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**Production of Radionuclides and Their Migration in Groundwater:
A Fermilab Case History and Model**

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**PRODUCTION OF RADIONUCLIDES AND THEIR MIGRATION IN
GROUNDWATER:
A FERMILAB CASE HISTORY & MODEL**

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ABSTRACT

Particle accelerators that are buried underground like those at Fermilab create a condition where soil can be activated. Naturally percolating groundwater becomes contaminated by leaching out some of the radioactivity as it migrates through the soil to the underlying aquifer. The Fermilab Concentration Model was formulated to account for and combine the fundamental processes of production, leaching, and migration. Its general features are described, then site-specific data from one of the target stations are used to make calculations and compare them to regulatory limits and DOE guidelines.

INTRODUCTION AND SITE CHARACTERISTICS

Fermilab is a national laboratory managed by Universities Research Association, Inc. for the U. S. Department of Energy. It is located in the greater Chicago area and covers 6800 acres. The major activity of the laboratory is to provide resources to conduct basic research in high-energy physics. A series of particle accelerators are used to probe the structure of the fundamental constituents of matter inside the atom. The particle accelerator (known as a proton synchrotron) is about 4 miles in length and is buried about 20 feet below the earth's surface to take advantage of low cost soil shielding. While this results in an almost negligible radiation exposure to personnel at ground level, it creates a condition underground where soil can be activated. Some of this activation can be dissolved by the natural percolating groundwater and transported to the underlying aquifer.

The region surrounding the laboratory is rapidly changing from farming to residential use. The 1990 census showed there were about 4000 people living within a 2 mile radius of the site, and about 20000 within a 3 mile radius [Grobe 1993]. Much of the land (about 25%) within the boundaries of Fermilab remains in crop production, but also includes open areas consisting of forests, prairie, grasslands and wetlands. The Fox River is about 1.5 miles from the western boundary, while the DuPage River is about 0.5 miles from the eastern boundary. The on-site geology is characterized by an upper layer of approximately 70 feet of glacial deposits, mainly silty clays. Below this layer is about 140 feet of Silurian Dolomite. Being weathered and fractured at the top, it yields sufficient quantities of water to be a groundwater aquifer. In fact, the primary drinking water supply for Fermilab is a well that taps into this Silurian aquifer. Figures 1 and 2 show elevation profiles for the lab's two high intensity target stations. Deposits from

advancing and receding glaciers have left mostly horizontal layers, predominantly clay. However, there are isolated areas of non-interconnected sand and gravel.

MODELING THE RELEVANT PROCESSES

Radiation is produced whenever energetic particles strike an object and initiate a nuclear cascade of secondary particles. A zone of activation is created which includes nearby soil. The water content of radioactive soil in its ionized state is available to exchange with normal groundwater, thereby making the groundwater radioactive. As it migrates downward to the aquifer, it becomes a potential source of contamination. To quantitatively estimate the concentration reaching the aquifer requires knowledge of the seepage velocity through the underlying geology. A mathematical model of the migration process is used to compare the final concentration with the regulatory limits. It is important to be able to make this comparison to assure that drinking water supplies are not being degraded and are available for use by future generations.

Our information about the interaction of particles with one another and their products is described by a parameter known as the inelastic cross section, σ . In a reaction, $p + N \rightarrow A + X$, with the proton as the projectile and the nucleus as the target, σ represents the process where the composition and/or energy of the target nucleus N changes into a final state with nucleus A and X (everything else, which may consist of one or more distinct parts). Since the exact dynamic details for producing the reaction products are not easily described, it is convenient to define σ for the production of A as the total probability for obtaining A nuclei per proton per target particle. It is an experimentally measured number which is statistical in nature and tells how many A 's are "made" for a given number of incident projectiles "hitting" the target. Studies have shown the principal radionuclides of concern from particle accelerators are H^3 and Na^{22} because they are the only ones that are both produced in significant abundance and are sufficiently leachable from the specific soil present at our site [Borak 1972].

The leaching process occurs when "clean" groundwater becomes activated as it filters through radioactive soil. When groundwater has established equilibrium with all of the soil solutes, the amount of radioactivity leached out of a material (clay, sand, till, rock) is a function of (a) the amount of water that has moved through the material, and (b) the grain size of the material. Not all of the produced activity is picked up and transported as the groundwater migrates. In particular, Na^{22} measurements made with Fermilab soils yielded 1% for dolomite, 7% for sand or gravel, and 15% for glacial till [Malensek 1993]. These asymptotic values are reached very quickly, as can be seen in Figure 3. In addition, leaching measurements give information on the average velocity of ion movement for each isotope. It is found that H^3 moves at the same rate as groundwater while Na^{22} moves 44% as fast [Borak 1972].

The equations for the fundamental laws which govern groundwater migration in a saturated continuum have been extensively investigated and are understood quite well, both theoretically and experimentally. For fluid flow, the process responsible for

moving fluid mass into and out of a volume element is simply flow in response to a potential gradient. The transport of a solute (under one-dimensional flow with three-dimensional dispersion) is a second order differential equation in space and time, but also incorporates the two crucial process of dispersion and radioactive decay:

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - v_x \frac{\partial C}{\partial x} - \lambda C$$

where C is the concentration, D the coefficient of dispersion, v the seepage velocity and λ is the reciprocal of the mean lifetime. Over the years, numeric and analytical models have been developed to solve the transport equation. The advent of powerful desktop computers, along with the experience gained in landfill design/operation have improved the quality of work in this field. Case studies provide a useful way to illustrate the application of models in understanding and predicting contaminant migrations. Models that are supplemented by carefully conceived field work provide an independent confirmation of subsurface conditions.

FERMILAB CASE HISTORY: THE CONCENTRATION MODEL

Prior to 1995, Fermilab used a simple model for groundwater migration. It took the concentration at the aquifer to be the sum of all the radionuclides produced and leached in the soil divided by the daily amount of water used by one person at their residence. Credit was taken for radioactive decay as it traveled through the glacial till to the aquifer at a rate of 2.2 meters/year for H³ and 1.0 meter/year for Na²².

A new "Concentration Model" was formulated by combining the results from an in-house group at the lab having expertise in production and leaching, and a hired consultant who had experience in groundwater migration. It was presented to the E S & H section; they made slight modifications, and it was adopted as policy by the management in December of 1994 [Cossairt 1994]. Although the model is quite general in its overall approach and state-of-the-art analytical methods were applied, to be most useful the parameters need to be derived from site-specific data. Information from over 250 boreholes at Fermilab was used, and a complete set of water levels were taken to determine the natural gradient. The consultant evaluated the hydrogeologic data, incorporated it into a computer migration model called "PATCH3D" and simulated how the concentration would evolve as the groundwater moved to the aquifer. This was only part of a comprehensive migration study which is shown in Figure 4. An initial "patch" migrates vertically through the till; it has an increased size when it reaches the till/dolomite interface because of transverse dispersion. Mixing takes place as the "patch" penetrates the boundary and changes direction. Finally, the "patch" migrates horizontally in the dolomite with three-dimensional dispersion. However, this paper will restrict its discussion to the first process--vertical migration in the glacial till.

The Concentration Model calculates an average initial concentration, C₀, in the zone of activated soil, then transports it to the aquifer where the concentration is C₀*R_i where R_i is the reduction factor for the ith nuclide. The equations are,

$$C_0 = \frac{N_p * (0.019 * S_{\max}) * K_i * L_i * (1 - e^{-t/\tau_i})}{1.17 \times 10^6 * \rho * w_i} ; \quad C_{\text{aquifer}} = C_0 * R_i$$

where

- N_p = number of incident protons per year,
- S_{\max} = maximum stars per cm^3 per proton produce in the soil,
- K_i = production fraction for the i th nuclide,
- L_i = leachable fraction for the i th nuclide,
- τ_i = the mean lifetime for the i th nuclide,
- w_i = the weight of water divided by the weight of soil that corresponds to a specific percentage of leaching for the i th nuclide,
- t = the irradiation time,
- ρ = density of soil,
- 1.17×10^6 = converts disintegrations per second into picoCuries per year,
- R_i = reduction factor while migrating from the till to the aquifer for the i th nuclide.

As already mentioned, there are only two radionuclides of interest that result from accelerator operations, $i=1$ for H^3 and $i=2$ for Na^{22} . A discussion of further details of each of the variables in the equation will highlight the model. It has these features:

- Given the chemical composition and geometry of the target material, Fermilab has two computer programs (CASIM and MARS) which use the Monte Carlo technique to calculate the inelastic interactions (the so-called star density) produced in a unit volume of soil per incident proton. Both programs follow the development of internuclear cascades and compute the star density as a function of location throughout the materials. The choice of which program to use depends primarily on the energy of interest (MARS has a much lower threshold) and the amount of computer time needed to obtain reasonable statistics.
- The average star density in the soil is related to the maximum star density through the numerical factor 0.019, which is characteristic of that found outside a beam absorber after integrating over a volume that essentially encompasses all the activity (the cutoff is made at 99% in each dimension).
- Multiplying by the number of protons and the production fraction gives the total amount of each nuclide that is created in the soil. The value of N_p is taken as an average over a three year period to moderate the cycle of operations.
- Of the total amount produced, a fraction of each radionuclide is leached from the soil by passing a given amount of water through it. The curve which relates the percentage leached from the soil into the water has already been briefly discussed and is shown in Figure 3. Essentially it is possible to extract the asymptotic value L_i for a ratio of weights, $w_1 = 0.5$ for H^3 and $w_2 = 1.0$ for Na^{22} .
- For a constant rate of production (i.e. a constant beam intensity), soil activity builds up over time to a saturation value, when the number of radionuclides

being produced per unit time equals the number being lost due to decay. The time dependence of this buildup is $(1-\exp(-t/\tau_i))$.

- To completely separate production from migration, the activation zone is excluded when determining the distance to the aquifer (which is at an elevation of 677 feet at Fermilab): $d = (\text{elevation at the bottom edge of the concrete} - 6 \text{ feet} - 677 \text{ feet})$. See Figure 5.
- The reduction factor R_i is a fit to the results of the computer program PATCH3D for a vertical seepage velocity of 0.15 meters/year.

A reality check for each of the processes considered must point back to measurements in the field for any model to have credibility. The leaching curve is already based on site specific data. Agreement between CASIM results and data are typically within a factor of two to three. PATCH3D compares favorably with the Gaussian Source Model of Huyakorn and AT123D for Yeh which evaluate test problems against data. Since the seepage velocity has the largest uncertainty and affects the migration calculation the most, considerable effort was made to find a value that was representative of the places radionuclides are created. A location known as AP0 is the target station that consistently receives the highest number of protons, so its characteristics were the ones used. The seepage velocity is a combination of gradient i , the hydraulic conductivity K , and the effective porosity n .

$$v = \frac{i * K}{n}$$

The consultant's report used data over an eight year period, 1984-1992, to determine a range for the gradient, $i = 0.28-0.56$. Using the moisture content of ten bore samples gave an effective porosity $n = 0.36-0.40$. Ten measurements of the hydraulic conductivity were made before construction began at AP0, covering the full range of depth into the glacial till--13 feet to 65 feet below the surface. K varied from $1.36E-8$ to $2.84E-7$ cm/sec [Cossairt 1994].

To be conservative, the extreme end of the ranges was taken for each variable to give a seepage velocity of approximately 0.15 meter/year. Using $v = 0.15$ meters/year for H^3 and 0.066 meters/year (44% of the tritium value) for Na^{22} , PATCH3D (includes dispersion) was run for several target stations which are at varying heights from the aquifer--8.5 meters to 17.7 meters. As a practical matter, the longitudinal coefficient of dispersion is about 1/10 the length of the flow path [Fetter 1988]. Transverse dispersion is usually about 10% of the longitudinal dispersion, but can be more like 30% if "transverse" is in the direction of gravity. Dispersion produces a mixing zone between the contaminated water and the native groundwater. It occurs because of mechanical mixing in the fluid; some water molecules and solute molecules travel more rapidly than the average linear velocity and some travel more slowly. The solute therefore spreads out in the direction of flow.

Taking the time dependence of buildup as $(1-\exp(-t/\tau_i))$ means the steady state is achieved after about 50 years of running for tritium. It is therefore not necessary to make calculations for longer running times. A family of "50 year curves" was made for

all of the lab's representative target stations, and because they have varying depths to the aquifer D_x changes about a factor of two. Target Station N is at the greatest distance, so its value of C_0/C (at the same distance) always remains higher than the other target stations. It is therefore appropriate, simplest, and safest to take the reduction factor R_i for Target Station N and apply it to all others. A fit for each radionuclide was made using the boundary condition $R(d=0.0) = 1.0$. It gives:

$$R(H^3) = \exp\left(-d(\text{meters})/3.5\right) ; \quad R(Na^{22}) = \exp\left(-d(\text{meters})/0.80\right)$$

A valuable by-product of the Concentration Model is the ability to calculate concentrations at sump locations. As Figure 5 illustrates, typically there is a gravel region with underdrains that surround an enclosure which collects water and directs it to a sump pit. Since water travels through gravel relatively quickly (as evidenced by the fact that sump pumps turn on shortly after a rainfall), it is appropriate to calculate the sump concentration with $t = 1$ year as,

$$C_0 = \frac{N_p * \langle S \rangle_{\text{gravel}} * K_i * L_i * (1 - e^{-1 \text{ yr} / \tau_i})}{1.17 \times 10^6 * \rho * w_i}$$

Note that $(0.019 * S_{\text{max}})$ has been replaced by $\langle S \rangle_{\text{gravel}}$, the star density averaged over the gravel region. L_i is the same as before for H^3 , but changes from 15% to 7% for Na^{22} .

APPLICATION OF THE MODEL TO AP0

The AP0 target station consists of a concrete enclosure 11.5 feet wide, with three-foot-thick walls and floor (see Figure 5). The steel shielding thickness within the target vault is typically 3 feet on the sides, 4.5 feet below and 5 feet above. Stone fill and gravel form a backfill layer that is typically 5 feet thick. This, together with underdrains, relieves the hydraulic pressure on the walls. Figures 6 and 7 show the results of PATCH3D under several conditions, (1) after 50 years of running, $D_x=d/10$, (2) after 25 years of running, $D_x=d/10$, and (3) no dispersion but 50 years of running.

The effect of dispersion can be seen dramatically in Figure 6 by comparing the no dispersion curve ($D=0.$) with those above it. In Figure 7, the no dispersion curve is dominated by radioactive decay--the short half-life of Na^{22} . In fact, the curving down of the concentration ratio from 25 years, (7.5 meters of travel for H^3 and 3.3 meters for Na^{22}) as compared to 50 years is due to the longitudinal dispersion. When the "front" moves by several years later, the concentration increases.

The AP0 geometry was input to CASIM. S_{max} and $\langle S \rangle_{\text{gravel}}$, are obtained from the same computer run by finding the maximum value at the bottom/side (whichever is highest) of the outside of the concrete for the former, and averaging over the gravel volume for the latter. To be consistent throughout the Model, the irradiation time is the time it takes to cross the 6 foot activation zone divided by the seepage velocity. [The lab policy as adopted in late 1994 took $t = \infty$.] The parameters used in the equations of the previous section are summarized in the following table:

Parameter	Units	Value	
Np(average of 1994-1996)	protons/yr	1.2E19	
S _{max}	stars/cm ³ -proton	2.26E-10	
<S> gravel	stars/cm ³ -proton	2.95E-11	
Soil density	gm/cm ³	2.25	
Distance to aquifer, d	meters	12.2	
Isotope		H ³	Na ²²
Radionuclide production	atoms/star	0.075	0.02
Leachable fraction (90%)	--	0.90	0.135*
Water fraction (90% leaching)	--	0.27	0.52
Irradiation time, t	years	12*	27*
Mean lifetime, τ	years	17.7	3.75
Reduction factor (exponent constant)	meters	3.5	0.80

* t is the time to cross the 6 ft activation zone; t = 1 for sumps and 0.135 changes to 0.063 (sand or gravel).

For H³ this gives C₀ = 2.44 pCi/ml, C_{aquifer} = 0.075 pCi/ml, and C_{sump} = 1.85 pCi/ml. The Na²² values are C₀ = 0.10 pCi/ml, C_{aquifer} = 2.4x10⁻⁸ pCi/ml, and C_{sump} = 0.076 pCi/ml. The tritium concentration limit of 20 pCi/ml in Class I groundwater is explicitly given in the regulations. No Na²² concentration limit in Class I ground water is explicitly stated in State of Illinois or Federal regulations. The guideline of 0.4 pCi/ml used at Fermilab is based on the Derived Concentration Guide value in DOE Order 5400.5. This order also specifies the concentration guidelines for surface water discharge (sumps), which are 2000 pCi/ml for tritium and 10 pCi/ml for Na²². Under the State of Illinois definitions, the aquifer under Fermilab is Class I Ground Water. It is independent of whether the water resource is presently being used or not. Another important point is the "non-degradation clause" and is usually interpreted to mean that nothing may be done that could result in concentrations that exceed the regulatory limits at the location where contamination enters the resource groundwater. The AP0 concentrations are well below the regulations/guidelines.

REFERENCES

- Borak, T.B. et al. 1972. The Underground Migration of Radionuclides Produced in Soil Near High Energy Proton Accelerators, Health Physics 23: 684-687.
- Cossairt, J.D. 1994. Environmental Protection Note 8. Fermilab Internal Report: 29.
- Fetter, C.W. 1988. Applied Hydrogeology. New York, NY: Macmillan, 394.
- Freeze, R.A. and Cherry, J.A. 1979 Groundwater. Englewood Cliffs, NJ: Prentice-Hall.
- Grobe, D.W. 1993. Site Environmental Report. Fermilab Internal Report: 69.
- Malensek, A.J. et al., 1993. TM-1851. Fermilab Internal Report: 23-24.
- Sudicky, E.A. et al., 1988. PATCH3D. Waterloo, Ontario, Canada: Univ. of Waterloo.

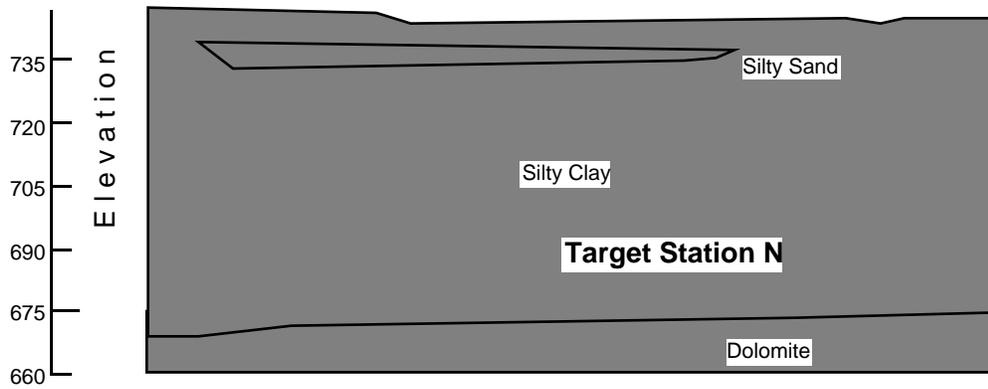


FIGURE 1

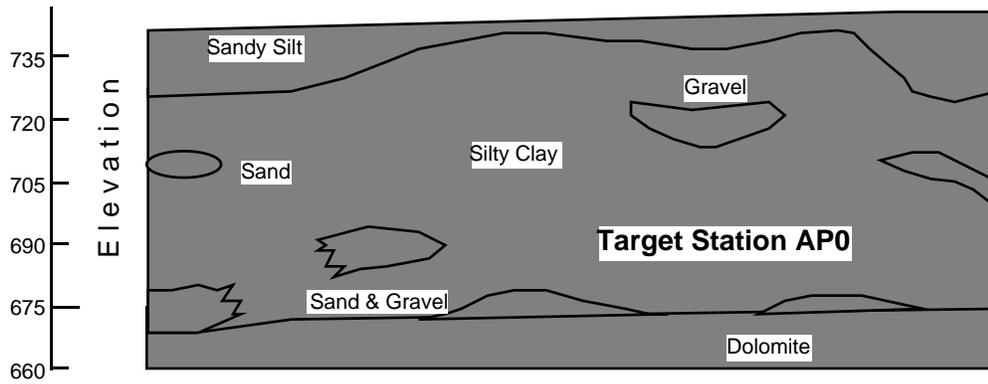


FIGURE 2

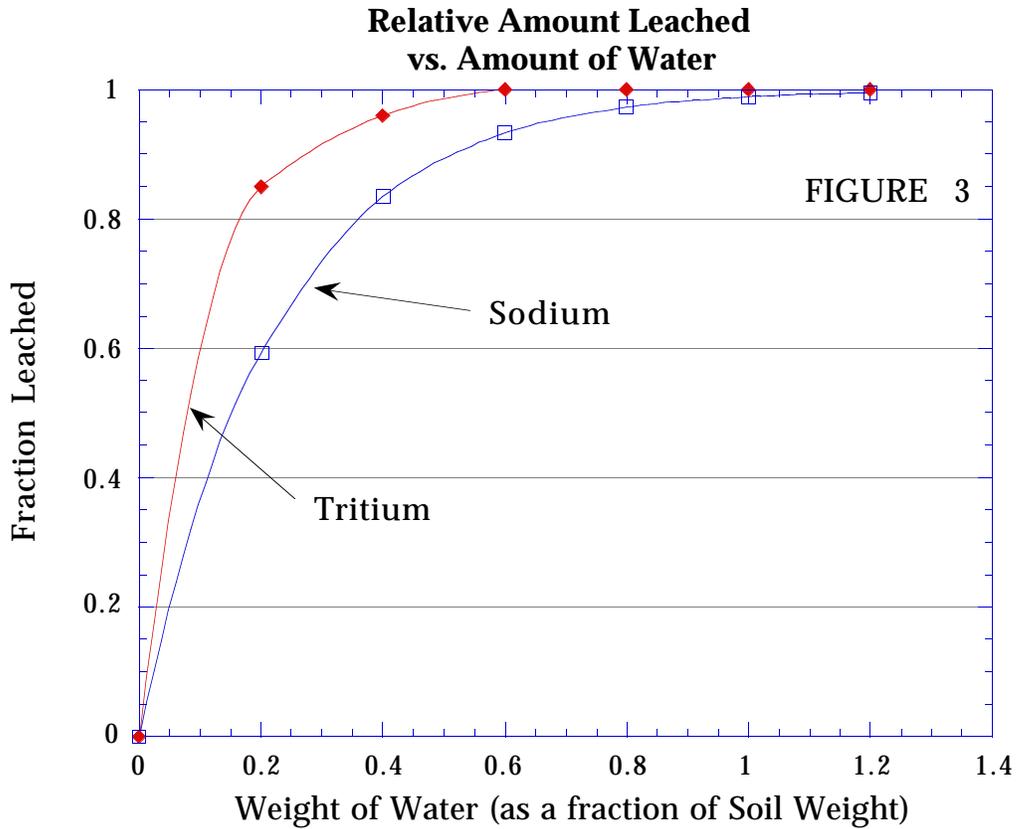


FIGURE 3

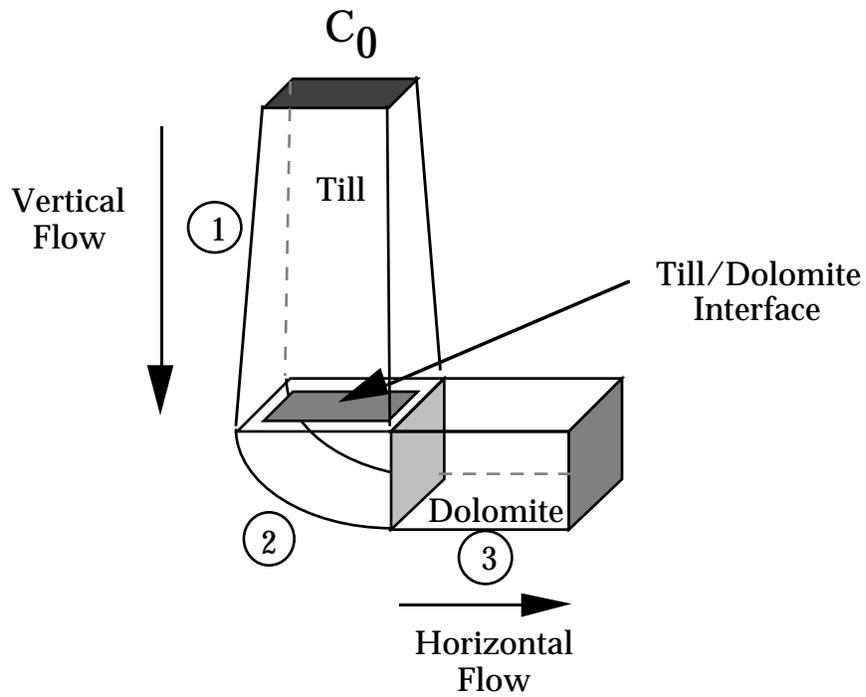


FIGURE 4 Schematic Illustration of the Concentration Model

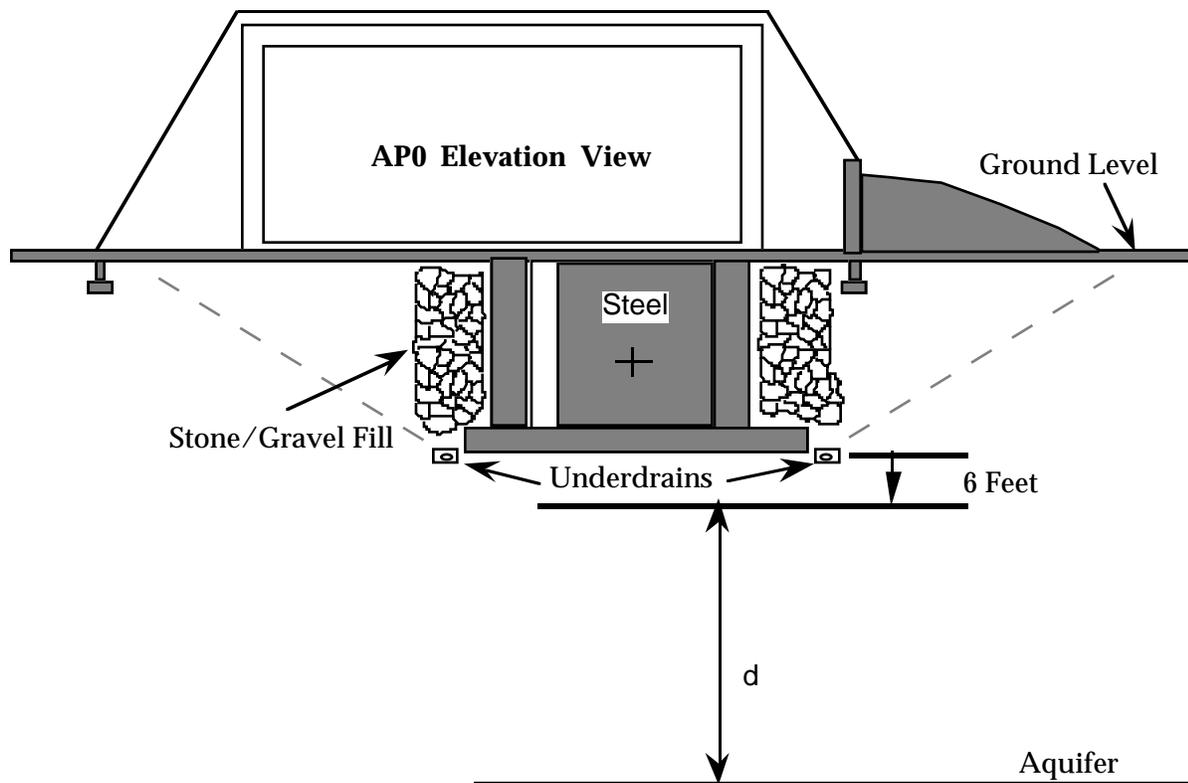


FIGURE 5 Elevation Profile of Target Station AP0

