross sections.
- b. High Energy Reactions. Neutrons of more than a few MeV may produce reactions of the type (n, p) , (n, pn) , (n, α) , etc.

Thermal neutron activities may be readily calculated from known thermal flux densities and the cross sections tabulated by Hughes and Schwartz(16). Estimates of induced activity by the high energy reactions is a little more difficult because the production cross section is a function of neutron energy. The evaluation of specific activities thus requires knowledge of cross sections and neutron spectra. It is appropriate to note at this point that high energy cross sections may be calculated from fundamental nuclear theory to a fair accuracy(17, 18). In studies such as this, where high precision is not required, neutron cross sections may thus be evaluated with sufficient accuracy, in most cases to within less than a factor of 2, in the absence of experimental data. Bruninx(19) has tabulated high energy nuclear reaction cross sections for charged particle energies greater than 50 MeV which are of value in estimating nuclide yields.

The three important sources of radionuclide production in an earth shield are neutron spallation in the water, neutron absorption, and spallation in the soil. Neutron capture in the water is unimportant, while neutron reactions in dissolved impurities in the water are generally inconsequential because of their low concentration.

4.2. Neutron Spallation Processes in Water

Table II lists the possible spallation products from O-16 produced

directly in water, listed in order of increasing half-life. We have seen that the short-lived nuclides will be of little consequence, but Be-7 or H-3 may be produced in sufficient concentrations to cause concern. Be-7 is readily absorbed on rock surfaces, but H-3 passes freely through ground-water systems and is usually the oxygen spallation product of greatest concern.

The specific activity of a nuclide produced by neutron spallation in the accelerator shield is given by

$$S_0(i) = \sum_j N_j \int_{E_{ij}}^{E_{\max}} \sigma_{ij}(E) \phi(E) dE, \quad (6)$$

where

- $S_0(i)$ is the specific activity of the nuclide i ,
- N_j is the number of target nuclei of type j ,
- $\sigma_{ij}(E)$ is the cross section for production of the radionuclide i from target nucleus j by a neutron of energy E ,
- E_{ij} is the threshold energy for the reaction,
- $\phi(E)dE$ is the neutron differential energy spectrum.

Table II. Spallation products from O-16.

| <u>Nuclide</u> | <u>Half-life</u> |
|----------------|------------------|
| C-10 | 19 sec |
| O-14 | 71 sec |
| O-15 | 124 sec |
| N-13 | 10 min |
| C-11 | 20.5 min |
| Be-7 | 53 days |
| H-3 | 12.2 years |

Measurements of Be-7 and H-3 production in water in a variety of accelerator spectra(20) and by monoenergetic protons (19,20) suggests it appropriate to use production cross sections of 10 mb and 30 mb independent of energy for Be-7 and H-3 production for oxygen spallation, respectively.

For an energy-independent cross section Eq. (6) reduces to:

$$S_0(i) = \sum_j N_j \sigma_{ij} \int_{E_{ij}}^{E_{\max}} \phi(E) dE \quad (7)$$

$$= \sum_j N_j \sigma_{ij} \Phi \text{ (for } E > E_{ij} \text{)} \quad (8)$$

where

$\Phi(E > E_{ij})$ is the integral flux density of neutrons of energy greater than E_{ij} .

4.3. Radionuclide Production in Earth

The problem of evaluating the production of radionuclides in earth is extremely complex. Gabriel et al (10, 11) have reported some approximate calculations of the production of radionuclides in earth but to quote the authors, ". . . there is considerable uncertainty in the activity . . . where it is largest." Refinements in the calculations is to be anticipated but in the meantime direct experimental measurements must be used. Hoyer (9) has measured Na-22 and Ca-45 produced in the earth shield of the CPS, and Borak et al. (21) of ANL and NAL have reported preliminary measurements in glacial till. These two studies naturally show differences in the radionuclides produced in largest quantities, depending upon the chemical composition of the soil irradiated. Because the neutron flux distribution in the CPS earth shield is well understood (12), the data due to Hoyer (9), who has studied radionuclides having half lives larger than two days, will be discussed in some detail.

From the known chemical composition of the earth and measured flux densities (12, 22) the specific activity of earth near target regions was calculated. Table III lists the radionuclides identified.

To check his calculations, Hoyer compared his estimates with measured values of the specific activity of Ca-45 and Na-22 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. But, it seems reasonable to assume from these measurements that the values calculated by Hoyer are unlikely to err on the low side.

4.4. Total Production of Radionuclides by a High Energy Accelerator

Once the radionuclide yield per unit flux density in water and

Table III. Long lived radionuclides produced in CPS earth shield (after Hoyer).

| Radionuclides | Half-life | MPC $\mu\text{c/ml}$ | Production reaction | Saturation activity per unit flux density $\mu\text{c cm}^2 \text{ sec/g n}$ |
|---------------|-----------|-------------------------|---|--|
| P-32 | 14.3 d | 2×10^{-5} | S-32(n, p)P-32 | 3.3×10^{-11} |
| Fe-59 | 45 d | 6×10^{-5} | Fe-58(n, γ)Fe-59 | 2.8×10^{-11} |
| Be-7 | 53 d | 2×10^{-3} | O-16(n, spall)Be-7 | $< 4.3 \times 10^{-9}$ |
| Sc-46 | 84 d | 4×10^{-5} | Mn-55(n, spall)Sc-46 | $< 5.5 \times 10^{-13}$ |
| S-35 | 87 d | 6×10^{-5} | S-34(n, γ)S-35 | $< 1.1 \times 10^{-12}$ |
| Ca-45 | 153 d | 9×10^{-6} | Ca-44(n, γ)Ca-45 | 6.2×10^{-10} |
| Mn-54 | 278 d | 1×10^{-4} | Fe-54(n, p)Mn-54 Mn-55(n, 2n)Mn-54 | 3.3×10^{-10} |
| Na-22 | 2.6 y | 4×10^{-5} | Na-23(n, 2n)Na-22 Al-27(n, spall)Na-22 | 2.33×10^{-10} 5.75×10^{-10} |

ground is known, the total production in an accelerator shield may be estimated.

If the distribution of specific activity of a given radionuclide in an accelerator shield can be estimated, then the total production of that nuclide may be obtained by spatial integration. A proton accelerator may be considered, for the purpose of calculating induced activity, as a source of neutrons. In the high-energy strong-focusing proton synchrotrons it is sufficiently accurate to ignore the radial curvature of the accelerator. Thus, if the specific activity at a position (r, z) in the shield is S(r, z) at a given time, then the total quantity of the nuclide contained in the shield, Q, is given by

$$Q = \iint 2\pi r S(r, z) dr dz, \quad (9)$$

with appropriate limits to the integration. We can write

$$S(r, z) = (f\rho \frac{L}{A}) \sigma \Phi_{E_0}(r, z), \quad (10)$$

where f = fraction by weight of target element in shield,
 ρ = shield density,
 L = Avogadro's number,
 A = atomic weight of target element,
 σ = production cross section,
 $\Phi_{E_0}(r, z)$ = integral neutron flux density $E > E_0$ at the point r, z .

For nuclides produced by spallation in water we have

$$S(r, z) = \left(\frac{L}{M}\right) \sigma \Phi_{E_0}(r, z), \quad (11)$$

where M = the molecular weight of water.

Thus the total quantity of a radionuclide at saturation Q_0 is given by

$$Q_0 = \left(\frac{f\rho L}{A}\right) \sigma \int_a^\infty \int_0^\infty 2\pi r \Phi(r, z) dr dz, \quad (12)$$

where a = effective radius of the accelerator room (cylindrical geometry assumed)

If the distribution $\Phi(r, z)$ is known, it follows Q_0 may be evaluated.

Measurements of the spatial and energy distribution of neutrons in the CERN proton synchrotron earth shield have been reported in great detail elsewhere (12). Briefly, measurements of neutron flux densities were made with various threshold detectors at many positions in the shield. For all neutron energies of concern to us here and, with accuracy sufficient for our purposes, the flux-density distribution in the shield region around an internal target may be conveniently written

$$\Phi(r, z) = \left(\frac{a}{r}\right) \Phi_{\max} \exp\left(-\frac{r-a}{\lambda}\right) \exp(-z/\mu), \quad (13)$$

where cylindrical geometry is assumed and $\Phi(r, z)$ is the neutron flux density of neutrons of energy greater than E_0 at the position r, z . Thus substituting for $\Phi(r, z)$ into Eq. (12) and integrating, we obtain

$$Q_0 = 2\pi f \sigma \left(\frac{\rho L}{A}\right) (a\mu\lambda) \Phi_{\max} \text{ (for } E > E_0 \text{) dis/sec.} \quad (14)$$

Measurements at CERN gave values

$$\begin{aligned} a &= 390 \text{ cm,} \\ \mu &= 650 \text{ cm,} \end{aligned}$$

* This model simplifies the actual situation because the maximum flux density observed in the shield does not occur at the point $r = a, z = 0$. It is understood, however, that the maximum flux density observed will be substituted for Φ_{\max} .

$$\begin{aligned}\lambda &= 53 \text{ cm,} \\ \rho &= 2.16 \text{ g/cm}^3 ,\end{aligned}$$

and substituting into Eq. (14) we obtain

$$Q_0 = 1.10 \times 10^5 \left(\frac{f}{A}\right) \sigma \Phi_{\max} \text{ (for } E > E_0) \text{ dis/sec,} \quad (15)$$

when σ is in millibarns.

The measurements at CERN also support the view that the number of secondary particles generated in the shield is proportional to beam power. Thus Q_0 may be expressed in terms of beam power lost to the accelerator shield:

$$\begin{aligned}Q_0 &= 1.10 \times 10^5 \left(\frac{f}{A}\right) P \sigma \eta(E_0) \text{ dis/sec,} \\ &= 2.97 \times 10^{-6} \left(\frac{f}{A}\right) P \sigma \eta(E_0) \text{ Ci}\end{aligned} \quad (16)$$

where P is the beam power lost to the shield in units of GeV/sec,

$\eta(E_0)$ are values of maximum integral neutron flux density, $\Phi_{\max}(\text{for } E > E_0)$, observed in the CERN PS accelerator shield per GeV/sec.

Values of $\eta(E_0)$ based on observation at CERN are given in Table IV.

In calculating the radionuclide production the following relationships between total quantity of nuclides in the shield, at saturation, and the maximum specific activity of the nuclide in rock or water are helpful. For nuclides produced in rock,

$$\left(\frac{Q_0}{S_{\max}}\right) = 2 \pi a \mu \lambda ; \quad (17)$$

for nuclides produced in water,

$$\left(\frac{Q_0}{S_{\max}}\right) = 2 \pi f_w^* \rho a \mu \lambda, \quad (18)$$

where f_w is the fraction by weight of water in the rock. Substituting for a , μ and λ , we get for the CPS:

* $f_w = 0.15$ at CERN(12).

$$\text{in rock, } \left(\frac{Q_0}{S_{\max}} \right) = 8.44 \times 10^7 \text{ g,} \quad (17a)$$

$$\text{in water, } \left(\frac{Q_0}{S_{\max}} \right) = 2.74 \times 10^7 \text{ ml.} \quad (18a)$$

Finally, if the specific activity of a nuclide in rock is measured directly and expressed as χ $\mu\text{C/g/n/cm}^2 \text{ sec}$, the maximum specific activity of that nuclide appearing in the accelerator shield will be:

$$S_{\max} = P \eta(E_0) \chi \text{ } \mu\text{Ci,} \quad (19)$$

and
$$Q_0 = 8.44 \times 10^7 P \eta(E_0) \chi \text{ } \mu\text{Ci,} \quad (20)$$

Table IV. Values of $\eta(E_0)$.^a

| Neutron energy (MeV) | $\eta(E_0)$ (n/cm ² GeV) ($\times 10^{-8}$) |
|----------------------|--|
| 1 | 3.6 |
| 2 | 3.5 |
| 3 | 3.4 |
| 4 | 3.3 |
| 6 | 3.1 |
| 8 | 3.0 |
| 10 | 2.8 |
| 20 | 2.4 |
| 30 | 2.1 |
| 40 | 1.9 |
| 50 | 1.8 |

a. Based on measurements at 10^{12} p/sec and 25 GeV.

The values of $\eta(E_0)$ given in Table IV are based on an estimate of a maximum flux density of 6×10^5 particles/cm² sec of energy greater than 20 MeV produced in the earth shield of the CPS with a single target in operation at a beam intensity of 10^{12} protons sec⁻¹ at 25 GeV, and the measured neutron spectrum in the earth(12). The corresponding maximum thermal neutron flux density was 2.5×10^5 n/cm² sec.

Thus, using Eq. (16) the production of Be-7 and H-3 in the ground water of an accelerator shield may be calculated. Equation (20) and the

data of Table III give the production of nuclides in the earth.

Based on these discussions estimates of radionuclide production in the shield of a 500 GeV accelerator with intensity 10^{13} protons/sec, of which 10% is lost to the shield due to inefficient beam extraction are given in Table V.

Since overall beam extraction efficiencies of 95% are confidently predicted and efficiencies better than 99% expected at some laboratories (26), these estimates represent an upper limit of the induced activities to be anticipated in practice. (The extracted beam will be disposed in beam dumps specially designed to contain the induced radioactivity.)

Table V. Radionuclide production in accelerator shield.

| <u>Nuclide</u> | <u>Mean life (days)</u> | <u>MPC ($\mu\text{c/ml}$)</u> | <u>Saturated activity (Ci)</u> |
|----------------|-----------------------------|--|------------------------------------|
| P-32 | 20.6 | 2×10^{-5} | 4.2×10^{-2} |
| Fe-59 | 64.9 | 6×10^{-5} | 1.2×10^{-2} |
| Be-7 | 77.0 | 2×10^{-3} | 4.4 |
| Sc-46 | 121 | 4×10^{-5} | 5.6×10^{-4} |
| S-35 | 126 | 6×10^{-5} | 4.6×10^{-4} |
| Ca-45 | 221 | 9×10^{-6} | 2.6×10^{-1} |
| Mn-54 | 401 | 1×10^{-4} | 4.2×10^{-1} |
| Na-22 | 1370 | 4×10^{-5} | 8.9×10^{-1} |
| H-3 | 6428 | 3×10^{-3} | 9.0 |

These calculations assume:

- a. That the radial dimensions of the 500-GeV accelerator are identical with the CPS.
- b. Equilibrium spectra similar to those measured in the CPS shield will be produced.

- c. The chemical composition and water content of the earth shield is similar to that at the CPS.

5. WATER TRANSPORT, DILUTION, AND CHEMICAL SORPTION

We assume that rainfall absorbed into the earth moves vertically downward past the accelerator to the water table. An estimate of the residence time of water flowing through regions of high flux density may be obtained from the size of the activation zone and the rate of inflow of water from rainfall. Estimates so obtained indicate water residence times of ~ 2000 days, but hydrological studies at accelerator sites will be needed to establish reliable values.

After leaving the activation zone water will continue to move downward to the water table and then be transported horizontally to the edge of the accelerator site. Radioactive decay subsequent to production is usually of importance only for shorter-lived nuclides. Thus, for example, in a journey of 7 days Be-7 will have decayed by a negligible amount. On the other hand, for short-lived nuclides, e.g., C-11, O-15, and N-13, the decay factor is very large (approx. 10^{15}), making them of no account.

If we assume that the activity produced in the activation zone is released steadily during the residence time of water in the activation zone then using Eq. (1) the rate of release of activity at the perimeter of the accelerator site, R , is given by:

$$R = \frac{\epsilon Q_0 (1 - e^{-T/t}) e^{-t/\tau}}{T} \text{ Ci/day} \quad (21)$$

Thus if the quantity of water pumped from the site is $P \text{ cm}^3/\text{day}$ using Eq. (3) the specific activity of water reaching the water supply in MPC is given by:

$$S_{\text{MPC}} = \frac{10^6}{P} \sum_i \frac{\epsilon_i Q_i (1 - e^{-T/\tau_i}) e^{-t/\tau_i}}{M_i T} \quad (22)$$

with Q_i in curies
 M_i in $\mu\text{c}/\text{cm}^3$
 $T, t,$ and τ_i in days
 P cm^3/day

Figure 1 shows the concentration calculated as a function of residence time, if it is assumed that all the activity produced in earth and water can be released directly to the ground water. A value of 10^{10} ml/day was used for the total volume of water abstracted from the accelerator site area and a transport time in the water table of 7 days

assumed. Radionuclide production data was taken from Table V.

Inspection of Fig. 1 shows that the specific activity of the water never exceeds 0.03 MPC and decreases only very slowly with residence time up to ~ 1000 days.

Chemical sorption will prevent several of the nuclides from appearing in the ground water and considerably reduce the MPC actually observed.

Mawson (24) has recently reviewed experience of the movement of radioactive wastes buried in the ground. He says, "with few exceptions, adsorption and exchange processes occur between the radionuclides and constituents of the soil." He concludes his review by saying, "The burial of radioactive waste is usually a very safe operation. If the site is selected with care any radionuclides that enter the soil will progress quite slowly down to the water table. Once in the ground water they will move faster, but still at a rate of one to several orders of magnitude less than the rate of movement of the ground water. These statements apply to most cations--many anions move at about the same speed as the ground water."

Be-7 has been recognized as being produced in a form very strongly absorbed onto surfaces in carrier free solutions (25). Similarly, studies by Blythe (26) show that P-32 is predominantly fixed in chalk soils in the form of calcium phosphate. Borak et al (21) report that of the long-lived radionuclides produced in glacial till, only H-3, Na-22, Ca-45, and Nm-54 were found in leach water, while Thomas (8) has reported the solubility of gross γ -activity produced in earth as $<1\%$. Detailed sorption studies may be necessary for particular accelerator sites, but available information suggests many accelerator nuclides will be strongly retained. Tritium may pass freely into the ground water.

6. CONCLUSIONS

General consideration of the total quantity of radionuclides produced in the earth shield of high energy accelerators suggest no serious contamination problems in volumes of water comparable with rainfall on the site. More precise estimates indicate maximum specific activities in ground water of $\sim 3 \times 10^{-2}$ MPC if all long-lived nuclides are released. However, chemical sorption plays an extremely important role in limiting the release of radionuclides to the general environment and if only tritium is mobile levels of $\sim 10^{-5}$ MPC are estimated.

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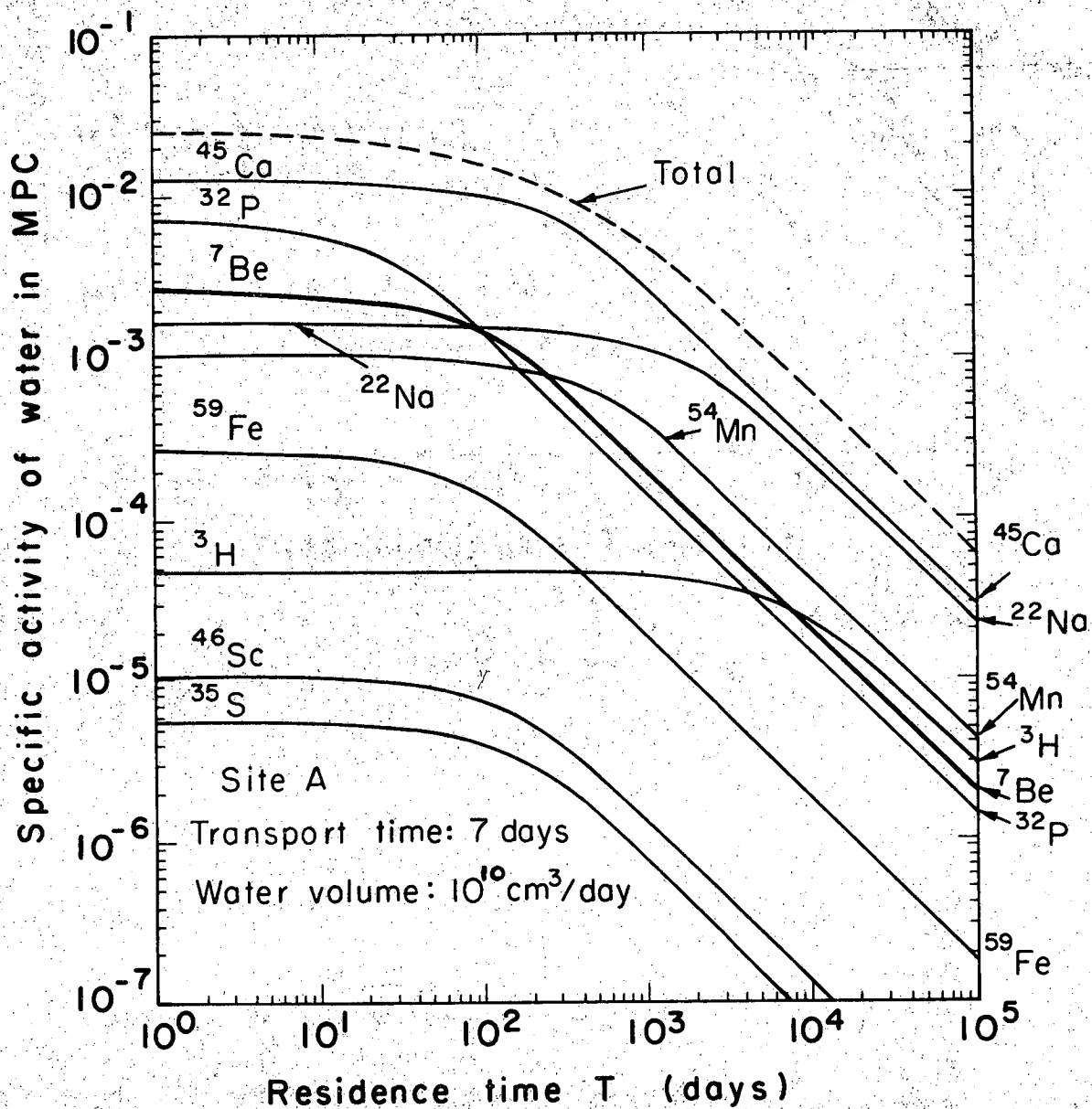
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FIGURE CAPTIONS

Figure 1. Specific activity of ground water at accelerator site perimeter.



XBL7010-4056

Figure 1

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