RADIOACTIVE GROUND WATER PRODUCED IN THE VICINITY OF BEAM DUMPS

SUMMARY

In this note we estimate the induced activity in the ground water near the 12° End Station A beam dump and we discuss the possibility of contamination of the public water supply. The results apply to other beam dumps as well.

Certain assumptions regarding ground water hydrology for this area are made, 5,6 the main one being that the ground water flows at less than 1 ft./day as it travels from the accelerator to San Francisquito Creek. A radical change in these assumptions could effect the calculation considerably. Consequently, an attempt is made to isolate the nuclear physics from the hydrology.

It is assumed that the electron-photon shower is completely attenuated within the dump complex itself, and that the water activity is caused by secondary particles (n, p, π , etc.) emanating from the shower core. Both capture and spallation reactions with the ground water and its impurities are considered, although activation of the earth is not.

The conclusion reached is that the concentration of radioactive nuclides that reaches the public water supply is well below the standards adopted by SLAC, 17 the State of California, 1 and the National Bureau of Standards. 2

Care must be taken, however, to insure that no drainage pipes intercept the channel of ground water flow. If such is the case, the isotopes with short half-lives will have to be re-considered.

I. INTRODUCTION

The ground water in the vicinity of the beam dumps will become somewhat radioactive. The purpose of this calculation is to estimate this activity and to discuss the means by which it might get to the general public.

It is assumed that the electron beam and its subsequent cascade shower will be sufficiently attenuated in the water-copper dump, and that the induced activity in the ground and external water will only be due to neutrons and other secondary particles $(p, \pi, etc.)$.

The 12° dump located upstream from End Station A has been used for this calculation; the general dimensions of the "mousehole" in which the dump is positioned are shown in Fig. 1.

A. HYDROLOGY

A few interesting opinions and facts concerning the hydrology of this area have been related to me by Professor Stanley Davis of the Stanford University Geology Department and by members of the U.S. Geological Survey in Menlo Park:

- 1. The ground water flow in the vicinity of the accelerator is rather slow and it would probably take 3 years⁶ to 10 years⁵ (at minimum) for any radioactivity to reach the creek.
- 2. There will certainly be dilution at the creek itself. 5
- 3. However, not much more than 50% dilution will occur in the ground water as it travels from the accelerator to the creek.⁵
- 4. The water table probably lies between 20 and 40 feet below the surface and generally follows the contours of the hills.^{5,6}
- 5. The minimum flow-rate of San Francisquito Creek, in the vicinity of the accelerator, is about 0.1 ft³/sec.⁶
- 6. By constructing a well near the beam dump, the water flow could be effectively confined and monitored.⁵,6

B. HEALTH PHYSICS

The "maximum permissible concentration," hereafter called MPC, for various isotopes in both air and water are tabulated by the NBS, 2 the State Department of Health, 1 and the Navy Department. 7 It should be pointed out, however, that the 168 hour/week (non-occupational) MPC values listed by the State of California and the Navy are a factor of 10 smaller than those listed by the NBS which are for occupational exposure. The SLAC tolerance 17 for the 168 hour/week (non-occupational) is 5 times smaller than the state. The State of California MPC values are, for the purposes of this calculation, sufficient.

C. NUCLEAR PHYSICS

One approach to the nuclear physics is to make the following assumptions:

- 1. The low-energy component is made up of giant resonance nuctrons; however, the high-energy flux consists of all secondary nucleons and mesons, other than the giant resonance spectrum, with energies $0 < E \le E_0$ (beam).
- 2. The high-energy flux can cause spallation of the water and impurities, leading to tritium and other radioactive products.
- 3. The giant resonance neutron flux, however, can only lead to a radioactive daughter through a <u>capture reaction</u> after the neutron is thermalized.

Two facts should be mentioned. First, the giant resonance spectrum essentially peaks at \sim 4 MeV and falls off at zero and 12 MeV (see Ref. 13-17 in SIAC-TN-65-11). Furthermore, the threshold energy (Laboratory) for the reaction n + 0¹⁶ \rightarrow H³ + N¹⁴ is 15.37 MeV.

Other (n,t) reactions have comparable thresholds. This suggests that the low-energy flux does not significantly produce tritium by means of spallation, and hence, assumption 3 above seems reasonable.

II. CALCULATIONS

- A. TRITIUM PRODUCTION BY THERMAL NEUTRON CAPTURE ON DEUTERIUM Assumptions:
 - 1. The majority of the neutrons come from the excitation of the giant resonance, and are distributed isotropically in the laboratory.
 - 2. No attenuation or build-up of these neutrons occurs in the dump or concrete.
 - 3. All of these neutrons are thermal when they arrive at the outside of the concrete, and the flux remains constant thereafter.
 - 4. The ground contains 15% water by weight (which may be somewhat high).*

Then, the flux just outside the concrete is:

$$\Phi = \frac{Q}{4\pi r^2} = 4.28 \times 10^9 \text{n/cm}^2 \text{-sec per MW}$$

where we have taken r = 5 feet = 152.4 cm (see Fig. 1) and⁸

$$Q = \frac{0.2 \text{ neutrons}}{\text{BeV(absorbed)}} \times \frac{6.25 \times 10^{15} \text{ BeV/sec}}{1 \text{ MW}}$$
$$= 1.25 \times 10^{15} \text{ neutrons/sec per MW}$$

The saturation activity in the ground water (15%) is:

$$R = \Phi \left(\frac{n}{\text{cm}^2 - \text{sec}}\right) \sigma \left(\frac{\text{cm}^2}{\text{D atom}}\right) n \left(\frac{\text{\# of D atoms}}{\text{ml of H O}}\right) \times \frac{1 \text{ curie}}{3.7 \times 10^{10} \text{ d/sec}} \times 15\%$$

We will use:

$$\sigma_{\rm D}$$
 = 0.5 mb % abundance of D = 0.015%

Whether to use weight % or mol % is not important here. Certainly 100% water (wt. or mol) would be the extreme case---a factor of 7 from that used above!

Then

$$n = \frac{2 \text{ atoms of D}}{D_2 0 \text{ molecule}} \times \frac{1.5 \times 10^{-4} \text{ D}_2 0 \text{ molecules}}{H_2 0 \text{ molecules}} \times \frac{6.02 \times 10^{23} \text{ (molecules of H}_2 0 / \text{mole)}}{18 (\text{gms/mole})}$$

$$\times \frac{1 \text{ gm}}{\text{ml}} = 1.00 \times 10^{19} \text{ atoms of D/ml}$$

and

$$R = \frac{4.28 \times 10^{9} \times 5 \times 10^{-28} \times 1.00 \times 10^{19}}{3.7 \times 10^{10}} \times 0.15 = 8.7 \times 10^{-11} \text{curies/ml per MW}$$

$$\simeq 10^{-4} \ \mu\text{Ci/ml per MW}$$

which is below the maximum permissible concentration of $3\times10^{-3}~\mu\text{Ci/ml.}^1$ Hence, because of the rather conservative assumptions, we will neglect this contribution.

B. ACTIVATION OF THE IMPURITIES IN THE GROUND-WATER FROM THERMAL NEUTRON CAPTURE

The impurities in the water near the beam end of the accelerator have been reported by Sokol. 9,10 He finds:

TABLE I

Impurity	η, ppm (by weight)
Ca	48
Mg	92 60
Na	60
HCO3	290
Cl	100
SO ₄ SiO ₂	240
	22
B	1.6
F	0.1

We will assume the following:

- 1. Φ (at r = 5 feet) = 4.28×10^9 n/cm²-sec per MW, previously calculated in A above with no build-up or attenuation.
- 2. $\lambda_{\rm attenuation}$ (concrete) = 6 inches³ and 1.5 feet of concrete (see Fig. 1), which gives an attenuation = $e^{-18/6} = 5 \times 10^{-2}$.
- 3. Build-up factor of 3. (3)
- 4. No attenuation of the flux in the earth itself.

Hence, we will take $\Phi = 4.28 \times 10^9 \times 5 \times 10^{-2} \times 3 = 6.4 \times 10^8$ n/cm²-sec per MW. The saturation activity is again given by

R (curies/ml) per MW =
$$\Phi \sigma n/3.7 \times 10^{10}$$

where

$$n \left(\frac{\text{atoms of impurity}}{\text{ml of H}_2 0} \right) = \frac{\eta(\text{ppm}) \times 10^{-6} \times 6.02 \times 10^{23} \times \text{ isotopic abundance}(\%) \times 10^{-2}}{A}$$

A = atomic weight of impurity in gms.

 $\sigma = cross section in cm²/atom$

or

$$R(\mu Ci/ml)$$
 per MW = 1.04 × 10⁻⁴ $\frac{\eta(ppm) \sigma(barns) Abd(\%)}{A (gms)}$

The concentrations are given in Table II along with the MPC values.

If we neglect all activities with half-lives shorter than 1 year, only the ${\rm Cl}^{36}$, ${\rm C}^{14}$ and ${\rm Be}^{10}$ concentrations show up, where we have assumed that it will take 10 years at minimum for the ground water to reach San Francisquito Creek. The other activities were included for completeness. The ${\rm C}^{14}$ activity is well below MPC and, although the MPC is not listed, the ${\rm Be}^{10}$ activity will never be near saturation due to the long (~ 10^6 year) half-life.

Now, the non-steady state activity (per ml of H_2O) is approximately

$$R(t) \simeq R \left[\frac{0.693 \ t}{\frac{T_1}{2}} \right]$$

where

R = saturation activity (per ml of H_2 0) t = irradiation time $T_{\frac{1}{2}}$ = half-life

It seems reasonable to assume that the ground water moves fast enough so that the irradiation time, t, is \lesssim one year. Take, for ${\rm Cl}^{36}$

t = 1 year

$$T_{\frac{1}{2}} = 3.2 \times 10^5 \text{ year}$$

R = $6.7 \times 10^{-1} \mu \text{Ci/ml}$

TABLE II

CONCENTRATION OF RADIOACTIVE IMPURITIES IN THE GROUND-WATER FORMED BY THERMAL NEUTRON CAPTURE*

Impurity	η(ppm) ⁹ ,10	A(gms)	Reaction, Decay, etc.	Half-Life of Unstable Daughter	oact(barns)16	Isotope Abundance(%) ¹⁶	R(μCi/ml) (Saturation)	MPC(μCi/ml) ¹
Boron	1.6	10.81	$B^{10}(n,p)Be^{10} \rightarrow (\beta^-)B^{10}(stable)$	$2.5 \times 10^6 y$	< 0.2	20	6.3×10^{-5}	NL
and the second second			$B^{11}(n,\gamma)B^{12} \rightarrow (\beta^-,\gamma)C^{12}(\text{stable})$	0.022s	0.050	20	6.3×10^{-5}	NL
Carbon	57.1 (calc.)	12.01	$C^{13}(n,\gamma)C^{14} \rightarrow (\beta^-)N^{14}(\text{stable})$	$5.6 \times 10^{3} \text{y}$	9 × 10 ⁻⁴	1.11	5.0×10^{-7}	8 × 10 ⁻⁴
Fluorine	0.1	19.00	$F^{19}(n,\gamma)F^{20} \rightarrow (\beta^-,\gamma)Ne^{20}(\text{stable})$	10.7s	9 × 10 ⁻³	100	5.0 × 10 ⁻⁷	NL
Scaium	60 .	22.99	$Na^{23}(n,\gamma)Na^{24} \rightarrow (\beta^{-},\gamma)Mg^{24}(stable)$	15 h	0.536	100	1.5 × 10 ⁻²	2 × 10 ⁻⁴
Magnesium	92	24.31	$Mg^{26}(n,\gamma)Mg^{27} \rightarrow (\beta^{-},\gamma)Al^{27}(stable)$	9.45m	0.027	11.29	1.2×10^{-4}	NL
Silicon	10.3 (calc.)	28.09	$Si^{30}(n,\gamma)Si^{31} \rightarrow (\beta^-,\gamma)P^{31}(stable)$	2.65h	0.110	3.05	1.3 × 10 ⁻⁵	9 × 10 ⁻⁴
Sulfur	80.1 (calc.)	32.06	$S^{33}(n,p)P^{33} \rightarrow (\beta^{-})S^{33}(stable)$	24.4d	0.015	0.75	3.0 × 10 ⁻⁶	NL
			$S^{34}(n,\gamma)S^{35} \rightarrow (\beta^{-})Cl^{35}(stable)$	87.1d	0.26	4.215	2.9 × 10 ⁻⁴	6 × 10 ⁻⁵
			$S^{36}(n,\gamma)S^{37} \rightarrow (\beta^-,\gamma)Cl^{37}(stable)$	5.04m	0.14	0.017	6.3 × 10 ⁻⁷	NL
Chlorine	100	35 -45	$Cl^{35}(n,\gamma)Cl^{36} \rightarrow (\beta^-,\gamma)Ar^{36}(stable)$	$3.2 \times 10^{5} \text{y}$	30	75.4	6.7 × 10 ⁻¹	8 × 10 ⁻⁵
			$Cl^{35}(n,p)S^{35} \rightarrow (\beta^*)Cl^{35}(stable)$	87.1d	0.19	75.4	4.2×10^{-3}	6 × 10 ⁻⁵
			$Cl^{35}(n,\alpha)P^{32} \rightarrow (\beta^{-})S^{32}(\text{stable})$	14.3d	< 5 × 10 ⁻⁵	75.4	1.1 × 10 ⁻⁶	2 × 10 ⁻⁵
	·		$Cl^{37}(n,\gamma)Cl^{38} \rightarrow (\beta^-,\gamma)Ar^{38}(stable)$	37.5m	0.56	24.6	4.1×10^{-3}	4×10^{-4}
Calcium	48	40.08	$Ca^{44}(n,\gamma)Ca^{45} \rightarrow (\beta^{-})Sc^{45}(stable)$	164 d	0.67	2.06	1.8 × 10 ⁻⁴	9 × 10 ⁻⁶
			$Ca^{46}(n,\gamma)Ca^{47} \rightarrow (\beta^-,\gamma)Sc^{47}$	4.9 d	0.25	3.3×10^{-3}	1.0×10^{-7}	5 × 10 ⁻⁵
			$Sc^{47} \rightarrow (\beta^{-}, \gamma) Ti^{47} (stable)$	3.43a				9 × 10 ⁻⁵

NOTE: NL means not listed

^{*}Per MW of beam power.

Then,

$$R(t) \simeq 1.5 \times 10^{-6} \mu Ci/ml$$

which is below the MPC of 8×10^{-5} and hence, we will neglect ${\rm Cl}^{36}$ also.

TRITIUM PRODUCTION FROM HIGH-ENERGY NUCLEAR REACTIONS WITH OXYGEN 16 Following a calculation by DeStaebler, 11 the saturation activity for tritium production by high-energy secondary particles is:

S.A.(Ci/MW) = saturation activity =
$$\frac{E_{o}}{E_{m}} \left(\frac{particles}{sec-MW} \right) \times \frac{\sigma}{\sigma_{o}} \left(\frac{fraction of interaction of interaction section in the second of the se$$

$$\times \frac{0.15}{3.7 \times 10^{10}}$$

where

F = fraction of incident energy which goes into high energy nuclear particles 12 = 0.2%

 $E_0I = incident power = 6.25 \times 10^{18} MeV/sec-MW$

 E_m = average energy of particles = 100 MeV

 σ = cross section for H³ production from 0¹⁶ = 35 mb (13)

 $\sigma_{\rm o}$ = total removal cross section = 291 mb (for $\lambda_{\rm o}$ = 137 g-cm⁻² and using $A_{ave}(earth) = 24$ as calculated from Ref. 15).

and where 0.15 comes from the assumption of 15% water by weight.

Hence, S.A. = 61 Ci/MW. The activity in one year is:

activity
$$\simeq$$
 S.A. $\left(\frac{0.693 \text{ t}}{T_{\frac{1}{2}}}\right) = 61 \times \frac{0.693 \times 1}{12.3} = 3.4 \text{ Ci/year-MW}$

Now, the minimum creek flow in San Francisquito Creek is $\sim 0.1 \text{ ft}^3/\text{sec}$ or 8.9×10^{10} ml/year.⁶ Furthermore, Section 30269 of reference 1 states that concentrations may be averaged over periods not greater than one year. If we average over one year we get:

$$R \simeq 4 \times 10^{-5} \mu \text{Ci/ml-MW}$$

which is below the MPC of $3 \times 10^{-3} \, \mu \text{Ci/ml}$.

Furthermore, we haven't taken into account:

- 1. Radioactive decay as the water moves from the dump to the creek (a period of about 10 years⁵,⁶).
- 2. Further dilution in traveling to the creek ($\sim 50\%$ at most⁵).
- 3. The average creek flow certainly must be greater than the minimum of 0.1 ft³/sec.

All of these will make R even smaller.

Hence, it appears as though tritium will not be too important by this mechanism either.

D. ACTIVATION OF THE WATER IMPURITIES BY HIGH-ENERGY REACTIONS Table II suggests that we only need to consider those radioactive products with A < 50. Furthermore, assuming that the ground water flow is slow, we can make a restriction on the half-life:

122 days
$$(\frac{1}{3} \text{ year}) \leq T_{\frac{1}{2}} \leq 100 \text{ years}$$

where the upper-limit is chosen under the assumption that saturation conditions won't be realized for isotopes with very long half-lives.

By carefully examining the nuclide chart with these restrictions, we are left with:

$$H^3$$
 (12.3 years) $\rightarrow \beta^-$
 V^{49} (330 days) \rightarrow electron capture
 Na^{22} (2.6 years) $\rightarrow \beta^+$, γ
 Ar^{42} (3.5 years) $\rightarrow \beta^-$
 Ca^{45} (164 days) $\rightarrow \beta^-$

Now, tritium has already been dealt with, and $\,{\mbox{V}}^{49}\,$ does not give off any radiation.

Also, $_{11}$ Na 22 , $_{18}$ Ar 42 and $_{20}$ Ca 45 must be created by bombarding isotopes having Z \geq 11, 18, and 20, respectively. By searching Ref. 13, keeping in mind that the only elements to consider are listed in Table I, we find only one cross section:

$$Na^{23}$$
 (p,pn) Na^{22}
 σ = 31 mb for E_p = 5.7 BeV

Using this cross section, 60 ppm by weight (Table I), and calculating in a manner similar to that done in Part C above, it is easy to show that the concentration in the creek, averaged over one year, is approximately:

$$R \simeq 1 \times 10^{-8} \, \mu \text{Ci/ml}$$

which is small indeed! (The MPC value is $4 \times 10^{-5} \, \mu \text{Ci/ml}$ for Na^{22} .)

I don't believe that high-energy spallation reactions with the water impurities will give significant concentration. If cross sections larger than 30 mb do exist, they should be quite evident. Even so, it would take an extremely large cross section in order to make these concentrations significant. This is because of the low ppm values of elements in the water.

III. CONCLUSIONS

If the main ground water assumption prevails (i.e., a very slow flow-rate) only small amounts of radioactive ground water will get into the public water supply. Should this assumption be wrong, we might have a more serious problem involving activities with relatively short half-lives. The only way I can imagine this happening is if a drainage pipe intercepted the ground water discharge channel to the creek.

One question is still unanswered. What about the nuclear reaction products made in the dirt which gets into the water? I don't believe that this will be important for the following reasons:

- 1. There's a sort of equilibrium between what's made in dirt getting into water and vice versa.
- 2. A spectrographic analysis of earth samples taken from the BSY area and just recently made available to me by the Hazelton Nuclear Science Corporation indicates that the ground does not contain appreciable amounts of high Z material. Thus, the argument about the few medium and low Z activities with 1/3 year $\leq T_{\frac{1}{2}} \leq 100$ years is still valid.

It should be mentioned that there is one method of insuring that no activated ground water ever reaches San Francisquito Creek; that is, to drill a sump near the creek-side of each dump or "hot-spot," thus

capturing the flow. If the water is monitored and found safe, it need not be pumped out. The flow would then resume its original path to San Francisquito Creek. If it is found to be quite active, it can be stored or possibly diluted and dumped into the sewer.

Several ground water calculations and experiments associated with underground nuclear blasts have been made. 18-21 Although some of the radioactive products found are identical with those found here, a worthwhile comparison seems fruitless at this time. For one thing, the neutron spectrum from nuclear detonations is either fission or fusion (depending on the bomb), and we must consider the high energy flux in addition. Also, it appears as though the nuclear blast itself tends to fuse the silican into glass, which hinders the flow of activated water.

Finally, a map of the local area has been included for convenience (Fig. 2).

ACKNOWLEDGMENT

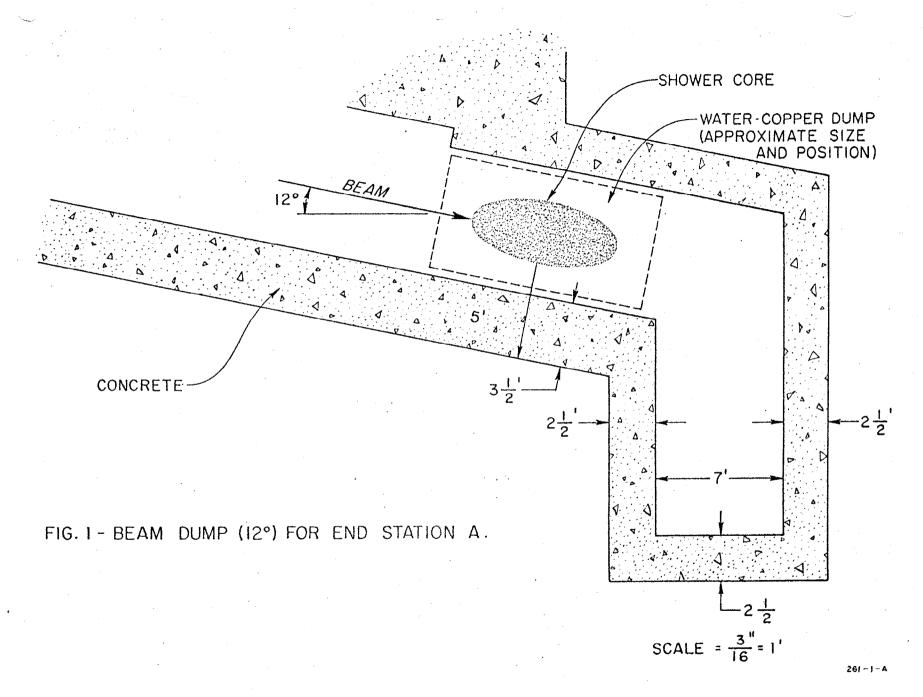
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(TRACED FROM DRAWING NO. D-505-311 SHEET NO. 5-11



Fig. 2