Fermilab
ES&H Section

EP Note 17
THE CONCENTRATION MODEL REVISITED

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June 24, 1999

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I. Introduction

During 1997 a number of committees set up by the Directorate were charged with, among other things, reviewing calculations of soil and groundwater in the vicinity of various target stations around the Laboratory. The final reports from some of the committees (Ho97, Ed97) recommended clarification and modification of aspects of the Concentration Model (Ma93), which is currently the methodology in use at Fermilab for estimating radionuclide production in soil and migration in the groundwater. This document represents an attempt to answer some of the questions raised in the various committee reports, clarify the procedures for determining the parameters of the Model (and suggest the use of defaults when precise information is not available), and discuss the uncertainties in the model.

II. The Concentration Model

The concentration of a radionuclide i in the ground water in soil or other medium immediately outside the region of a loss of accelerator beam (such as a beam targeting enclosure) is given, in units of pCi/ml, by

$$C_{0i} = \frac{N_p \cdot < S > \cdot K_i \cdot L_i}{(1.17\times10^6)\cdot \rho_s \cdot w_i}[1 - e^{-t_{ir}/\tau_i}]. \qquad (1)$$

Here N_p is the number of protons per year at the source, <S> is the star density in stars/cm^3 averaged over a volume surrounding the source out to an appropriate boundary, K_i is the production yield of the isotope i in atoms/star, L_i is the fraction of isotope i produced in the soil that is leachable by water, ρ_s is the density of the soil in g/cm^3, w_i is the ratio of the weight of water to the weight of soil that corresponds to the leaching fraction for the ith nuclide, t_{ir} is the irradiation time, and τ_i is the mean-life of the ith radionuclide. The numerical factor converts disintegrations/sec to pCi and years to seconds. The final concentration at a point down-gradient in the aquifer is

$$C_{fi} = R_i\, C_{0i}, \qquad (2)$$

where R_i is the reduction factor for the ith nuclide due to vertical transport through the glacial till deposits and horizontal transport in the aquifer. In TM-1851 (Ma93), R_i is broken into a product of three reduction factors, R(till), R(mix), and R(dolomite). As discussed in EP Note 8 (Co94), R(mix) and R(dolomite) will usually be taken to be unity, so that the reduction factor that gives the final concentration is R_i = R(till).

In the original description of the Concentration Model in TM-1851, and in the subsequent Environmental Protection Note EP-8, recommendations are given for the values of the parameters specified in Eqs. 1 and 2 above. Woodward-Clyde Consultants (WCC93) did model calculations simulating radionuclide transport that incorporated average site geologic, hydrologic and geochemical conditions. Average hydraulic conductivities were determined and with estimated vertical gradients led to a suggested vertical seepage velocity. Based on this, a conservative "one size fits all approach" was recommended for all target stations within typical Fermilab soil (a mostly silty clay glacial till).

Specifically, the calculation was based on the saturation activity for the given radionuclide (that is, the term within the brackets in Eq. 1 was replaced by unity) that is produced by the average number of protons delivered to the target per year. The star density <S> was an average of the maximum star density S_{max} in the unprotected soil outside of the targeting enclosure over a volume that contains 93% of the total radioactivity; this was out to where the star density fell to 1% of S_{max}, and was called the "99% volume" in Ma93. Here the dependence of star density fall-off on r and z were based on a CASIM (Va75) calculation for 1000 GeV protons incident on a solid concrete cylinder. The values of K_i for the two leachable isotopes ^3H and ^{22}Na in soil are taken to be the values that have traditionally been used at Fermilab (see, e.g., Go78), as is the soil density ρ_s. The recommended value of the leachability fraction was taken to be 0.9 for ^3H and 0.135 for ^{22}Na, and the weights of water relative to the soil weight needed to obtain those fractions were taken from curves that arose from controlled columnar leaching studies by Baker (Ba75) with sand and gravel (not glacial till).

The reduction parameter R_i for both leachable nuclides for glacial till was based on the results of the Woodward-Clyde calculations with the computer program PATCH3D (Su88), a three dimensional analytical solution for radionuclide transport in ground water. In this program a 3.7 m by 3.7 m "patch", approximately representing the size of the region over which the star density was averaged[1], was transported vertically through the soil. This was done at each of six targeting stations at the Laboratory with, as much as possible, site specific values of dispersivity and for two average vertical groundwater seepage velocities, 2 and 40 cm/year. Later, a value of 15 cm/year was chosen as the representative seepage velocity to be used in all cases. The PATCH3D calculation was parametrized as an exponential in distance to the aquifer (Co94) for use in all future analyses.

In the present report we discuss each of the parameters in Eq. 1 separately. Suggested values for these quantities are given, and where appropriate the uncertainties in these are discussed. In general, in order to minimize uncertainties we strongly recommend that for all future construction the local hydrogeological parameters such as hydraulic conductivity, porosity, and hydraulic gradient necessary to characterize a specific targeting location be obtained from analyses of soil or rock borings and water levels in wells. It is further urged that radionuclide production cross section, leaching fraction, and

[1] This patch size arises from projecting the "99% volume" in each dimension onto the r-z plane. See Ma-93.

the conversion of radionuclide concentration in soil, or other media, to that in water be determined experimentally by activation studies of irradiated samples, if at all possible.

III. The Parameters of the Model

1. N_p and the Buildup Factor

The number of protons per year incident on the source target station should be representative of the average annual proton delivery. It has previously been suggested (Co94) that this average be taken over a three-year period. The quantity in the square brackets in Eq. 1 is the buildup factor for the production of a given radioisotope with mean life τ_i. The determination of a value for the irradiation time t_{ir} must be based on judgment as to a reasonable lifetime of the particular facility. Irradiation of the target such that a particular radionuclide activity will saturate is clearly the conservative choice but is, in all likelihood, an unreasonable assumption. For example, to saturate the ^3H activity takes about 80 years of constant irradiation.

2. The Average Star Density <S>

The star density in the unprotected soil region outside of the targeting enclosure can be determined from a computer program for the simulation of hadronic and electromagnetic cascades in matter. At Fermilab, CASIM has traditionally been used in these model simulations. Recently the Monte Carlo program MARS (Mo95) has become the primary shielding code. The output supplies tables and contour plots of star density, along with uncertainties, as a function of rectangular or cylindrical coordinates. For most applications at hadron accelerators, the output of these two codes is generally in acceptable agreement.

As mentioned, TM-1851 (Ma93) prescribed a "one-size fits all" cookbook value for the average star density, <S>. This prescription, discussed above, based as it is on a solid cylindrical concrete dump, does not necessarily describe those contours that arise from beam incident on other target configurations. Instead, the procedure should be modified as follows.

As before, the average star density <S> can be obtained from star density contour plots in r and z, based on MARS, by going "out" in r and z from their values for the star density S_{max} at the boundary right outside the enclosure in which the beam loss occurs to those values of r and z at which S has dropped to 1% of its maximum value. The total stars in this volume can be obtained by multiplying the stars in each (r,z)-bin by the volume of each bin and summing over the number of bins within the region. Then, <S>, the value to be used in Eq. 1, is equal to the total stars within that region divided by the volume enclosed by the limiting values of r and z. The calculation should be performed such that star density statistical accuracy is acceptable at all values of r and z, if at all possible. If necessary, however, an extrapolation out to the 1%-of-S_{max} level in r and z from small values of both coordinates where S-values are well-defined may be performed. Typical

bin-to-bin statistical uncertainties in star densities calculated with both CASIM and MARS are 30%. This will lead to uncertainties of about 10-15% for total stars, and on the value of <S>.

3. Leaching L and w, and Radionuclide Production K

The preferred values for the quantities L_i, w_i and K_i for the two radioisotopes ^3H and ^{22}Na that enter into expression Eq. 1 for initial radionuclide concentration should be determined from activation and leaching experiments on soil (or other media) samples obtained at the particular targeting locations. The media samples should be subjected to chemical analysis, including the determination of the fractional amount of water.

Borak, et al (Bo72) describe in some detail the techniques and considerations that are important to the determination of the induced activity in media samples that are irradiated in hadron beams, as well as to the associated batch process leaching studies. In particular, for example, it is important to preserve the water present in the samples during the irradiation time. Wehmann and Childress (We99), on the basis of the Borak, et al results for Fermilab soils, have suggested that the leaching of ^3H from irradiated soil or rock involves a different mechanism than that for ^{22}Na. The sodium apparently dissolves in the water, and the process can go in the "other direction" when, e.g., radioactive leach water is batch processed with non-irradiated soil. With ^3H, on the other hand, We99 suggest that it enters the leach water in two ways; one, from the tritium produced directly in the irradiated water in the soil, and two, from entrapment in the water in the soil due to triton recoil after production in the soil itself. This process is not reversible; that is, as was observed by Borak, et al, none of the ^3H in the radioactive leach waters was transferred to non-irradiated soil when batch processed together. In other words, the leachable (or transferable) activity appears to be associated with the amount of water in the soil (or other media) at the time of irradiation.

For ^{22}Na, the value of L_{22} in the leach water as a fraction of the activity in the irradiated sample can easily be obtained by use of batch processing techniques because it is straightforward to measure its activity in both water and the sample. For ^3H, on the other hand, the activity can not be measured directly in the sample because of its low end point beta energy. Thus, measurements of tritium in the "leach" water represents only a determination of the tritium that can be transferable (or "leached"). In effect, then, what is measured in a batch process leaching study is the product of K_3, tritium production in atoms/star, and L_3 together.

In batch processing studies the fraction of weight of water to weight of sample w_i is usually not determined. The transport of radionuclides through soil or rock occurs, generally speaking, under conditions in which the medium is saturated with water. Since the maximum amount of water contained in saturated media occurs when the voids or pores are filled, it is this water that is available for the leaching of the radioactivity. As mentioned by Borak, et al (Bo72), and stated above, particularly for the case of ^3H, the

leachable activity is associated with the amount of water in the soil at the time of irradiation.

Porosity is defined as the fraction (by volume) of the soil or rock that is void of material (see, e.g., Fe88). It is reasonable therefore to use the porosity for the particular medium to determine the value of the water content of the soil. Thus, with W_w and W_s equal to the weight of water in the soil sample and the weight of the "wet" sample, respectively, and V_w and V_s the equivalent volumes,

$$w = W_w/W_s = V_w \rho_w/(V_s \rho_s) = V_v/(V_s \rho_s) = n/\rho_s, \qquad (3)$$

where ρ_w, the density of water, is set equal to1 g/cm^3, V_v is the volume of the void, which from above is equal to V_w, and $n=V_v/V_s$ is the fractional porosity. Bulk (measured) porosity ranges from 0.2-0.35 for mixed sand and gravel, 0.33-0.6 for clay, 0.1-0.2 for glacial till, and 0.01-0.3 for limestone and dolomite (Fe88), although effective porosity[2] may under some circumstances be even smaller. For fine grained materials, such as clays and glacial tills, effective porosity is equal to bulk porosity (Fe98).

It is proposed, therefore, that in the calculation of the initial concentration, Eq. 1, the measured values of K_i and L_i (or the product L_iK_i), if available, be used for each radionuclide i. For w_i the value associated with the porosity of the medium (see Eq.3 above) should be used. The quantity K_i for a given radionuclide can be obtained from the macroscopic cross section for production of the nuclide by dividing by the total inelastic cross section for hadrons incident on the medium. The macroscopic cross section is directly related to the measured activity of the sample (see Bo72, e.g.).

If, on the other hand, neither the leachable fraction nor the nuclide production K_i has been directly measured for the particular medium at the targeting location, then, following Wehmann and Childress (We99) and Malensek, et al (Ma93), the measurements of Borak, et al for Fermilab glacial till should be used. That is, for ^{3}H the product of K_3L_3 should be taken as 0.075 atoms/star, while for ^{22}Na, K_{22}=0.02 atoms/star and L_{22}=0.15. For both nuclides take the value of w_i as 0.14, which is the fraction (by weight) of water in the glacial till sample, as shown in Table 1 in Bo72. The uncertainties in these K_i values have been estimated in the AP0 Review Committee Report (Ho97). Statistical errors are on the order of ±(30-40)% with systematic uncertainties that are hard to quantify. Finally, it should be emphasized again that, as mentioned, it is highly desirable to determine those parameters characteristic of each targeting location by sampling and measurement. The default values discussed here should be used only as a last resort.

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[2] Fluids flowing through pores may not move through the entire volume of pore space. There may be non interconnected as well as dead end pores. Effective porosity is defined as the porosity available for fluid flow. See Fe88.

4. Propagation to the Aquifer, R_i

The movement of radionuclides in ground water is controlled by advection (bulk movement of the ground water) and dispersion. Woodward-Clyde (WCC93) recommended the use of a three-dimensional analytic solution computer program called PATCH3D (Su88) to solve the advection-dispersion equation. In their report they presented the results for the transport of ^{22}Na and ^{3}H through soil to determine R(till) for each isotope at six targeting locations at the Lab. As mentioned, the calculations were done by transporting a uniform rectangular patch (3.7 m by 3.7 m) containing the initial radionuclide concentration in a direction perpendicular to the patch downward to the dolomite aquifer utilizing a range of values of the characteristics for the glacial till based on a site wide average obtained from bore hole measurements and water levels in monitoring wells.

The program PATCH3D is available for use at Fermilab. The input requires, along with the properties of the particular radionuclide and distance to the aquifer from the source, values of the vertical seepage velocity within the medium and of both longitudinal (vertical) and horizontal dispersivity. (Molecular diffusion is very much smaller and can be neglected). Ideally, the program should be run with such input parameters derived from local hydrogeologic properties of the medium such as hydraulic conductivity, porosity and gradients obtained from bore hole and water level data in the vicinity of the targeting station, and with a rectangular patch source based on the size of the projected area derived from the volume over which the star density is averaged. If necessary, when the medium through which the water and contaminant are flowing consists of layers with different hydrogeologic properties, the program can be run for each layer separately (taking any change in patch size due to the dispersivity into account) with the final reduction factor R a product of the R's of the separate layers[3]. It should be noted that it is necessary to design a facility (that is, a new target location) based on the maximum activity of the radionuclides reaching the aquifer, even though this situation may not occur until several years after accelerator operations at the given location have been terminated.

According to Woodward-Clyde, and others, it is not possible to accurately calculate dispersivities from the measured soil properties. Empirical estimates of dispersivity based on total travel distance in the geologic medium are usually made. Therefore, in PATCH3D calculations, longitudinal (vertical) dispersivity should be taken as 0.1 of the distance to the aquifer, and the horizontal dispersivity as 0.1 of the longitudinal dispersivity. While we strongly recommend that vertical seepage velocities be calculated from analyses of local soil borings particularly for any new construction, a default value of 0.15 m/yr has been well documented (see Co94, Appendix 1) if such soil boring data does not exist.

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[3] See, e.g., some recent calculations (Va98) at a location near the Booster extraction point to the Main Injector.

IV. Discussion

1. Concentration in Sumps

At many targeting locations around Fermilab underdrains located in granular fill at small distances below the concrete enclosures can collect radioactive water leached from the media and transport it to surface sumps and retention pits. Ignoring possible non-steady-state behavior arising from the fact that this region may not satisfy saturated flow conditions, Eq. 1 can be used for calculating radionuclide concentrations in sump water with redefinition of some of the parameters discussed in Sects. II and III. The AP0 Review Committee Report (Ho97) has identified two cases - annual flushing of activity into the underdrains and sumps, and cumulative buildup over several years prior to flushing. Both have been discussed quite extensively in the AP0 Report, and the annual flushing case is thought to be most appropriate since sumps run many times during the course of a year.

For sumps, then, for this case, the concentration can be calculated as in Eq. 1 with t_{ir} taken as equal to 1 year (at most). The average star density <S> is calculated as discussed in Sect. III.2 above but with S-values obtained by going out in r and z from the values at the location of S_{max} to those values of r and z at the elevation of the underdrain, <u>not</u> to those coordinates at which S has dropped to 0.01 S_{max}. Leaching and production parameters are, as before, those appropriate for the given medium.

The calculation of expected radionuclide concentration in sump water for new construction projects allows an estimate of whether the shielding is sufficient to keep water discharges below the regulatory limits for surface water. It is in this spirit that such calculations are useful. Thus, for example, if the concentration estimate is over the limit then the sumps and underdrains might be relocated so that the regulatory limit on concentration is met.

On the other hand, sumps should not be used as "monitoring wells" during running periods. It is not always clear whether all the water flowing into the sumps is directly related to beam targeting at that specific location. Thus, spills of closed loop cooling water systems as well as contributions from adjacent target locations and other sources of input water can contribute to measured concentrations. It was in fact mostly the uncertainties in the comparisons between calculated and sampled sump concentrations that prompted this current reexamination of the Concentration Model.

2. Material Parameters for use in MARS

The computer program CASIM required a series of parameters, input by the user, that describe the medium in which the hadronic and electromagnetic cascades develop. A report, TM-1898, was prepared (Ma94) based on input to CASIM, that describes procedures for obtaining the appropriate parameters for an arbitrary material be it an

element, compound or mixture. In MARS, on the other hand, most of the needed parameters are calculated from standard algorithms given only the atomic weights, atomic numbers, and densities of the elements within the material.

3. Summary

It should be emphasized that in this report the basic structure of the Concentration Model remains the same. That is, no attempt is made to derive the radionuclide concentration in the aquifer or some other location below the source from a more fundamental description based on continuous star production and leaching processes within the framework of the three dimensional flow and contaminant transport equations associated with the ground water system. The uncertainties in the geological parameters probably justify the model approach over the increased complexity of a more basic description.

As seen in Sect. III, most of the uncertainty associated with the parameters of the model arises in connection with the leaching of the radionuclides from the soil or other medium. It is important therefore to perform realistic measurements followed by activation analysis on the various Fermilab soils and rock in order to determine the values of K, L and w for each radionuclide. It is strongly recommended that such studies be initiated in the near future.

Finally, it is appropriate to emphasize again that hydrogeological properties characteristic of the specific targeting or beam loss location be used to determine the input parameters (including the true distance to the dolomite aquifer from the source "patch") for the PATCH3D calculation. These properties should be obtained from analyses of local soil or rock borings and water levels in wells. This especially holds true for any non-standard location such as NuMI, where the target hall is directly in the dolomite aquifer. In fact, in such cases it is especially important that the values for all of the parameters in Eq. 1 be determined from measurements within the specific medium and not from any "one-size-fits-all" formula.

The authors would like to acknowledge, with thanks, the comments and suggestions from Kamran Vaziri in the preparation of the final version of this report.

V. References

Ba75 S. I. Baker, "Soil Activation Measurements at Fermilab." Proceedings of the Third Environmental Protection Conference, Chicago, Il.,US Energy Research and Development Administration Publication ERDA-92, Vol. 1, Sept. 1975.

Ba97 S. I. Baker, J. S. Bull, and D. L. Goss, "Leaching of Accelerator-Produced Radionuclides." Health Physics 73, 504-508 (1997).

Bo72 T. B. Borak, M. Awschalom, W. Fairman, F. Iwami, and J. Sedlet, "The Underground Migration of Radionuclides Produced in Soil Near High Energy Proton Accelerators." Health Physics 23, 679-687 (1972).

Co94 J. D. Cossairt, "Use of a Concentration-Based Model for Calculating the Radioactivation of Soil and Groundwater at Fermilab." Internal Report Environmental Protection Note 8, Fermi National Accelerator Laboratory, Batavia, Il., December 1994.

Ed97 H. Edwards, T. Murphy, T. Toohig, F. Turkot, A. VanGinneken, and A. Wehmann, "C0 Main Ring-Tevatron Beam Dump." Review Committee Report, Fermi National Accelerator Laboratory, Batavia, Il., December 5. 1997.

Fe88 C. W. Fetter, Applied Hydrogeology, 2nd Ed. Macmillan Publishing Company, New York, 1988.

Fe98 C. W. Fetter, Private Communication, 1998.

Go78 P. J. Gollon, "Soil Activation Calculations for the Anti-Proton Target Area." Internal Report TM-816, Fermi National Accelerator Laboratory, Batavia, Il.,1978.

Ho97 C Hojvat, W. Freeman, F. Lange, A. Leveling, and A. Malensek, "AP0 Target Station." Review Committee Report, Fermi National Accelerator Laboratory, Batavia, Il., June 9,1997.

Ma93 A. J. Malensek, A. A. Wehmann, A. J. Elwyn, K. J. Moss, and P. M. Kesich, "Groundwater Migration of Radionuclides at Fermilab." Internal Report TM-1851, Fermi National Accelerator Laboratory, Batavia, Il., August 1993.

Ma94 A. J. Malensek and A. J. Elwyn, "CASIM Input Parameters for Various Materials." Internal Report TM-1898, Fermi National Accelerator Laboratory, Batavia, Il., July 1994.

Mo95 N. V. Mokhov, "The MARS Code System Users Guide, Version 13(95)." FN-628, Fermi National Accelerator Laboratory, Batavia, Il., April 1995; http://www-ap.fnal.gov/MARS.

Su88 E. A. Sudicky, T. D. Wadsworth, J. B. Kool, and P. S. Huyakorn, PATCH3D-Three-Dimensional Analytic Solution for Transport in a Finite Thickness Aquifer with First-Type Rectangular Patch Source. Prepared for Woodward Clyde Consultants, HydroGeologic Inc. Herndon, Va., January 1988.

Va75 A. VanGinneken and M. Awschalom, "Hadronic Cascades, Shielding, Energy Deposition," High Energy Particle Interactions in Large Targets, Vol. 1, Fermi National Accelerator Laboratory, Batavia, Il., 1975.

Va98 K. Vaziri and P. Kesich, "PATCH3D Reduction Factors for Tritium Migration in Booster Soil." Memorandum to Peter Kasper, Fermi National Accelerator Laboratory, Batavia, Il. July 1998.

WCC93 Woodward-Clyde Consultants, Summary of Radionuclide Transport Modeling for Ground Water at the Fermi National Accelerator Laboratory, Batavia, Il., Project 92C3073, Chicago, Il., August 1993.

We99 A. Wehmann and S. Childress, "Tritium Concentration in the Dolomitic Rock Adjacent to NuMI Beam Tunnels." Internal Report TM-2083, Fermi National Accelerator Laboratory, Batavia, IL, May,1999.