8.1 Introduction

Chapter 7 provided the tools needed to address the subject of induced radioactivity. In this chapter, the discussion of induced radioactivity at accelerators is continued to address their production and production and propagation in environmental media such as air, soil, rock, and water. Aspects pertinent to both occupational safety and environmental protection are covered. The chapter includes introductory material connecting meteorology and hydrogeology with such radioactivity.

8.2 Airborne Radioactivity

8.2.1 Production

Thomas and Stevenson have presented a very useful synopsis, largely followed here, of the production of radioactivity in air (Th88). This discussion was reprised by Swanson and Thomas (Sw90). The principal source of radioactivity in air at accelerators is due to the interaction of primary and secondary particles directly with the constituent target nuclei in the air in accelerator enclosures. Activated dust and gaseous emission from activated liquids are of much less importance. Table 8.1 gives the abundances and number densities of atoms of the most common stable isotopes found in the atmosphere both by volume percentage and in terms of the atomic density, N_j.

Table 8.1 Abundances of the Most Common Stable Nuclides in the Atmosphere

Isotope	Percentage by volume in the atmosphere (atoms)	N_j (atoms cm^{-3}) at room temperature
^{14}N	78.16	4.199×10^{19}
^{16}O	20.00	1.075×10^{19}
^{40}Ar	0.467	1.558×10^{17}
^{15}N	0.290	2.149×10^{16}
^{18}O	0.040	1.255×10^{17}

Patterson and Thomas (Pa73), have expanded general activation equation, Eq. (7.8), to derive the total specific activity, S (typically in units of Bq cm^{-3}) of an enclosed volume of radioactive air;

$$S = C\sum_i \left[\sum_j \phi_\gamma N_j \bar{\sigma}_{ij\gamma} + \sum_j \phi_{th} N_j \bar{\sigma}_{ij_{th}} + \sum_j \phi_{HE} N_j \bar{\sigma}_{ij_{HE}} \right] [1-\exp(-\lambda_i t_i)] \exp(-\lambda_i t_c), \tag{8.1}$$

where ϕ_γ, ϕ_{th}, and ϕ_{HE}, represent the average photon, thermal neutron and high energy flux densities. As before, in this equation t_i is the irradiation time while t_c represents the decay time. The $\bar{\sigma}_{ij}$ values are the corresponding cross sections averaged with the energy-dependent flux density over energy,

$$\bar{\sigma}_{ij} = \int_{E_{\min}}^{E_{\max}} dE \sigma_{ij_k}(E)\phi_k(E) \Big/ \int_{E_{\min}}^{E_{\max}} dE \phi_k(E), \tag{8.2}$$

where the limits of integration correspond to the three ranges in the summation. The constant, *C*, is the conversion to specific activity and is equal to unity for activity in Becquerels cm^{-3} if all the units of length implicit in the quantities in Eq. (8.1) are expressed in cm. The outer sum over index *i* includes all possible radionuclides produced and the sum over the index *j* sums over the parent atoms found in air. The flux densities are, without further information, the average over some relevant spatial volume.

Table 8.2 lists the radionuclides that can be produced from the principle constituents in air along with the reaction mechanisms associated with their production and an estimate of the average production cross section. The large cross sections for neutron-induced (n, γ) and (n, p) reactions are for captures of neutrons of thermal energies ($E_n \approx 0.025$ eV) while the remaining cross sections are generally the saturation cross sections found in the region above approximately a few 10's of MeV. The γ-induced reactions are present at virtually all accelerators and at most energies. The corresponding cross sections will, of course, be energy-dependent.

8.2.2 Accounting for Ventilation

Adjustments for the presence of ventilation can be quite conveniently made by for a given radionuclide by using an effective decay constant, λ', that includes the physical decay constant, λ, along with a ventilation term, *r*,

$$\lambda' = \lambda + r, \tag{8.3}$$

where $$r = \frac{D}{V},$$

with *D* being the ventilation rate in air volume per unit time and *V* being the enclosure volume. Thus *r* is the number of air changes per unit time. The applicable differential equation with ventilation included is, then,

$$\frac{dn'}{dt} = -\lambda' n'(t) + N\sigma\phi. \tag{8.4}$$

After an irradiation time, t_i, with no initial activation, the solution is

$$n'(t_i) = \frac{N\sigma\phi}{\lambda + r}\left\{1 - \exp\left[-(\lambda + r)t_i\right]\right\}. \tag{8.5}$$

So the specific activity is

$$a'(t_i) = \lambda n'(t_i) = \frac{\lambda N \sigma \phi}{\lambda + r}\left\{1 - \exp\left[-(\lambda + r)t_i\right]\right\}. \tag{8.6}$$

Table 8.2 Radionuclides with half-life > 1 minute that can be produced in air at accelerators. [Adapted from (Sw90).]

Radionuclide	Half-life	Emission	Parent Element	Production Mechanism	High Energy Cross Section (mb)
^{3}H	12.3 years	β^-	N	Spallation	30
			O	Spallation	30
^{7}Be	53.3 days	γ, elect. capt.	N	Spallation	10
			O	Spallation	5
			Ar	Spallation	0.6
^{11}C	20.4 minutes	β^+	N	Spallation	10
			O	Spallation	0.7
			Ar	Spallation	0.7
^{14}C	5730 years	β^-	N	(n,p)	1640[a]
^{13}N	9.96 minutes	β^+	N	Spallation	10
			N	(γ,n)	10
			O	Spallation	9
			Ar	Spallation	0.8
^{14}O	70.6 seconds	β^+,γ	O	Spallation	1
			Ar	Spallation	0.06
^{15}O	2.03 minutes	β^+	O	Spallation	40
			O	(γ,n)	10
			Ar	Spallation	
^{18}F	1.83 hours	β^+, elect. capt.	Ar	Spallation	6
^{24}Ne	3.4 minutes	β^-,γ	Ar	Spallation	0.12
^{22}Na	2.6 years	β^+,γ	Ar	Spallation	10
^{24}Na	15.0 hours	β^-	Ar	Spallation	7
^{27}Mg	9.46 minutes	β^-,γ	Ar	Spallation	2.5
^{28}Mg	20.9 hours	β^-,γ	Ar	Spallation	0.4
^{28}Al	2.25 minutes	β^-,γ	Ar	Spallation	13
^{29}Al	6.6 minutes	β^-,γ	Ar	Spallation	4
^{31}Si	2.62 hours	β^-,γ	Ar	Spallation	6
^{30}P	2.50 minutes	β^+,γ	Ar	Spallation	4.4
^{32}P	14.3 days	β^-	Ar	Spallation	25
^{33}P	25.3 days	β^-	Ar	Spallation	9
^{35}S	87.5 days	β^-	Ar	Spallation	23
^{34m}Cl	32.0 minutes	β^-,γ	Ar	Spallation	0.7
^{38}Cl	37.2 minutes	β^-,γ	Ar	(γ,pn)	4
^{39}Cl	55 minutes	β^-,γ	Ar	(γ,p)	7
^{41}Ar	1.8 hours	β^-,γ	Ar	(n,γ)	660[a]

[a] Thermal neutron capture cross section, see text.

But $N\sigma\phi$ is just the saturation concentration, a_{sat}, without mixing [see Eq. (7.7)]. Hence, with mixing the saturation concentration, a'_{sat} is

$$a'_{sat} = \frac{\lambda\, a_{sat}}{\lambda + r}. \tag{8.7}$$

Since low energy accelerators contain their beams in continuous vacuum systems, the activation of air at these machines is greatly minimized. At high energy accelerators, it is quite common to have air gaps at certain "interface points" and devices associated with beam targetry or beamline diagnostic instrumentation render continuous vacuum impractical. These "air gaps" are only characteristic of extracted beam lines while the beam in the accelerator proper is, of necessity, contained in continuous vacuum. In addition, the large multiplicity of secondary particles produced as a part of cascade processes, either electromagnetic or hadronic, can produce airborne radioactivity even where the beams themselves are contained in vacuum.

If the accelerator enclosures were completely sealed, there would be no releases to the outside world and the hazard of these airborne radionuclides would be entirely restricted to those who might have to enter the enclosures. This would, however, allow the longer-lived radionuclides to build up in accord with Eq. (8.1). Also, ventilation is generally needed to provide cooling of components and fresh breathing air for workers. Typically, the "residence time" of air in accelerator enclosures is limited to a range of time between approximately 30 minutes and one hour. Thus, the airborne radionuclides in the accelerator environment, in equilibrium, will have half-lives only up to the order of one hour. The residence time of the air in conjunction with the cross sections determines the radionuclides of importance.

8.2.3   Propagation of Radionuclides in the Environment

The other consideration concerning airborne radioactivity is that associated with the dose to members of the general public when radioactive materials are released to the atmosphere external to the accelerator enclosure. The U. S. Environmental Protection Agency (EPA) has placed an annual limit of 10 mrem on dose equivalent to members of the general public due to the operations of DOE facilities, comparable to limits applied to other facilities, and has also placed stringent regulations on how such releases are to be measured (CFR89). An annual dose equivalent of such small value is usually difficult, or impossible, to measure at distant locations. Thus, the standard practice is to measure the activity released and then use calculational models to estimate the maximum dose equivalent that actual members of the public could receive. The regulations prescribe the specific computer codes that must be used to perform these calculations through the use of a Gaussian plume model that combines input data on the release of radioactivity with meteorological information. Such computer modeling will not be described in detail here. Examples of such plume models are given in textbooks and the results depend on details of the meteorological conditions. A short synopsis is given here.

8.2.3.1 Propagation of Radioactivity - Tall Stacks

Concentrations of radionuclides at distant locations can be estimated analytically using the so-called **Sutton’s equation** to be described shortly. Due to the nature of accelerator operations, only "steady-state" conditions are considered since transient accidental releases from accelerators are unlikely. A good description that applies to rather tall (> 25 m) release points has been presented by Cember (Ce69). Such release points are commonly called **stacks**. More details have been provided by Slade (Sl68). The dispersion is mainly characterized by dilution of the radionuclides and their eventual return to ground level breathing zones. The meteorological conditions of major importance to this topic are illustrated in Fig. 8.1. Relevant **stability classes** have been developed that describe the various possibilities. Descriptions of these stability classes are given in Table 8.3.

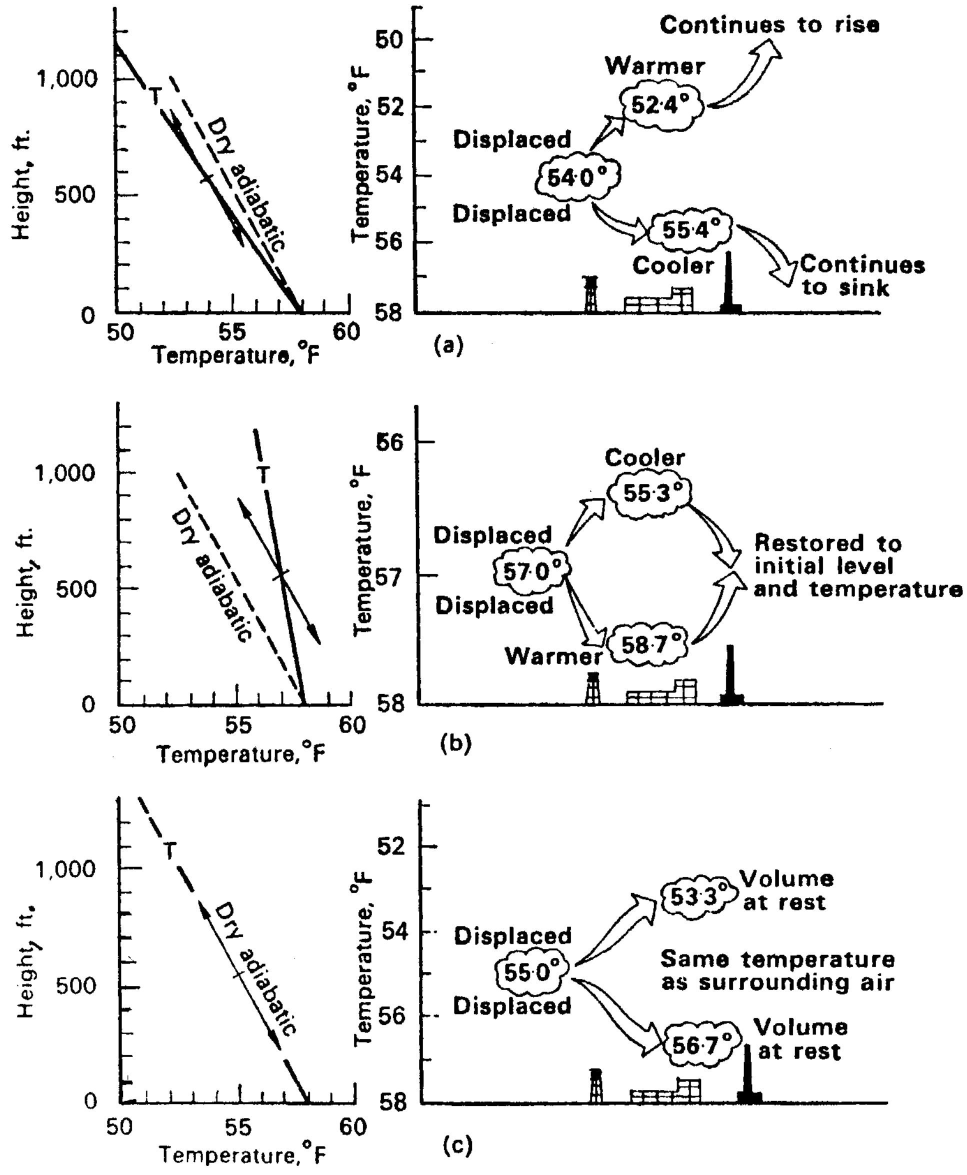

Fig. 8.1 Effect of atmospheric temperature gradient or lapse rate on a displaced volume of air for various conditions: **a)** Unstable lapse rate; **b)** Stable lapse rate; **c)** Neutral lapse rate [Reproduced from (Sl68).]

Table 8.3 Atmospheric stability classes for use with Sutton's equation [Adapted from (Sl68).]

stable:	No heat is gained or lost by a parcel of air that rises and expands adiabatically with falling temperature. The adiabatic cooling with rise normally corresponds to a gradient of 5.4 ^{o}F/1000 ft (1 ^{o}C/100 meters) for dry air and 3.5 ^{o}F/1000 ft (0.6 ^{o}C/100 meters) for moist air. If the atmospheric temperature gradient is less than adiabatic, but still negative, stability is achieved because a rising parcel cools faster than its surroundings and then tends to sink. A sinking parcel is warmer than its surroundings and thus is less dense and tends to rise. This restricts the width of the plume and consequently decreases dilution.
inversion:	If the temperature gradient is such that the temperature increases with height, then an inversion occurs. Rising effluent from a "stack" becomes much denser than its surroundings and thus sinks. The effluent is thus more limited in its ascent and this, too, serves to limit dilution.
superadiabatic:	If the rate of decrease of temperature with elevation is greater than that in adiabatic conditions, an unstable condition results which promotes the vertical dispersion, and hence dilution. A rising parcel does not cool fast enough due to its expansion and therefore remains warmer and continues to rise. Likewise, a falling parcel continues to fall.

Sutton's equation, as adapted here by Cember (Ce69) for consideration of short-lived radionuclides and for tall stacks, is:

$$\bar{c}(x,y) = \frac{2Q}{\pi C^2 \bar{u} x^{2-n}} \exp\left\{-\frac{\lambda}{\bar{u}}\sqrt{x^2+y^2}\right\} \exp\left\{-\frac{h^2+y^2}{C^2 x^{2-n}}\right\}, \qquad (8.8)$$

where the exponential involving the decay constant λ conservatively allows for radioactive decay in transit for a particular radionuclide and:

$\bar{c}(x,y)$ is the average concentration (activity per m^3),
Q is the emission rate of activity per second,
(x,y) are coordinates to the point of measurement from the foot of the stack (meters);
  x is along the centerline of the plume as determined by the wind direction (downwind), y is the transverse coordinate, and z is the vertical coordinate,
$\bar{u}$ is the mean wind speed, meters per second,
C is the virtual diffusion constant in lateral and vertical directions (see Table 8.4),
n is a dimensionless parameter related to the atmospheric conditions (Table 8.4),
h is the *effective* chimney height (if the gas has significant emission velocity)
  determined as follows from the actual chimney height h_a;

$$h = h_a + d\left(\frac{v}{\bar{u}}\right)^{1.4}\left(1+\frac{\Delta T}{T}\right). \qquad (8.9)$$

In Eq. (8.9) d is the outlet diameter in meters, v is the exit velocity of the gas (meters/sec), and $\Delta T/T$ is the difference between the temperature of the gas and the ambient outdoor temperature divided by the *absolute* temperature of the gas, T. Table 8.4 gives values for parameters to be used in Eq. (8.8).

Table 8.4 Diffusion (C^2) and Stability (n) parameters for Cember's version of Sutton's Equation, Eq. (8.8) [Adapted from (Ce69).]

Lapse Rate		**C^2 Effective Chimney Height, h, (meters)**			
	n	**25**	**50**	**75**	**100**
Superadiabatic	0.20	0.043	0.030	0.024	0.015
Stable	0.25	0.014	0.010	0.008	0.005
Moderate Inversion	0.33	0.006	0.004	0.003	0.002
Large Inversion	0.5	0.004	0.003	0.002	0.001

8.2.3.2 Propagation of Airborne Radioactivity - Short Stacks

The above representation of Sutton's equation is a useful one where tall stacks are involved. However, at typical accelerator facilities it is uncommon for stacks to be very tall. Again for purposes of this discussion, only steady state conditions continuous in time are treated here. For such calculations, the concentration as a function of coordinates (x,y,z), defined as for the tall stacks, is given by a somewhat different formulation of Sutton's equation which uses the same coordinate system;

$$\bar{c}(x,y,z)=\frac{Q}{2\pi\sigma_y\sigma_z\bar{u}}\left\{\exp\left[-\frac{\lambda}{\bar{u}}\sqrt{x^2+y^2}\right]\right\}\left\{\exp\left(-\frac{y^2}{2\sigma_y^2}\right)\right\}\left\{\exp\left[-\frac{(z-h)^2}{2\sigma_z^2}\right]+\exp\left[-\frac{(z+h)^2}{2\sigma_z^2}\right]\right\}. \tag{8.10}$$

For the common situation of interest where the receptor location of concern is at ground level ($z = 0$), this simplifies to

$$\bar{c}(x,y,0)=\frac{Q}{\pi\sigma_y\sigma_z\bar{u}}\left\{\exp\left[-\frac{\lambda}{\bar{u}}\sqrt{x^2+y^2}\right]\right\}\left\{\exp\left[-\left(\frac{y^2}{2\sigma_y^2}+\frac{h^2}{2\sigma_z^2}\right)\right]\right\}, \tag{8.11}$$

where the presence of the ground as a "barrier" to the flux is taken into account. In these equations, the quantity h is the elevation of the stack top above the ground in meters and $\sigma_y(x)$ and $\sigma_z(x)$ are the dispersion coefficients that have units of length (meters) and are implicitly functions of x. All other quantities are the same as given above for tall stacks. These variables are, of course, determined from the meteorological conditions.

Table 8.5 gives an alternative scheme for classifying the meteorological conditions. The classification may then be used with the curves in Figs. 8.2 and 8.3 to determine the values of σ_y and σ_z as a function of the coordinate x for use in Eqs. (8.10) and (8.11).

Table 8.5 Relation of turbulence types to weather conditions. [Adapted from (Sl68).]

A-Extremely unstable conditions	D-neutral conditions[a]
B-Moderately unstable conditions	E-Slightly stable conditions
C-Slightly unstable conditions	F-Moderately stable conditions

Surface Wind Speed (m/sec)	Daytime insolation			Nightime conditions	
	Strong	Moderate	Slight	Thin overcast or > 4/8 cloudiness[b]	< 3/8 cloudiness
<2	A	A-B	B		
2	A-B	B	C	E	F
4	B	B-C	C	D	E
6	C	C-D	D	D	D
>6	C	D	D	D	D

[a] Applicable to heavy overcast, day or night

[b] The degree of cloudiness is defined as that fraction of the sky above the local apparent horizon which is covered by clouds.

Airborne radioactivity releases can be minimized by:

- limiting the ventilation rates during operations when people are <u>not</u> present in the enclosure,
- delaying the actual emissions by requiring long pathways to the ventilation "stacks", and
- minimizing air gaps in the beam.

8.2.4 Radiation Protection Standards for Airborne Radioactivity

The airborne radioactivity is of primary concern to workers who might enter the enclosure to perform maintenance activities and thus are classified as "occupational workers". Since the principal radionuclides are of relative short half-life, the hazard is largely due to the "immersion" in a source of *external* dose rather than a gaseous ingestion hazard such as might be found in operations involving the processing of long-lived radioactive materials. The latter leads to long-term *internal* radiation exposures. Regulatory authorities, guided by recommendations of the International Commission on Radiation

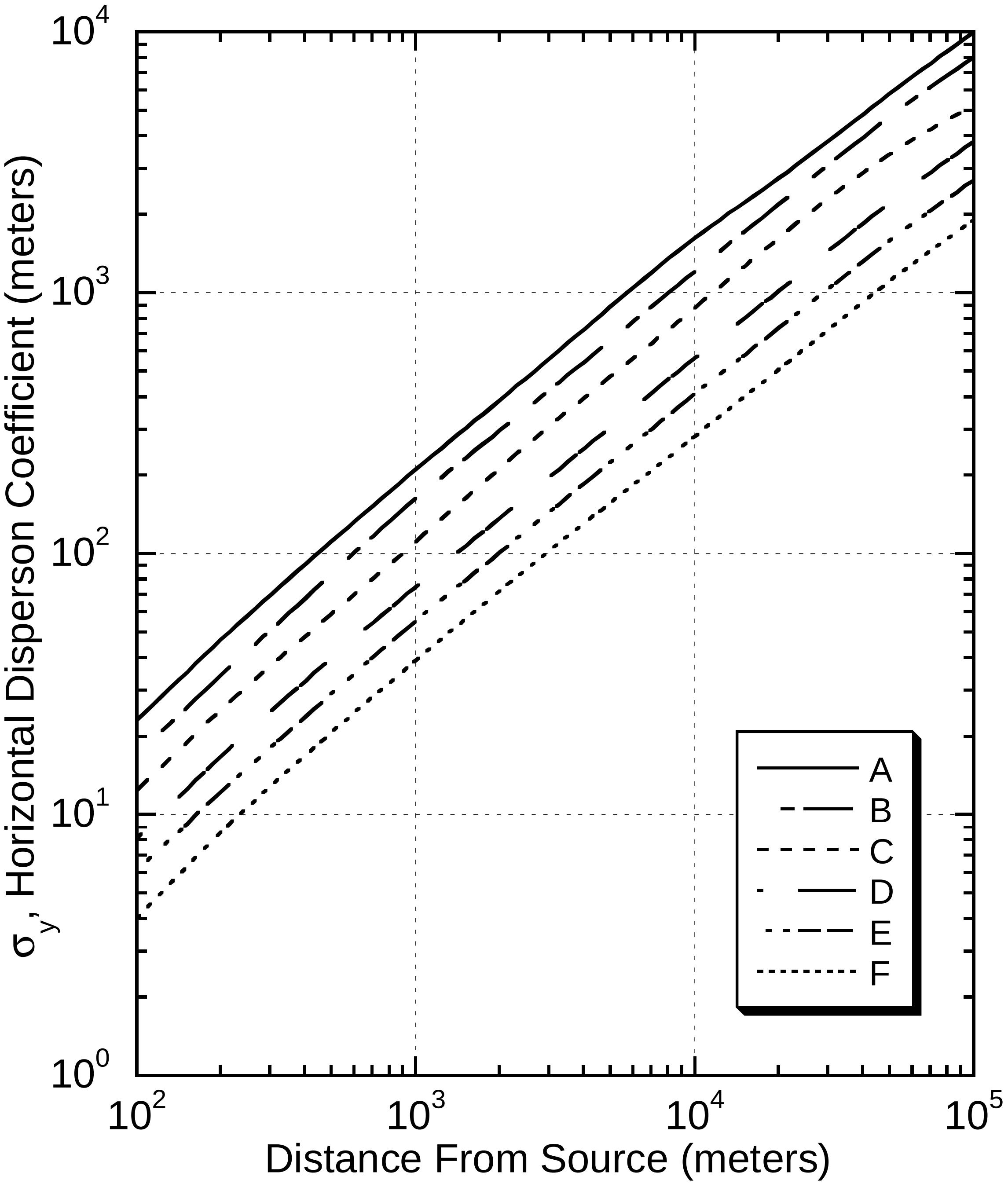

Fig. 8.2 Lateral diffusion, σ_y, as a function of downwind distance, x, source for turbulence types defined in Table 8.5. [Adapted from (Sl68).]

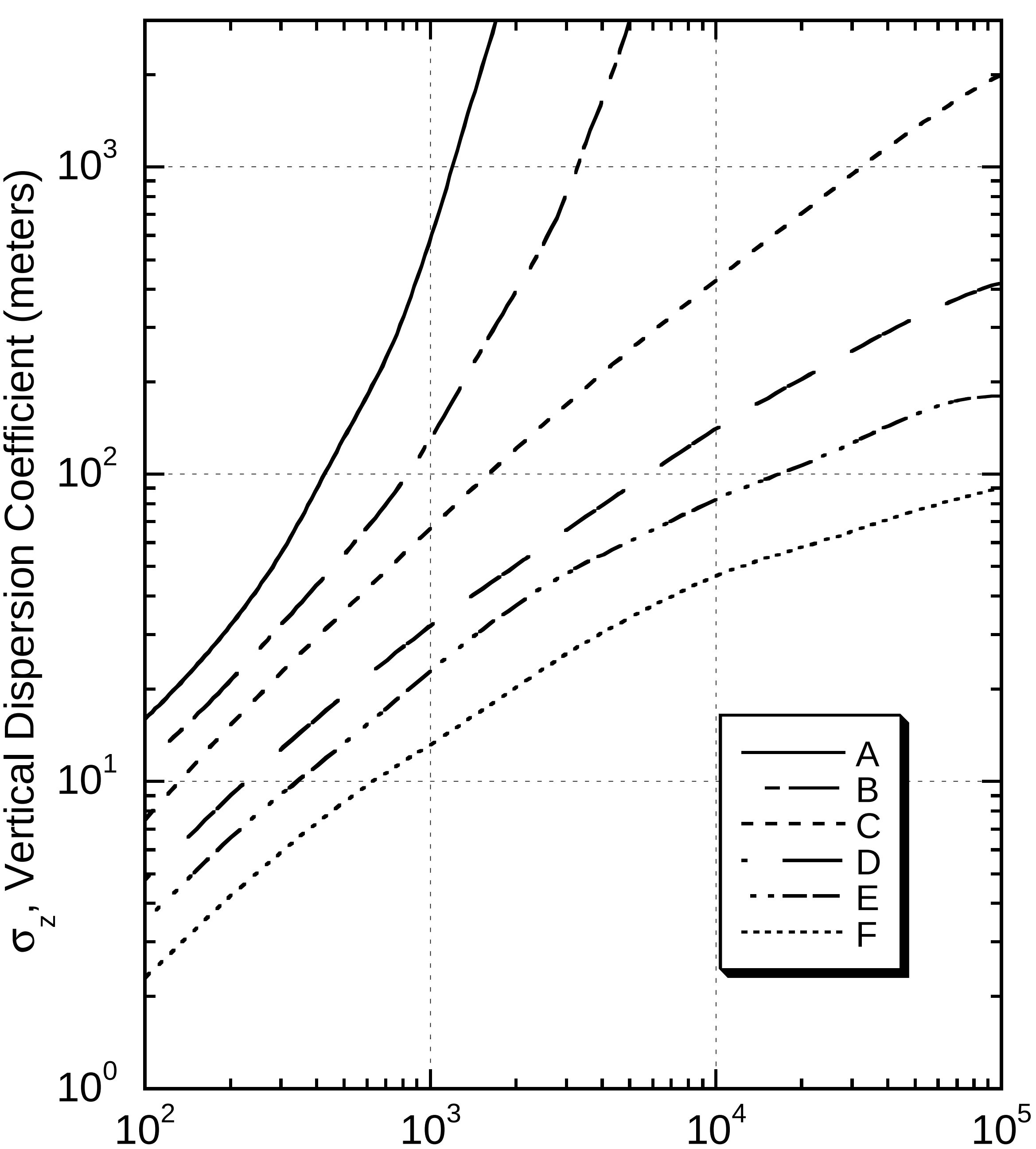

Fig. 8.3 Vertical diffusion, σ_Z, as a function of downwind distance, *x*, from source for turbulence defined in Table 8.5. [Adapted from (Sl68).]

Protection (ICRP) and National Council on Radiation Protection and Measurements (NCRP), have established **Derived Air Concentrations** (DACs) for radiation workers. These quantities have supplanted the **Maximum Permissible Concentrations** (MPCs) formerly employed for this purpose that reflected methodologies that have now been updated by the ICRP. DACs, as were MPCs, are based upon the receipt of 5000 mrem of dose equivalent if the entire working year (≈ 2000 hours, or 40 hours weekly) is spent working in a concentration corresponding to "1 DAC". A one DAC concentration is generally a quite large concentration that is rarely encountered in accelerator radiation environments. Similarly, for members of the general public, values of **Derived Concentration Guides** (DCGs) have been tabulated that would result in the receipt of 100 mrem of dose equivalent by an individual who spent all of the time in one year breathing such air. Table 8.6 gives representative values of these circumstance-dependent maximum concentrations, C_{max}, for accelerator-produced radionuclides in air based upon present U. S. Department of Energy Orders (DOE90) and Regulations (CFR93) along with companion values determined for accelerator-produced radionuclides not included in the cited references that have been calculated by Höfert (Hö69). For some radionuclides commonly found at accelerators, DOE regulations (CFR93) gives two values of DAC, one for air inhaled into the lungs and the other for immersion in an infinite cloud of γ-emitting radionuclides.

Table 8.6 DACs and DCGs (Air) for radiation workers and the general population. (μCi m^{-3}). These represent maximum concentrations for radionuclide *i*, $C_{max,i}$, depending upon the circumstances of exposure (see text).

Radionuclide	DAC-Radiation Worker 5 rem year^{-1}-40 hrs week^{-1}			DCG-General Population 0.1 rem year^{-1}-168 hrs week^{-1}	
	Inhaled Air (CFR93)	Immersion in infinite cloud (CFR93)	Immersion in 4 meter cloud (Hö69)	(DOE90)	(Hö69)
^{3}H	20			0.1	
^{7}Be	9			0.04	
^{11}C	200	4	59	1.0	
^{13}N		4	41		0.02
^{15}O		4	27		0.02
^{41}Ar		3	47		0.01
^{22}Na	0.3			0.001	

Immersion conditions are more likely to be the dominant exposure mechanism due to activated air at accelerators. However, for occupational exposures, the sizes of the "clouds" are not likely to be "infinite" but will be determined by the dimensions of the accelerator enclosures. Höfert's calculations are very important because they provide appropriate values for these accelerator-produced radionuclides. Höfert also recognized that such "immersion dose" is highly sensitive to the size of the cloud and that clouds of infinite extent are rare inside buildings at accelerators. He then calculated the equivalent of DACs for clouds of various sizes. Table 8.6 gives those for clouds of 4 meters radius that might be typical of an accelerator enclosure. For the general population, Höfert postulated an infinite cloud, since such exposure would presumably occur outdoors. Mixtures of radionuclides are commonly encountered. To account for the presence of multiple radionuclides, the set of individual radionuclide concentrations in the air, C_i, must satisfy the following inequality:

$$\sum_i \frac{C_i}{C_{\max,i}} \leq 1, \tag{8.12}$$

where $C_{max,i}$ is the regulatory standard for the i^{th} radionuclide, dependent upon the circumstances of the exposure.

8.2.5 Production of Airborne Radionuclides at Electron Accelerators

At electron accelerators, significant air activation will not occur without bremsstrahlung because the nuclear cross sections of *electrons* are about two orders of magnitude smaller than those of *photons* (Sw79a)[1]. This airborne radioactivity is generally short-lived and the concentrations, as shall be seen in what follows, are usually quickly reduced to levels where the exposure rates (R h^{-1}), or equivalently the absorbed dose rates (rads h^{-1}) are small compared to those due to the accelerator components. This result is because the radiation length of air is so much longer than that of any solid material (see Table 1.2).

Swanson (Sw79a) has calculated the saturation activities produced in air normalized to the electron beam power with the results provided in Table 8.7. The results of these calculations are normalized to unit path length and to beam power. To use them to determine the volume specific activity (e.g., Ci cm^{-3}) in a given accelerator enclosure, one must multiply the tabulated values by the available **bremsstrahlung path length**[2] and divide by the enclosure volume. The results found in this table were calculated in a manner completely analogous to those given in Table 7.2 for other materials. For energies close to the threshold of an individual reaction, the rise of activity with beam energy, E_o (see Section 7.3.2 and Fig. 3.7) must be considered. ^{41}Ar is produced in the thermal neutron capture (n, γ) reaction most copiously where there are high fluences of moderated neutrons present, typically near water-cooled targets and in concrete enclosures. ^{3}H and ^{7}Be are too long-lived to be produced in saturation and usually do not merit further consideration.

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[1] The reverse is true for toxic gas production, which occurs by a chemical, rather than nuclear, transformation and whose reaction rate is closely proportional to the integral dose to air. Such dose is generally higher if the primary electron beam does not strike a target to produce bremsstrahlung but rather is directly delivered to air. The production of such toxic gases, most notably oxone (O_3) is beyond the scope of this text but has been covered adequately by Swanson (Sw79a).

[2] The available path length would either be set by the physical dimensions of the room or, for a large room, by the attenuation length of the bremsstrahlung radiation in air.

Table 8.7 Saturation activities per unit path length and per unit beam power produced in air by an electron beam normalized to the beam power. See discussion in the text concerning the meaning of MPC values. "Cross section" refers to a spectrum-weighted value for photoneutron production. [Adapted from (Sw79a).]

Produced nuclide			Parent nuclide				Cross-section[b]	A_s[c]
Nuclide	$T_{\frac{1}{2}}$	MPC (μCi·cm^{-3})	f Abundance[a]	Nuclide	Reaction type	Threshold (MeV)	$\Sigma f\sigma_{-2}$ (μb·MeV^{-1})	Saturation activity (μCi·m^{-1}·kW^{-1})
H-3	12.262 a	2×10^{-3} [d]	1.562 0.424	N-14 O-16	(γ,H-3)	22.73 25.02	(3)	(140)
Be-7[f]	53.6 d	1×10^{-6} [d]	1.562 0.424	N-14 O-16	(γ,sp)[f]	27.81 31.86	(0.6)	(30)[f]
C-11	20.34 min	3×10^{-6} [e]	1.5×10^{-4}	C-12	(γ,n)	18.72	0.011	0.5
			1.562 0.424	N-14 O-16	(γ,sp)[f]	22.73 25.88	(6)	(300)[f]
N-13	9.96 min	2×10^{-6} [e]	1.562	N-14	(γ,n)	10.55	310	14000
O-15	123 s	2×10^{-6} [e]	0.424	O-16	(γ,n)	15.67	32	1500
N-16	7.14 s	5×10^{-7} [e]	4.0×10^{-4}	O-18	(γ,np)	21.81	(0.01)	(0.5)
Cl-38	37.29 min	2×10^{-6} [d]	4.6×10^{-3}	Ar-40	(γ,np)	20.59	0.13	6
Cl-39	55.5 min	3×10^{-6} [d]	4.6×10^{-3}	Ar-40	(γ,p)	12.52	0.86	40
Ar-41[g]	1.83 h	2×10^{-6} [e]	4.6×10^{-3}	Ar-40	(n,γ)	–	–	–[g]

[a] Fraction of air by volume, multiplied by atoms/molecule.

[b] Abundance f times integral cross-section σ_{-2}. Values in parentheses are rough estimates.

[c] Per bremsstrahlung pathlength in air (metres) and electron beam power (kW) incident on a thick high-Z target. Values in parentheses are rough estimates.

[d] Based on ICRP recommendation for radiation workers, 40-hour week, exposure from inhalation.

[e] Based on ICRP recommendation for radiation workers, 40-hour week, semi-infinite cloud.

[f] Spallation reaction.

[g] Neutron-capture reaction. Occurs where high neutron fluences are moderated by water or concrete shielding.

After calculating the production rates, one can then apply the general methodology presented in this chapter determine the concentrations within the accelerator enclosure and to estimate the effective dose equivalent rates at offsite locations as well as the status of compliance with applicable regulations.

8.2.6 Production of Airborne Radionuclides at Proton Accelerators

At proton accelerators, the excitation functions of the possible nuclear reactions listed in Table. 8.2 exemplified by those shown in Fig. 7.9 become important. In general, the positron emitters ^{11}C, ^{13}N, ^{15}O along with ^{41}Ar (produced by thermal neutron capture) are the nuclides most frequently seen. Work at Fermilab described by Butala et al. (Bu89) and Vaziri et al. (Va93 and Va96) has also confirmed these identifications and, additionally, detected ^{39}Cl. The determination of the relative contributions of the various positron emitters present must principally be done by fitting measured decay curves with a sum of exponential functions, each term of which represents one of the possible radionuclides present. This is a result of the fact that their γ-ray spectra are all dominated by 0.511 MeV photons from positron annihilation. The results of analysis of such decay curves have been discussed in various references (Th88, Sw90, Bu89, Va93, and Va96).

It was concluded by Butala et al. that the geometry of target stations significantly can affect the composition (Bu89). For example, high intensity targets immediately surrounded with large volumes of iron and concrete (in contact with the iron) produced much less ^{41}Ar than did other targets where the bulk iron shield was located in a open room with a layer of air between the iron and the concrete. Presumably, the open space provided opportunity for the large flux of low energy neutrons expected external to a pure iron shield (see Section 6.3.5) to "thermalize" and thus enhance the production of ^{41}Ar in the air space. The large cross section for the ^{40}Ar(n,γ)^{41}Ar reaction at thermal neutron energies (σ_{th} = 660 mb) also may have provided the photons necessary to enhance the (γ, p) and (γ, pn) reactions required to produce significant quantities of ^{39}Cl and ^{38}Cl, respectively. Some typical percentages of the various radionuclides, by activity concentration, released from high energy proton accelerators are given in Table 8.8.

Table 8.8 Measured radionuclide composition of typical airborne releases from proton accelerators

Situation	**Radionuclides (Activity Per Cent)**					
	^{11}C	**^{13}N**	**^{15}O**	**^{38}Cl**	**^{39}Cl**	**^{41}Ar**
CERN (Th88) 28 GeV protons	31.0	47.0	8.0			14.0
Fermilab (Bu89) 800 GeV protons						
(no gap between iron and concrete walls)	46.0	19.0	35.0			
(gap between iron and concrete walls)	42.0	14.0	0.0	0.0	10.0	34.0
Fermilab (Va93) 120 GeV protons	58.5	37.9		1.0	1.1	1.5
Fermilab (Va96) 120 GeV protons	64.6	30.5				5

After calculating the production rates, one can then apply the general methodology presented in this chapter to estimate the effective dose equivalent rates as well as the status of compliance with applicable regulations.

8.3 Water and Geological Media Activation

The protection of groundwater resources is a significant public concern that includes the need to assure protection of groundwater resources from contamination with radionuclides. In principal, radioactivity can be produced in soil or rock and in the water it contains. Sometimes the radioactivity produced in water is a matter of concern for occupational workers as well. In practice, it is not always a simple matter to separate these two sources. One could initiate calculations of groundwater activation at accelerators by starting from "first principles" and by using the activation formula, Eq. (7.8).

8.3.1 Water Activation at Electron Accelerators

As seen before, questions of radioactivation are generally less complex at electron accelerators. As was done for atmospheric activation and exhibited in Table 8.7, Swanson (Sw79a) has provided the results of calculations to address the production of radionuclides in water at electron accelerators. Such activation will principally occur in water used to cool magnets and beam absorbers and can become a radioactive waste issue. The results are, again, in the form of saturation activities normalized to the electron beam power absorbed in the water volume. Such activities, as before, are for infinite irradiation periods with no time allowed for decay. The results are given in Table 8.9, which also includes the point source specific gamma-ray constants, Γ_i, useful calculating exposure rates near such water as well as the obsolete maximum permissible concentrations in water (MPC_w's) and . From these results, it is clear that, aside from short-lived positron emitters, only ^3H and ^7Be are of importance. Table 8.9 gives the results due to interactions with ^{16}O found in water. Activity concentrations can be obtained by assuming rapid mixing of the saturated activity in the available volume of water. In principal, ^3H could be produced from the hydrogen in water by means of two sequential thermal neutron capture reactions, $^1H(n,\gamma)^2H$ followed by $^2H(n,\gamma)^3H$. However this is unimportant due to the fact that the cross sections for both thermal capture reactions involved are fractions of a millibarn.

In practice, due to the compactness of the shielding at electron accelerators compared with that found at proton and ion accelerators, soil activation is generally not a severe problem at such facilities.

Table 8.9 Saturation activities per unit beam power produced in ^{16}O as found in by an electron beam normalized to the beam power. "Cross section" refers to a spectrum-weighted value for photoneutron production. [Adapted from (Sw79a).]

Nuclide	$T_{\frac{1}{2}}$	MPC_W [a] ($\mu Ci \cdot cm^{-3}$)	Γ Specific gamma-ray constant ($(R \cdot h^{-1})(Ci \cdot m^{-2})^{-1}$)	Reaction type	Threshold (MeV)	Cross-section [b] σ_{-2} ($\mu b \cdot MeV^{-1}$)	$A_s^{b,c}$ Saturation activity ($Ci \cdot kW^{-1}$)
O-15	123 s	–	0.59 (β^+)	(γ,n)	15.67	75	9
O-14	70.91 s	–	1.60 (β^+)	(γ,2n)	28.89	(1)	(0.1)
N-13	9.96 min	–	0.59 (β^+)	(γ,2np)	25.02	0.9	0.1
C-11	20.34 min	–	0.59 (β^+)	(γ,3n2p)	25.88	3	0.4
C-10	19.48 s	–	1.01 (β^+)	(γ,4n2p)	38.10	(1)	(0.1)
Be-7	53.6 d	0.02	0.029 –	(γ,5n4p)	31.86	0.3	0.04
H-3	12.262 a	0.03	– (β^-)	(γ,H-3)	25.02	1.5	0.2

[a] ICRP recommendation for the general public, 168-hour week occupancy.

[b] Values in parentheses are rough estimates.

[c] Saturation activity in water per unit electron beam power. Assume 100% direct absorption of electron beam power in water. Activity in water will be less in most situations where the beam absorber is water-cooled metal. Values shown are obtained directly from Approximation A and apply at high energies. For $E_0 \gtrsim 50$ MeV, the value for O-15 may be reduced by a factor of two, and others by an even larger factor.

8.3.2 Water and Geological Media Activation at Proton Accelerators

8.3.2.1 Water Activation

At proton and ion accelerators, as with electron accelerators, radioactivity can be produced directly in water as a result of both proton and neutron interactions. Values for the relevant cross sections were given in Chapter 7. Equipped with knowledge of the beam energy and information about the energy spectra of neutrons that are present, one can proceed to calculate the radionuclides present. In general, the most important radionuclides, as is the situation with electron accelerators, are the interactions of the hadrons with the oxygen present in the water. As before, the production of ^{3}H from the hydrogen present in the water is possible, but is rendered sufficiently improbable due to the small cross sections of both of the thermal neutron capture reactions required to occur sequentially. For such calculations, the production of ^{3}H in water from atoms other than hydrogen is of special importance. Published experimental data of ^{3}H production reactions is surprisingly scarce. Konobeyev and Korovin (Ko93) have developed a method of globally fitting the existing cross section data on the production of ^{3}H due to neutron interactions with a variety of target elements found in soils. The results for protons are similar and the results are shown in Fig. 8.4.

8.3.2.2 Geological Media Activation

While calculating the production of radionuclides in soil, and in the water it contains, directly from known cross sections has an appeal due to its simplicity, in practice such calculations have been done more frequently by analyzing results obtained using irradiated samples. The work of Borak et al. (Bo72) is of singular importance in this regard. Borak et al. measured the radioactivity produced in soil by high energy hadrons by radiochemical analysis of soil samples irradiated near high energy synchrotrons; the 12 GeV Argonne ZGS and the 28 GeV Brookhaven AGS. The radionuclides ^{3}H, ^{7}Be, ^{22}Na, ^{45}Ca, ^{46}Sc, ^{48}V, ^{51}Cr, ^{54}Mn, ^{55}Fe, ^{59}Fe, and ^{60}Co were identified. Bench-top experiments were then performed to determine which radionuclides, and what fractions of them, could be leached by water. This study determined macroscopic production cross sections and ion velocities relative to ground water flow in soil. Of these nuclides, only ^{3}H, ^{22}Na, ^{45}Ca, and ^{54}Mn were observed in leach waters. The ^{3}H was assumed to be all leachable and was measured by driving it out of the sample by baking. Radionuclides with half lives exceeding 15 days were the only ones considered. The results were based upon the elemental composition of soil given in Table 8.10.

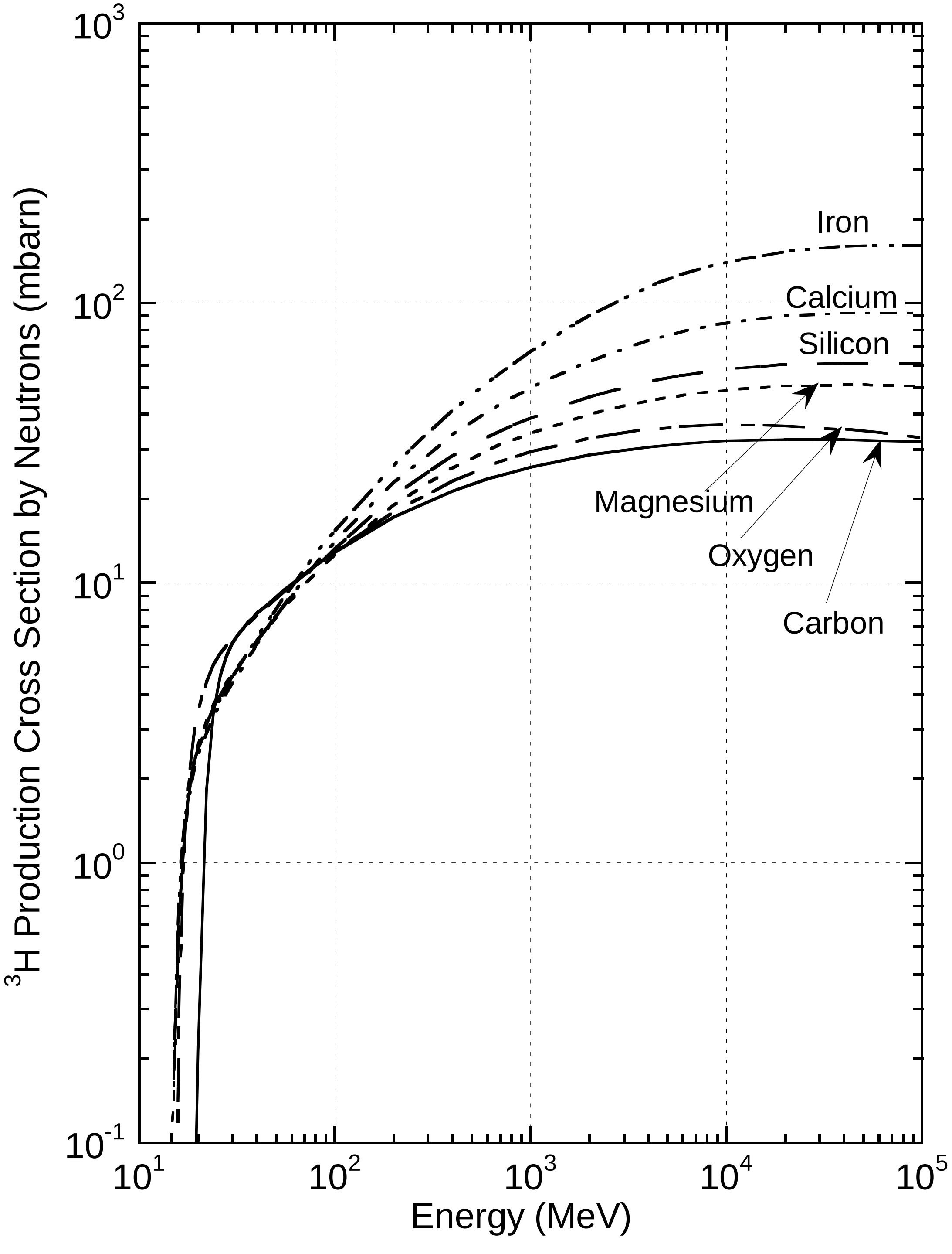

Fig. 8.4 Cross sections for the production of ^{3}H due to neutron bombardment of materials commonly found in soil and rock as a function of neutron energy. The calculations have been performed following the method of Konobeyev and Korovin (Ko93). Results for aluminum are quite similar to those found for silicon and the results for sodium are quite similar to those found for magnesium.

Table 8.10 Composition of Soils Typical of the Fermilab site. [Adapted from Bo72).]

	Elemental Composition of Soil*	
Element	***Z*, Atomic Number**	**% by Weight**
Silicon	14	14.47
Aluminum	13	2.44
Iron	26	1.11
Calcium	20	7
Magnesium	12	3.79
Carbon	6	5.12
Sodium	11	0.34
Potassium	19	0.814
Oxygen	8	≈ 64

*The mean moisture percentage was 13.15 ± 4.45 % and the mean pH was 7.6 ± 0.1.

Borak et al. measured specific activities at saturation, A_i, (Bq g^{-1}) which are related to the microscopic cross sections by means of the following equation,

$$A_i = \phi \sum_j n_j \sigma_{ij} \, , \tag{8.13}$$

where ϕ is the flux density (cm^{-2} s^{-1}), n_i is the number density of target nuclei of the j^{th} nuclide (g^{-1}) of the soil sample, and σ_{ij} (cm^2) is the effective cross section for the transformation from target nucleus *j* to radionuclide *i*. The summation is taken over the soil constituents. Borak et al. were able to directly measure the summations on the right hand side of Eq (8.13). These summations are the total **macroscopic cross sections** for each radionuclides of interest. Table 8.11 gives the results of the measurements of the macroscopic cross sections, denoted Σ (cm^2 g^{-1}), for each of the radionuclides identified in the various types of soils analyzed.

Table 8.11 Macroscopic cross section for soil normalized to unit flux of hadrons with kinetic energies greater than 30 MeV. [Adapted from (Bo72).]

Nuclide	**Glacial Till** Σ **(cm^2 g^{-1})**	**Gray Sandy Clay** Σ **(cm^2 g^{-1})**	**Red Sandy Clay** Σ **(cm^2 g^{-1})**	**Gray Clay** Σ **(cm^2 g^{-1})**
^{7}Be	2.9 x 10^{-4}	3.7 x 10^{-4}	3.2 x 10^{-4}	2.7 x 10^{-4}
^{51}Cr	1.7 x 10^{-5}	3.7 x 10^{-5}	2.8 x 10^{-5}	3.1 x 10^{-5}
^{22}Na	2.1 x 10^{-4}	2.3 x 10^{-4}	2.0 x 10^{-4}	1.6 x 10^{-4}
^{54}Mn	5.9 x 10^{-5}	4.1 x 10^{-5}	3.5 x 10^{-5}	3.7 x 10^{-5}
^{46}Sc	3.0 x 10^{-5}	1.3 x 10^{-5}	9.6 x 10^{-6}	1.1 x 10^{-5}
^{48}V	4.1 x 10^{-6}	1.1 x 10^{-5}	6.7 x 10^{-6}	7.4 x 10^{-6}
^{55}Fe	9.3 x 10^{-5}	1.2 x 10^{-4}	7.0 x 10^{-5}	2.1 x 10^{-4}
^{59}Fe	3.2 x 10^{-6}	1.7 x 10^{-6}	1.3 x 10^{-6}	1.6 x 10^{-6}
^{60}Co	3.3 x 10^{-5}	1.4 x 10^{-5}	1.1 x 10^{-5}	1.3 x 10^{-5}
^{45}Ca	1.6 x 10^{-4}	2.0 x 10^{-5}	3.0 x 10^{-5}	1.6 x 10^{-5}
^{3}H	8.2 x 10^{-4}	1.1 x 10^{-3}	3.3 x 10^{-4}	5.2 x 10^{-4}
^{3}H*	5.9 x 10^{-3}	5.9 x 10^{-3}	4.1 x 10^{-3}	4.4 x 10^{-3}

*Cross sections per gram of water in soil.

Borak et al. also measured some results concerning the **leachabilities** of the various elements from the soils studied. This was a measurement of the ability of water to remove a given radionuclide from the soil material. It is not related to nuclear properties but rather is related to chemical properties and processes, such as ion exchange. The results were reported by Borak et al. as follows:

^{3}H: The leaching process was able to collect all the tritium measured by the bake-out process. The average value of the macroscopic cross section in soil was found to be 5.1 x 10^{-3} cm^{2} g^{-1} of water. An important conclusion is that the tritium will migrate with the same velocity as any other water in the soil.

^{22}Na: Typically 10-20 % of this nuclide was leachable. On average, it appeared that the migration velocity of this nuclide is approximately 40% of that of water through the soil due to ion exchange processes.

^{45}Ca: At most 5 % of this nuclide was leached from the soil. The migration velocity was determined to be extremely small.

^{54}Mn: At most 2 % of this nuclide was leached from the soil. It was determined that this nuclide will not migrate significant distances.

Thus, based upon leachability considerations, ^{3}H and ^{22}Na are thus the most important radionuclides that can be produced in environmental media such as soil.

One can thus calculate the quantities of radionuclides that might pose a risk to groundwater in the environs of an accelerator. This can be done by using the cross sections directly, or, as demonstrated by Gollon (Go78) for high energy protons, by performing, for example, Monte Carlo calculations in which the total stars (or inelastic interactions above some threshold) produced in some volume of earth shielding are determined[3]. As in Eq. (7.35), the total number of atoms, K_i, of the i^{th} nuclide that can be produced per star in that same volume is given by

$$K_i = \frac{\Sigma_i}{\Sigma_{in}}, \tag{8.14}$$

where Σ_i is, as above, the macroscopic cross section (cm^{2} g^{-1}) for the i^{th} radionuclide and Σ_{in} is the total macroscopic inelastic cross section (cm^{2} g^{-1}) for soil. Gollon quoted a value of Σ_{in} = 1.1 x 10^{-2} cm^{2} g^{-1} for soil based upon the results of Borak et al.

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[3] Some Monte Carlo codes of more recent development can now calculate these quantities directly from the energy-dependent production cross sections. However, given the limited energy dependence at high energies, working with the total stars remains worthwhile as a means to achieve results rapidly, or as a "quality check" on the more complex computations.

Gollon used the following values of K_i for ^{3}H and ^{22}Na, respectively, as selected from Borak's paper for soils peculiar to Fermilab (glacial till):

$$K_3 = \frac{8.2\times10^{-4}}{1.1\times10^{-2}} = 0.075\text{, and} \tag{8.15a}$$

$$K_3 = \frac{2.1\times10^{-4}}{1.1\times10^{-2}} = 0.020\,. \tag{8.15b}$$

One can then calculate the total number of atoms of radionuclides produced during some time interval in some volume by simply multiplying these factors by the number of stars (or nonelastic interactions) in the same volume. The number of atoms then can be converted to activity using the decay constant. The above values of K_i are applicable to soils such as those found at Fermilab. For other soil compositions one may need to use cross sections for producing the radionuclides of interest in various target elements and perform an integration over the energy spectrum of incident hadrons. Figure 8.5 gives cross sections for producing ^{22}Na by interactions of hadrons with the various elements comprising soil due to Van Ginneken (Va71). This figure should be regarded as a companion to Fig 8.4.

8.3.3 Regulatory Standards

The quantity of ultimate concern, of course, is the resultant concentration in water. The water could be an actual or potential drinking water resource that might well be protected by regulatory authorities. The regulations may well differ between different governing jurisdictions. Such requirements were generally not developed for application to the operations of particle accelerators and generally need to be well-understood by the designers of accelerator facilities. The standards differ for drinking water supplies and surface water discharges. The allowable concentrations for surface waters are generally, but not always larger due to the likelihood that such discharges will most certainly be diluted significantly prior to the consumption by individuals. For public drinking water supplies, the U. S. Environmental Protection Agency (CFR76, reaffirmed in CFR00) limits such concentrations to those that would produce an annual dose equivalent of 4 mrem and specifically gives a limit of 20 pCi cm^{-3} for tritium as a legal limit. An explicit limit for ^{22}Na is not specified by USEPA. For surface water discharges, the U. S. Department of Energy (DOE90) has set forth **Derived Concentration Guides** which would result in an occupation worker receiving no more than 100 mrem yr^{-1} should they use such water during their working hours. The DOE DCGs are based upon a more up-to-date dosimetry methodology that results in a limit of 80 pCi cm^{-3} for ^{3}H and 0.4 pCi cm^{-3} for ^{22}Na in drinking water. Table 8.12 lists these concentration limits, $C_{max,i}$. For purposes of this discussion, surface water discharges include those that add water to streams, ponds, etc. while drinking water standards apply to water that could potentially end up in a source of drinking water such as a public, or private well.

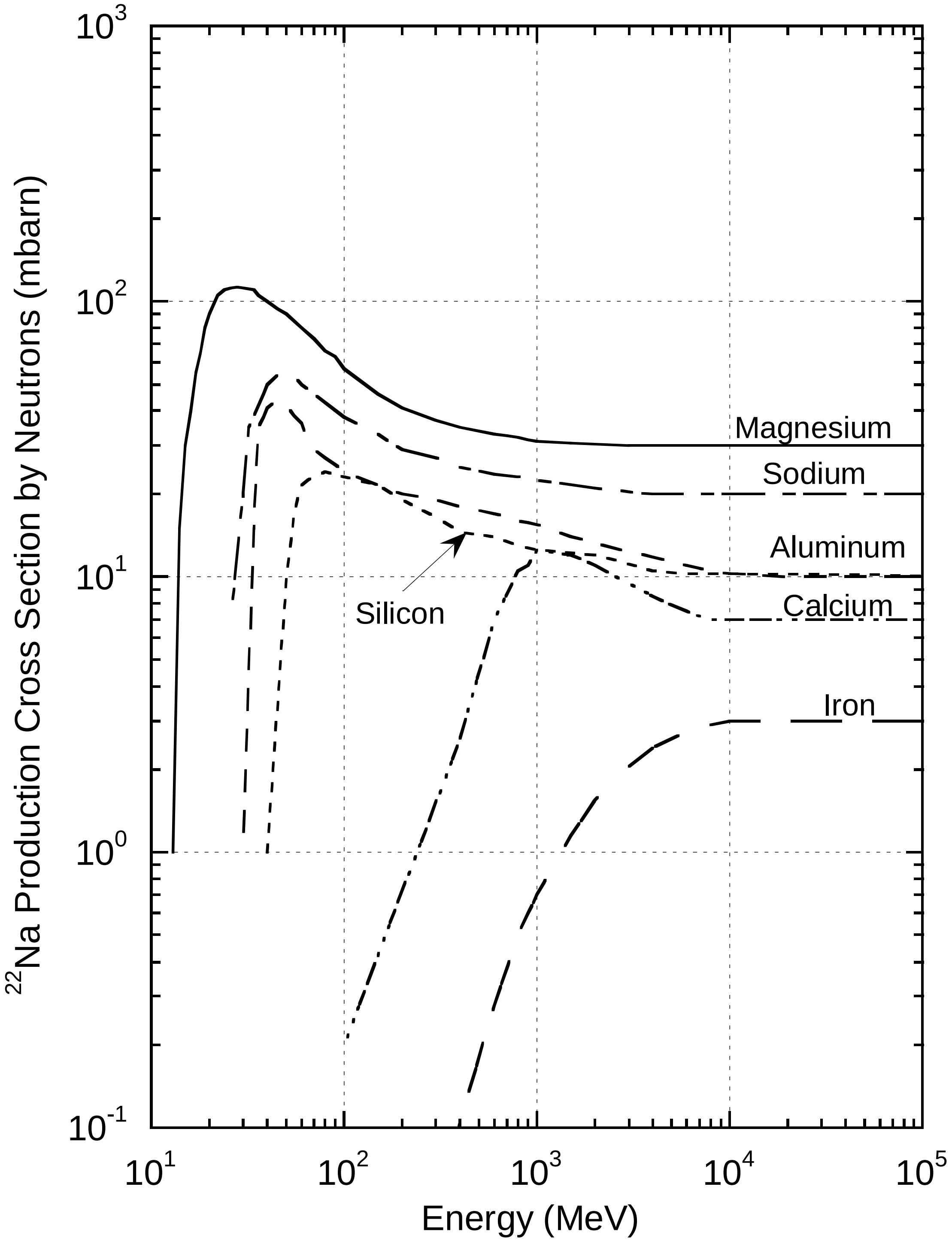

Fig. 8.5 Cross sections for the production of ^{22}Na due to neutron bombardment of materials commonly found in soil and rock as a function of neutron energy. Results for potassium are quite similar to those found for calcium. [Adapted from (Va71)].

Table 8.12 Concentration Limits for ^{3}H and ^{22}Na in surface water discharges and in drinking water

Radionuclides	Half Life (years)	Concentration Limit, $C_{max,i}$ (pCi cm^{-3})	
		Surface Water	Drinking Water
^{3}H	12.32	2000[a]	20[b]
^{22}Na	2.604	10[a]	0.4[a]

[a]Value taken from (DOE90)
[b]Value taken from (CFR76 CFR00). A value of 80 pCi cm^{-3} results from (DOE90).

In exact analog with the situation found with airborne radioactivity, to account for the presence of multiple radionuclides, the set of radionuclide concentrations in the water, C_i, must satisfy the following inequality, where $C_{max,i}$ is the regulatory standard for the i^{th} radionuclide for the particular circumstances of exposure;

$$\sum_i \frac{C_i}{C_{\max,i}} \leq 1. \tag{8.16}$$

8.3.4 The Propagation of Radionuclides Through Geological Media

The methods for calculating these concentrations in actual environmental media will vary with the regulatory authority and the "conservatism" of the institution. The most conservative assumption is to assume that saturation values of production are reached. This is equivalent to assuming that the accelerator will operate “forever” in a static configuration and that the water in its vicinity never moves. The former is an extremely unrealistic assumption and it is questionable that the "motionless" water in such a medium actually comprises a potential source of useable drinking water. However, for this static case, the activity concentration C_i of radionuclide i, under such conditions can be calculated by means of following formula:

$$C_i = \frac{N_p K_i L_i S_{ave}}{1.17\times10^{6}\rho w_i}\left\{1-\exp(t_{irrad}/\tau_i)\right\}\exp(-t_c/\tau_i)\ (\text{pCi cm}^{-3}), \tag{8.17}$$

where,

- N_p is the number if incident particles delivered per year,
- K_i is as above,
- L_i is the fraction of the radionuclide of interest that is leachable,
- S_{ave} is the average star density (stars cm^{-3}) in the volume of interest per incident particle,
- ρ is the density of the medium (g cm^{-3}),
- w_i is the mass (grams) of water per unit mass (grams) of medium required to leach some specified fraction of the leachable radioactivity and is, thus, linked to the value of L_i.
- t_{irrad} is the irradiation time,
- t_c is the “cooling” time once the irradiation is suspended, and
- τ_i is the mean life of the i^{th} radionuclide.

The constant in the denominator contains the unit conversions needed to yield pCi cm^{-3}. For a given medium, the ratio L_i/w_i should be determined by measurements specific to the local media. An important quantity is the **effective porosity**, p, which represents the volume fraction of the material that is available to water movement. It is given by,

$$p = \rho w_i. \tag{8.18}$$

The effective porosity is essentially equal to the pore volume of the material for soils but for consolidated materials (i.e., rock) it does not include sealed pores through which movement is not allowed. This provides a means by which “worst case” estimates may be made. For realistic estimates some method of taking into account water movement must be used.

At Fermilab, a simple model allowing for some movement and further dilution of water was employed for many years (Go78). In this model, called the **single resident model** for reasons that shall become obvious, the vertical migration of water was assumed to be 2.2 m yr^{-1}. In the standard clays present, this migration velocity is conservative, likely high, by at least an order of magnitude. Its use crudely allowed for the presence of cracks and fissures through which more rapid propagation of water might be possible. The tritium vertical velocity was taken to have this value while the results obtained by Borak et al. (Bo72) were used to obtain a reduced value of about one m yr^{-1} for ^{22}Na. Only the leachable fraction of the ^{22}Na is included. The procedure then allowed for decay during the downward migration of the total inventory of radionuclides produced in one year, integrated over the entire volume of the irradiated material, to the highest aquifer below the location of the irradiation. At that point, it was assumed that the radionuclides were rapidly transported horizontally to a shallow well where it was assumed that the flow of water collecting the radionuclides is entirely used by a single user who consumes a volume of 150 liters per day. This value, a minimal one, was taken from results achieved by municipalities that needed to ration public water consumption during severe drought conditions. Thus the annual production, as transported vertically, was diluted into the 5.5 x 10^{7} cm^{3} yr^{-1} that this represents. This simple model is generally conservative but does, in fact, neglect that fact that the water movement may not be uniform from year-to-year. It also did not use the fact that the radionuclides are initially distributed over a considerable volume as they are produced.

It is clear that better methods are warranted and a new model has been developed for use at Fermilab (Ma93). The **concentration model** now in use at Fermilab calculates the production of the radionuclides of concern in accordance with Eq. (8.17). The result, then, provides an initial concentration that is available for further migration, decay, and dilution. The concentration after migration is, then, calculated by using up-to-date modeling techniques to calculate the reduction in the concentration due to dilution, diffusion, and radioactive decay. At the point of concern, usually the location of an aquifer producing water suitable for consumption as a supply of drinking water, the concentrations calculated are then substituted into Eq. (8.16) in order to determine if a shielding design is adequate.

To do these calculations properly requires a detailed knowledge of the media involved. Some principles will be given here. In situations where a definite potential gradient, often called the **hydraulic gradient**, *dh/dx*, is applied to water in a medium, the rate of flow is said to be **advective**. Under such conditions and in situations where only one dimensional coordinate is important, the average linear velocity (or seepage velocity), *v*, is given by the application of **Darcy's Law** as (Fe88),

$$v = \frac{K}{p}\frac{dh}{dx}, \tag{8.19}$$

where *p* is, as above, the effective porosity. More complicated situations involving two and three dimensions are addressable using the mathematical language of vector calculus. The derivative is the gradient of the hydraulic head in the material. *K* in this equation represents the **hydraulic conductivity.** This quantity is a function of the material and its moisture content. All of the factors in this equation can, and generally should, be determined empirically for the medium and location under consideration. Typical values of *K* are given in Table 8.13 and have been given by Batu (Ba98).

Table 8.13 Examples of typical values of hydraulic conductivity [Adapted from (Ba98).]

Group	**Porous Materials**	**Range of *K* values (cm s^{-1})**
Igneous Rocks	Weathered granite	(3.3 - 52) x 10^{-4}
	Weathered gabbro	(0.5 - 3.8) x 10^{-4}
	Basalt	(0.2 - 4250) x 10^{-6}
Sedimentary Materials	Sandstone (fine)	(0.5 - 2250) x 10^{-6}
	Siltstone	(0.1 - 142) x 10^{-8}
	Sand (fine)	(0.2 - 189) x 10^{-4}
	Sand (medium)	(0.9-567) x 10^{-4}
	Sand (coarse)	(0.9- 6610) x 10^{-4}
	Limestone and dolomite	(0.4 -2000) x 10^{-7}
	Karst limestone	(1- 20000) x 10^{-4}
	Gravel	(0.3 - 31.2) x 10^{-1}
	Silt	(0.09-7090) x 10^{-7}
	Clay	0.1 - 47) x 10^{-8}
Metamorphic Rocks	Schist	(0.002 - 1130) x 10^{-6}

Darcy's Law can, then, be used to determine the rate of migration of a contaminant, in this case, radioactivity from one point to another. During the time of migration, the concentration would be reduced due to radioactive decay. This method would be particularly applicable to determining the migration of contamination due to spills.
One often encounters the problem of calculating the concentration of radionuclides at some location as a function of time during, or after, a period of irradiation comparable to the mean lives of the radionuclides of concerns. At a given location in such a medium,

denoted by the coordinate x, one needs to solve the following continuity equation that can be thought of as an extension of Eq. (7.4), for situations where the velocity of water movement, v, can be thought of as slowly varying or a constant over time and some volume of space.

$$\frac{\partial C_i}{\partial t} + v\frac{\partial C_i}{\partial x} + \lambda_i C_i(x,t) = \frac{L_i}{w_i'} Q_i(x,t)\,, \tag{8.20}$$

where all variables are as in Eq. (8.17) except that λ_i is the decay constant of the i^{th} radionuclide, x is the spatial coordinate, t is the time, w_i' is the water content of the media per unit volume of media. The quantity $Q_i(x,t)$ represents the production of the i^{th} radionuclide and is equivalent to the factor $N_pS_{ave}/(1.17 \times 10^6\rho)$. It includes any time-dependence in the delivery of beam. The middle term in the left-hand side of the equation takes care of movement from a point of one concentration to another at the seepage velocity v of the particular radionuclide i. One can commonly describe the spatial dependence of the production factor as,

$$Q_i(x,t) = Q_{oi}(t)\exp(-\xi x)\,, \tag{8.21}$$

where the spatial distribution of the production follows an exponential dependence as is typical in a large shield.

Mokhov (Mo97) has solved this equation for the typical initial conditions of $C_i(x,0)=0$ and $x \geq 0$, $t \geq 0$.

In general,

$$C_i(x,t) = \frac{L_i}{w_i}\int_o^t dt' Q_i(x - vt', t')\exp(-\lambda t')\,, \tag{8.22}$$

and for an exponential spatial dependence as in Eq. (8.21) this becomes:

$$C_i(x,t) = Q_{oi}(t)\frac{L_i}{w_i}\frac{1}{\eta_i}\exp(-\xi x)[\exp(\eta_i \tau) - 1]\,,$$

with

$$\eta_i = \xi v - \lambda_i\,,$$
$$\tau = t \text{ for } t < x/v, \text{ and}$$
$$\tau = x/v \text{ for } t \geq x/v. \tag{8.23}$$

$C_i(x,t)$ has a maximum at $x_{i,\max}$ given by,

$$x_{i,\max} = -\frac{v}{\lambda_i}\frac{\ln\left(\frac{\xi v}{\lambda_i}\right)}{1 - \frac{\xi v}{\lambda_i}}. \tag{8.24}$$

In using these results, one must take care that the algebraic signs of the coordinates *x* relative to that of *v* are properly taken into account. In situations where the seepage velocity is extremely slow, **diffusion** becomes the dominant mechanism for water flow and dilution. Mathematically, a second partial derivative with respect to the spatial coordinate is added to Eq. (8.20). Examples are provided by Fetter (Fe88). Computer software has been written to address this topic such as the one produced by Sudicky, et al. (Su88).

As a further example of methodologies that can be employed in solving such problems, Jackson (Ja87) has estimated the dilution for a shallow uncased well in an aquifer a distance *r* from a beam loss point also in the aquifer. The loss point was assumed to be within the drawdown zone of the well. This was performed for a simple geology that involved a single uniform stratum of earth above some level of impervious stratum. Fig. 8.6 shows the situation described by this model. Here, a given well is modeled by using the profile of depth of water *h(r)* at distance r from the well. *h(r)* is determined by the depth of a test well at radius *r* from the well under consideration and represents the hydraulic potential. The well is assumed to supply a volume *Q* of water per day. The flux of water is determined by the gradient relation, equivalent to Darcy's Law,

$$S_r = k\frac{dh(r)}{dr}\ , \tag{8.25}$$

where S_r is the inward flux at radius *r* and *k* is a constant with dimensions of volume per unit time per unit area and is characteristic of the soil. Conservation of water yields the steady-state equation:

$$Q = 2\pi rh(r)S_r = 2\pi rkh\frac{dh}{dr} = \pi k\frac{d\left(h^2\right)}{d\left(\ln r\right)}\ . \tag{8.26}$$

The quantity $2\pi rh\dfrac{dh}{dr}$ corresponds to the rate of change of volume of the cylindrical shell of height *h* (i.e., the **head**) with respect to *r*. This equation has the solution;

$$Q\ln\left(\frac{r}{r_o}\right) = \pi\,k\left\{h^2(r) - h_o^2\right\}, \tag{8.27}$$

where r_o is the radius of the well and h_o is the height of water above the impervious stratum at the well. If *H* is the depth of the impervious layer below the water table unperturbed by the wells, the radius of influence *R* of the well can be defined by the relation:

$$\ln\frac{R}{r_o} = \frac{\pi\,k\left\{H^2 - h_o^2\right\}}{Q}\ . \tag{8.28}$$

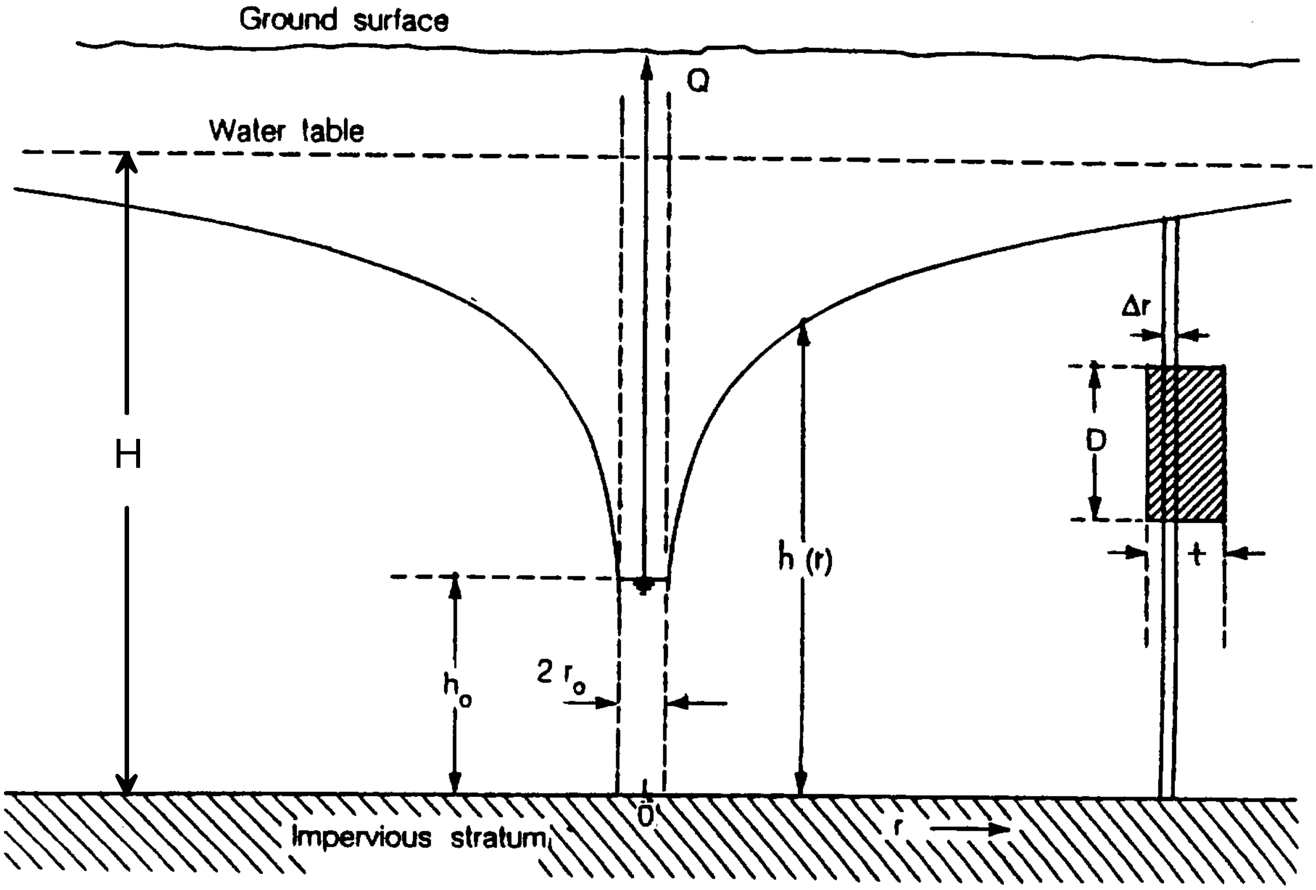

Fig. 8.6 Hydrogeological model of a shallow well in proximity to an accelerator tunnel where a beam loss occurs. The radioactivated region is represented in cross section by the shaded rectangle to the right. *h* represents the elevation of the water table above the impervious stratum as a function of *r* while the water table is a distance *H* above the impervious stratum where the water table is not perturbed by wells. [Adapted from (Ja87).]

However, the detailed solution is not necessary. Now, suppose that there is a well a distance *r* away from the region of deposition of radioactivity near an accelerator. We also assume that the activation zone lies below the water table and that the deposition region lies within the radius of influence of the well. This assumption leads to higher concentrations than would be obtained if the activation zone were totally, or partially, above the water table. The amount of activity drawn into the well is determined by the rate of pumping *Q* and the necessary total flow through a cylinder of radius r and height *h(r)* as we have seen. Let ΔV be the volume of soil yielding *Q* gallons of water. The cylindrical shell providing this amount of water will be of radial thickness Δr, where $\Delta V = 2\pi r h(r)\Delta r$. The fraction *F* of the volume of activity included in this shell can be said to be given by:

$$F = \frac{\Delta r}{t} = \frac{2\pi\, rh\Delta r}{2\pi\, rht} = \frac{\Delta V}{2\pi\, rht}\,, \qquad (8.29)$$

provided that $\Delta r < t$.

If the activated region contains leachable activity, *A* (either total activity or that of a particular radionuclide of interest), the corresponding specific activity, *a*, in water drawn from the well is thus given by:

$$a = F\frac{A}{Q} = F\frac{A}{p\Delta V} = \left[\frac{\Delta V}{2\pi\, r\, h\, t}\right]\frac{1}{p}\left[\frac{1}{\Delta V}\right]A = \frac{1}{2\pi\, r\, t\, D}\frac{f}{p}A \quad , \tag{8.30}$$

where $f = D/h$ is the fraction of the total height of the cylindrical shell occupied by the activated region and p is the **porosity** of the soil. The pumping volume Q is implicit in f. Porosity values vary considerably but in general are in the range,

$$0.2 < p < 0.35. \tag{8.31}$$

Thus, this formula may be used to obtain an estimate of the specific activity as a function of distance from the well, although it is perhaps not too useful for applications to beam losses far from the well. By definition, $f \leq 1$ and the lower value of porosity can be used to obtain upper limit estimates of the concentration. It must be emphasized that this model depends upon uniformity of water conduction by the strata. The presence of "cracks" or more complex geological strata, of course, can provide much more rapid movement that is not well-described by this simple model.

Problems

1. A 20 m long air gap has a beam of 10^{12} s^{-1} of high energy protons passing through it. First, calculate the production rate of ^{11}C in the gap at equilibrium if one approximates air in the gap by nitrogen and assumes σ (^{11}C) = 10 mb. Assume that there are no significant losses of beam by interaction after checking to see that this assumption is, in fact, true. Table 1.2 contains helpful information.

   a) If the air gap is in a 10 x 10 x 20 meter^3 enclosure with <u>no</u> ventilation, calculate the equilibrium concentration of ^{11}C in the room (in units of μCi m^{-3}) assuming extremely rapid mixing (i.e., no time allowed for decay while mixing occurs) of the enclosed air. Compare the concentration with the derived air concentration values in Table 8.6 and calculate, using simple scaling, the dose equivalent to a worker who spends full time in this room. (This is a purely hypothetical scenario due to the much larger hazards due to such an intense direct beam!)

   b) Calculate the concentration if two (2) air changes hr^{-1} are provided.

   c) Assume the exhaust of the ventilation described in part "b" is through a 10 cm radius stack 25 m tall. Calculate the air speed in the stack, and the emission rate in Ci s^{-1}. Then using Cember's version of Sutton's equation for tall stacks to estimate the concentration directly downwind at ground level, and hence the dose equivalent 1 km away with moderately stable meteorological conditions and an average wind speed of 10 km hr^{-1}.

   d) Perform the same calculation requested in "c" using the more general version of Sutton's equation appropriate to short stacks and assume the stack height to be 3 meters. All other conditions of the problems are the same as in "c".

2. In soil conditions similar to those at Fermilab, a volume of soil around a beam dump approximately 10 m wide by 10 m high by 20 m long is the scene of a star production rate (averaged over the year) of 0.02 stars proton^{-1} at a beam intensity of 10^{12} protons $^{-1}$.

   a) Calculate the annual production of ^3H ($t_{1/2}$ = 12.3 years), the saturated activity (in Bq & Ci), and the average saturated specific activity in the above volume's water (assume 10% water content by volume).

   b) Use the older Fermilab single residence model to calculate the concentration at the nearest well. Assume the activation region (beam loss point) is 50 m above the aquifer and the usual migration velocities.

   c) "Conservatively" apply the "Jackson Model" to estimate the concentration at a well 100 meters distant from the center of the activation region.