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Tritium Production in the Dolomitic Rock Adjacent to NuMI Beam Tunnels

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Abstract

This paper is a discussion of tritium production levels we might expect in the Dolomitic rock adjacent to NuMI beam tunnels.

Contents

1 Introduction

In order to assess the tritium concentration to be expected in NuMI monitoring wells we need both a model for tritium production in resource groundwater and some idea of the motion of the groundwater in the vicinity of the beam tunnels. Both these subjects have received attention in earlier papers and reports (References [1] & [2] & references cited therein).

In this paper we provide a new interpretation of tritium production, which we believe is more consistent with existing data. In developing this, we have closely examined data for tritium production in glacial till and in rock samples. A review of this data has led us to a model which provides an interpretation of tritium produced in either material, as well as an explanation of the anomolously low tritium production seen in a NuMI Dolomite sample irradiation test.

We then compare tritium production expectations, under static conditions, with earlier NuMI calculations and those for a CERN design. Finally, we point out that groundwater in the rock surrounding the NuMI tunnels should not be static (in the time period of extended NuMI beam operation), and we present our insight regarding the expected benefits (i.e. lower radionuclide concentrations) of the motion of the groundwater.

2 Tritium Production in Groundwater

The paper by Borak et. al. [3] describes the results of irradiation of soil samples near two proton synchrotrons[1]. The irradiation resulted in induced radioactivity, which was measured by radiochemical analysis. In addition, studies were made to see which of the radionuclides could be leached by water.

A careful reading of the Borak et. al. paper convinces us that tritium is a special case. Besides the fact that the soil samples could not be directly counted for ^{3}H (due to the low beta endpoint energy (0.02 MeV)) their paper found that the transferable ^{3}H appears to be associated with the amount of

[1]The Alternating Gradient Synchrotron (AGS) at the Brookhaven National Laboratory (BNL) and the Zero Gradient Synchrotron (ZGS) at the Argonne National Laboratory (ANL).

water in the soil at the time of irradiation. They also found that a sample of well water simultaneously exposed at the BNL AGS acquired a tritium activity that was 1/3 the activity of the water in the soil samples. These observations lead us to conclude that ionized tritium is produced from both soil molecules and the oxygen atoms in water. The tritium that is produced from soil molecules, and which becomes bound in a water molecule, has engaged in a two-step process—production and then entrapment. The typical range of the ionized tritium[2] is significantly less than 1 cm (see Figure 1); therefore the soil molecule and water molecule must be in proximity for entrapment in water to occur. Tritium produced by spallation from the oxygen atom in a water molecule, which ends up bound in a water molecule, also results from a two-step process. If one assumes that this two-step process is the only method by which tritium ends up bound to a water molecule, it would explain the Borak et. al. observation that none of the ^{3}H in the radioactive leach waters was transferred to non-irradiated soil when the two were batch processed together[3].

The other radionuclide, ^{22}Na, found to be important in the Borak et. al. paper can be leached[4] from irradiated soil by non-radioactive leach water, and does go in the other direction when radioactive leach water is batch processed with non-irradiated soil. Borak et. al. calculate the distribution coefficient, K_d, for ^{22}Na, and explain how its value is associated with the retardation of the movement of ^{22}Na with groundwater. Because tritium was found by Borak et. al. to have a distribution coefficient, K_d, with value zero, it suffers no retardation effect and travels with the velocity of the groundwater.

In the case of tritium, the transferable tritium is present in the water in the soil samples at the end of the irradiation; the batch processing with "leach water" mixes the "leach water" with the water in the soil sample and tritium enters the "leach water" by simple mixing[5]—not processes involving

[2]From the PDG booklet [4]we can find a value for $\frac{R}{M} = 1\frac{g}{cm^2}\,\mathrm{GeV}^{-1}$ for the range of a heavy charged particle in Carbon at $\beta\gamma$ (= p/Mc) $= 2.5 \times 10^{-1}$. For tritium M = 2.817 GeV/c^2. This works out to be a range of 2.8 g/cm^2 in Carbon for ionized tritium having an energy of 88 MeV.

[3]A corollary is that a leaching process does not remove a substantial amount of tritium from soil (or from rock). Water which becomes tritiated has to be present in the sample during the irradiation.

[4]This leaching process is presumed to be chemical in nature. The sodium becomes a dissolved ion in water.

[5]Borak et. al. used a quantity of "leach water" that was 10 times the weight of their

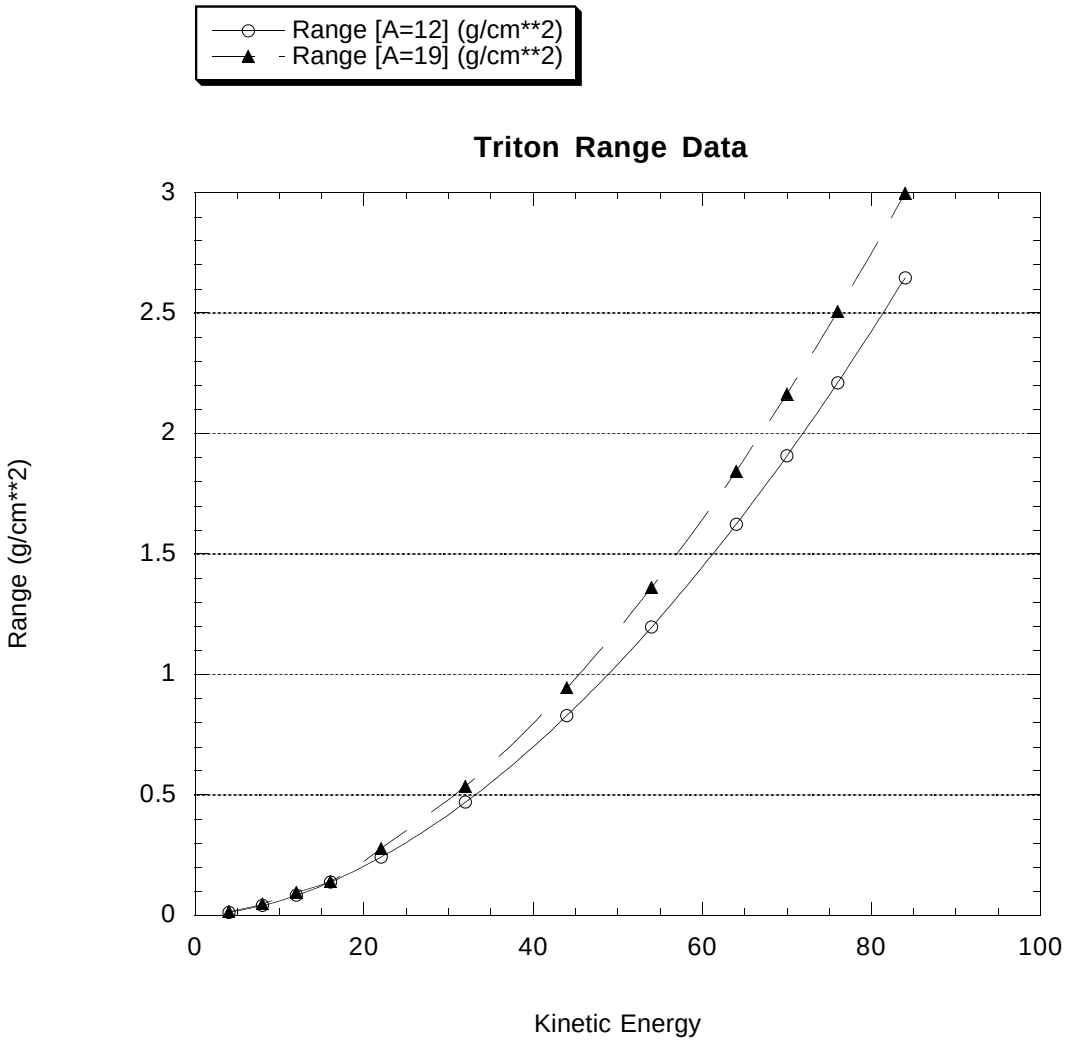

Figure 1: Triton range in Carbon (A=12) and in material with A=19 (close to soil weighted A value of 20.51). Units are g/cm^2 and MeV. The values come from reference [5] (courtesy of Alex Elwyn).

any tritium bound to soil molecules. With this point of view in mind, it is extremely important for a tritium measurement to preserve the water present in the sample at the time of irradiation.

Borak et. al. [3] irradiated a sample of well water at the BNL AGS, together with the soil samples denoted as B-1, B-2, B-3 (three depths from borings made along the proposed neutrino beam line). They also irradiated a sample of glacial till at the ANL ZGS; this was denoted as sample A-1 and came from beam elevation in the vicinity of the main ring injection-extraction gallery (approximately 20 ft below ground level[6]).

Borak et. al. observed that ^{3}H is produced in both water and soil. In the case of soil, the water and the soil are intimately mixed. In rock, a good portion of the water resides in fractures in the rock. To obtain the rate of tritium production in rock, one can expose rock samples to a monitored hadron flux—such as was done for NuMI[7].

2.1 Interpreting the Borak et. al. water sample activity

As already indicated, Borak et. al. tells us that the water sample irradiated simultaneously with samples B-1, B-2, & B-3 acquired a tritium activity that was 1/3 the activity of the water in the soil samples. We can interpret this to mean that the water sample was sized and exposed such that its tritium

samples. They observed that, once corrected for dilution, the "leach waters" accounted for all of the ^{3}H measured by the bake out process. Baker et. al. [6] used a quantity of "leach water" that was equal in weight to the weight of their samples, and did not account for all of the ^{3}H from their bake out measurement in all cases. The amount of water used may have been a factor in how much tritium could be removed by the "leaching" process. Had Baker et. al. used the larger quantity of water, the fraction of tritium "leached" after 1 hour of stirring might have been significantly higher—particularly for the sample they describe as "Fermilab soil".

[6]It is indicated in Table 4 of Borak et. al. that Sample B-3 was taken at a depth of 15-22 feet; that sample is characterized as "Gray clay". Sample B-1 was at depth 3-6 feet and is characterized as "Gray sandy clay"; sample B-2 was at depth 6-12 feet and is characterized as "Red sandy clay".

[7]The water that would be found in fractures should also be included appropriately in the exposure. It is questionable that the measurements done for NuMI took proper account of water that would have been found in the rock fractures. Dave Boehnlein has confirmed that the rock samples used had been left unsealed in a dry environment for months prior to irradiation. It is presumed that much of the original water contained within the samples evaporated away in this dry environment.

activity was due mostly to production of tritium in the ^{16}O atoms in the water molecules. It is our hypothesis that there was a limited amount of other material close enough to all the water molecules to have tritium produced in that material and then displace a hydrogen atom in nearby water—such as there is in the case of water dispersed in the pores of soil. The near presence of other, more dense material could significantly impact the results seen.

3 Normalized activity from Borak et. al.

In Table 4 of Borak et. al. [3] the values for tritium activity were normalized both to the amount of soil and to the amount of water contained in the soil. The values normalized to the amount of water in the soil varied significantly less from sample to sample than did the activity values normalized to the amount of soil. The activity data is plotted in Figure 2. The authors comment that this observation verified their conclusion that the transferable ^{3}H appears to be associated with the amount of water in the soil at the time of irradiation.

The Borak et. al. "leaching" measurements utilized 10 parts by weight of water to one part soil. If leaching from soil to water had a major role in the resultant tritium levels in water, we would expect very different distributions for the plots in Fig. 2. The expectation would be for leached activity levels normalized to grams of soil which would be largely independent of the % water fraction present during exposure. Similarly, when plotted normalized to the % water fraction in the soil during activation the distribution would vary inversely with the % fraction of water in the sample. The small variation (20%) seen in the lower left plot of Fig. 2 has a slope much smaller, and in the opposite direction from, that which would be expected for ^{3}H activity due to a leaching mechanism.

The ^{3}H activity (at saturation) per g of H_2O averaged for the four soil samples in Table 4 of Borak et. al. is 1.4×10^{-1} $\frac{pCi}{g}$. This is for a unit flux of hadrons (units cm^{-2} sec^{-1}), with an energy threshold of 30 MeV.

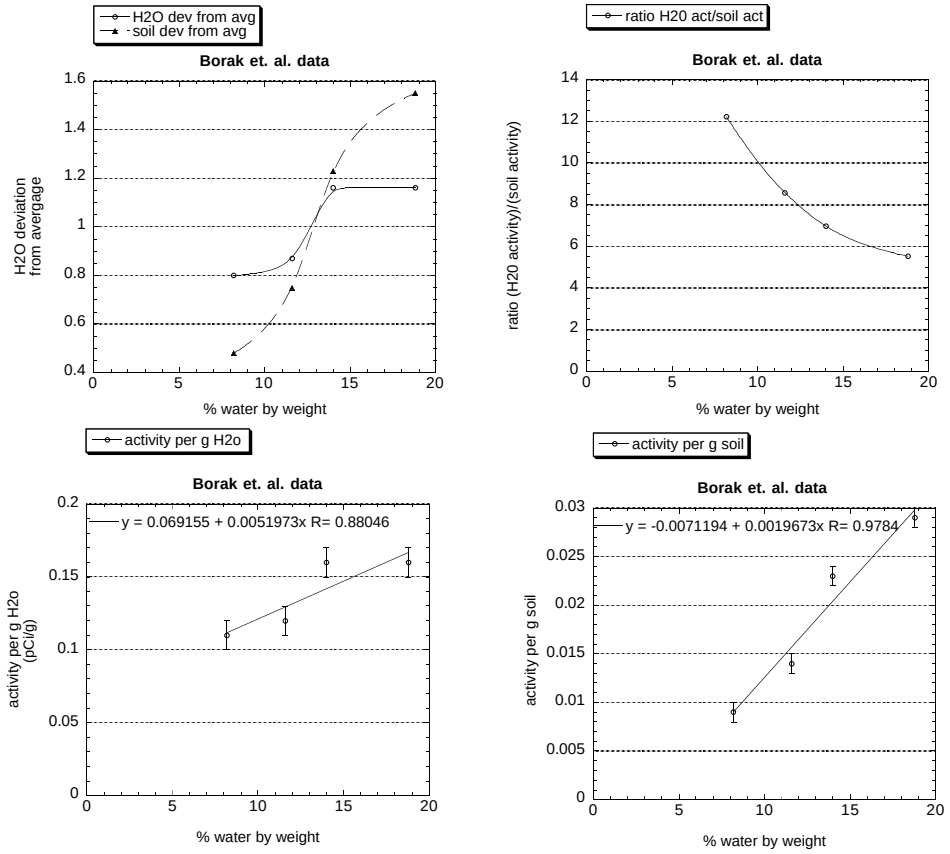

Figure 2: When tritium activity in the four Borak et. al samples is normalized to the soil weight, the largest deviation from the average is 59%; when tritium activity is normalized to the weight of the water in the sample, the largest deviation from the average is 20%. This is shown in the top left plot. The percentage of water by weight in the four samples varied from 8.2% to 18.8%. The top right plot shows the ratio of activity normalized to grams of water and activity normalized to grams of soil. The bottom left plot shows the activity levels normalized to the weight of the water found in the soil samples; the bottom right plot shows the activity levels normalized to the weight of the soil samples. The error bars shown on the bottom two plots are simply $\pm$ 0.01 on the left and $\pm$ 0.001 on the right, and represent the level of the least significant digit in the data values.

3.1 Projection for NuMI Dolomite

To use the number of 1.4×10^{-1} $\frac{pCi}{g}$ for NuMI Dolomite calculations (in particular for the decay tunnel), we make the observation[8] that it is the average of $\frac{\sum_i n_i \sigma_{i3}}{(.037)}$. Equation 16 in Appendix B illustrates how we might convert the value of $\sum_i n_i \sigma_{i3}$ from one mix of elements (soil) to another (Dolomite). For

$$\frac{\left[\sum_i n_i \left(\frac{A_i}{16}\right)^{\alpha}\right]_{Dolomite}}{\left[\sum_i n_i \left(\frac{A_i}{16}\right)^{\alpha}\right]_{soil}}$$

we calculate 1.00011 for $\alpha = 2/3$ and 1.00307 for $\alpha = 0.8$. Thus, the mix of elements hardly matters, when considering soil or Dolomite.

In Appendix A.1 we discuss calculating the flux of hadrons for NuMI from star density values. Since star densities are obtained using a threshold kinetic energy[9] of 48 MeV it is necessary to increase the value for hadron flux to account for the lower threshold of 30 MeV that is included in the Borak et. al. flux normalization. This topic is discussed in Appendix A.1.

Utilizing equations 7 & 9 in Appendix A.1 we then have

$$\phi_{30} \;=\; (1.456)(4223) \;=\; 6.148 \times 10^3 \quad \frac{particles}{cm^2\ sec} \tag{1}$$

Peak saturated tritium activity using the NuMI assumptions (from TM-2009, discussed in Appendix A.1) is then[10]

$$(.14) \times (6.148 \times 10^3) \;=\; 861\ \frac{pCi}{g} \tag{2}$$

This corresponds to the activity in the groundwater at the edge of the tunnel (where the Dolomite begins), under static conditions.

[8] See the discussion in Appendix B.

[9] CASIM ([7]) has a momentum cutoff of 300 MeV/c built in; MARS ([8]) has an option to choose to run with this same momentum threshold. For neutrons, a momentum threshold of 300 MeV/c corresponds to a kinetic energy of 48 MeV.

[10] In equation 2 the number 6.148×10^3 is unit-less, because it is being used as a ratio of the flux in equation 1 and the unit flux that is the case for Table 4 of Borak et. al. The units in equation 2 could have been written pCi/ml, since we are calculating activity in the water in the Dolomite.

3.2 Comparison with TM-2009

TM-2009 [2] calculated a ^{3}H activity[11] of 10 $\frac{pCi}{ml}$. This number is gotten by averaging star density in the rock between the tunnel wall and 1.5 meters further out (where the star density has decreased to 1% of its peak value). The average star density was calculated to be 0.19 times the peak star density. TM-2009 used a "90 %" leaching factor ω_i (equal to 0.27) to arrive at the tritium concentration of 10 $\frac{pCi}{ml}$. TM2009 [2] calculates a decay factor

$$(1 - e^{(-3.15\times10^8 * 1.79\times10^{-9})}) \; = \; .431$$

which represents the difference between ten years of running and saturation.

The use of a "90%" leaching value has been questioned by the various committees that the Fermilab Director appointed to review past targeting vulnerabilities. In response, an ad hoc groundwater working committee appointed by D. Cossairt is considering using a leaching factor ω_i that would correspond to the amount of water present in the medium. In the case of NuMI Dolomite, one such measure is the rock porosity, which has been measured as 0.19 (see Reference [9]). If one assumes this porosity is filled with water, then one calculates ω_i as

$$w_i \; = \; \frac{.19}{2.67} \; = \; 0.071 \tag{3}$$

The value of rock density being used here (2.67) is that from TM-2009. The ratio of these differing values of ω_i is 3.8.

In calculating a tritium activity of 10 pCi/ml, TM-2009 used a K value[12] of 0.03 (taken from TM-1851 [10]). The value of K = 0.03 was determined from preliminary results from the study described in Reference [6]. Based upon the considerations in this paper, we would use the value of 0.035—given in equation 17 in Appendix B.

Collecting together the factors, we scale the TM-2009 activity value of 10 pCi/ml in order to compare to the saturated peak value of activity at the

[11] This activity corresponds to "90% leaching". The TM-2009 number for "99% leaching" is 5.4 $\frac{pCi}{ml}$.

[12] Probability of producing one radionuclide atom per inelastic collision (star). See Reference [11] for early use of this notation.

tunnel wall in equation 2; we then have

$$\left[\frac{0.27}{.071}\right]\left[\frac{1}{.431}\right]\left[\frac{1}{0.19}\right]\left[\frac{0.035}{0.03}\right] \times 10 \;=\; 542 \text{ pCi/ml} \qquad (4)$$

If we wish to compare this to the 861 pCi/ml given in equation 2 (using the ^{3}H activity from Borak et. al.), one further factor is necessary, since the star density used to get the value in equation 4 had an energy threshold of 48 MeV. To make the comparison we need to use a star density value that has a threshold[13] of 30 MeV. The extra factor needed is given in equation 9 (Appendix A.1). Applying this factor we have

$$(542)\,(1.456) = 789 \text{ pCi/ml}$$

We note that the measured K values for tritium vary, both by material type, and in a manner correlated with the amount of water that existed in the material sample at the time of irradiation (see Tables 6 & 7 in Appendix B). Such a dependence on the amount of water[14] contained in the sample could explain the large variation in the K values seen in Reference [6]. Utilizing the ^{3}H activity measured by Borak et. al. [3] has the advantage (compared to using a K factor) that it starts out with a number that is already ^{3}H activity expressed per gram of water in a medium. As has been noted (see Section 3.1), to use the Reference [3] ^{3}H activity we had to obtain a number for the flux of hadrons with a 30 MeV threshold, and we had to extend it to a different medium.

3.3 Discussion of Uncertainties

As indicated in Figure 2, we have uncertainty in the value of 0.14 (used in equation 2) of the order of 20%, just due to the variation in the value of

[13]Regarding the 48 MeV threshold, Reference [12] says ". . . this threshold is introduced mainly for convenience of computation. At low energies the Hagedorn-Ranft production model is not valid. Furthermore the cross sections vary rapidly with energy in this region." However, Reference [13] assumes the non-elastic cross sections to be energy independent from about 30 MeV up to the highest energy considered in that reference. Examination of Reference [14] convinces us that the tritium cross sections of interest are sufficiently stable between 30 MeV and 48 MeV that we don't need to consider them as varying.

[14]Unfortunately References [6], [15], and [16] don't quote the fraction of water contained in the samples measured.

activity normalized to water for the four samples in Table 4 of Reference [3]. In extending the hadron energy spectrum from 48 MeV to 30 MeV we have relative uncertainties in the flux value[15] of order 10%, as discussed in Appendix A.2. The astute reader could make a list of other uncertainties (which are not considered in detail here).

4 Comparisons to Other Calculations

In Section 3.2 we have already compared two methods of calculating tritium concentration in groundwater. The first method utilized the measured activity (from Reference [3]) per unit flux (30 MeV threshold) per gram of water—adjusted to NuMI conditions as given in Reference [2]. The second method of calculation was that given in reference [2], which used the K value[16] of 0.03 ^{3}H atoms per star from Reference [10]. We have seen in Section 3 that the two methods of calculation can be made to agree to a reasonable level if the amount of "leach" water in Reference [2] is made equivalent to the amount of water that would fill the Dolomite porosity, rather than the amount of water[17] that is based upon a "90% leaching". In the second method we also made a small adjustment to the K factor.

We'd like to continue making comparisons and this time refer to CERN calculations for their proposed long baseline neutrino beam, as well as the NuMI Technical Design Report [1].

4.1 CERN Papers

One CERN paper of interest[18] is entitled "Initial Estimates of Radiological Parameters of Environmental Interest for the CERN/INFN Gran-Sasso Neutrino Project"[17]. On page 9 it discusses radionuclides produced in the rock ("Molasse") surrounding their underground tunnels. For ^{3}H it gives a value

[15]The flux was calculated in equation 1.

[16]See Table 6 for a compendium of K values.

[17]In Reference [10] these amounts were determined by percolation measurements made by Sam Baker on a column of Neutrino Area sand and gravel. These measurements were described in Reference [15].

[18]Reference [17] was kindly provided to us by Alex Elwyn. He also provided us with a subsidiary reference ([18]).

9×10^{-10} Bq/cm^3 in water[19], for a star density in the rock with value 1 cm^{-3}. It says that to this must be added the direct production of ^{3}H by spallation reactions in the oxygen of the water, which results in an additional 1×10^{-10} Bq/cm^3. For their conditions they quote 45 stars in rock per proton. In their Table 3 they list K = 0.05 ^{3}H atoms per star in their rock[20].

A second CERN paper [18] gives further information regarding the numbers found on page 9 of Reference [17]. It says that the proportion by volume of water in the Molasse is estimated to be 10%. It gives the number of nuclei of ^{3}H produced per star in water as 0.113 & further says that the interaction mean free path in water is approximately twice that in their rock.

With this information at hand we can verify their numbers as follows:

Spallation in Rock

$$\begin{aligned} .05 &= {}^3\text{H atoms per star in rock} \\ \text{star density} &= 1 \text{ cm}^{-3} \text{ (in rock)} \\ \left[\frac{.05}{(17.75)\,(3.15 \times 10^7)}\right] &= 8.9 \times 10^{-11} \text{ Bq/cm}^3 \text{ (normalized to 1 cm}^3 \text{ of rock)} \\ &= 8.9 \times 10^{-10} \text{ Bq/cm}^3 \text{ (normalized to 10\% water in rock)} \end{aligned}$$

where $\lambda = (17.75)^{-1}$ years[21].

Spallation from ^{16}O in water

$$\begin{aligned} 0.113 &= {}^3\text{H atoms per star in water} \\ 0.5 &= \text{star density per cm}^3 \text{ of water} \\ \left[\frac{(.113)(0.5)}{(17.75)\,(3.15 \times 10^7)}\right] &= 1 \times 10^{-10} \text{ Bq/cm}^3 \end{aligned}$$

[19] 1 Bq equals 1 disintegration s^{-1} (or $\frac{1}{3.7 \times 10^{10}}$ Ci.)

[20] From a private communication with one of the authors (G. R. Stevenson) J. D. Cossairt learned that this K value derives in some manner from Borak et. al. [3]. It is not based upon independent measurements. In equation 18 we compute a value of K = 0.048 for the Molasse, so we think we understand the published value of 0.05.

[21] $\lambda = \left(\frac{12.3 \text{ yr}}{ln\, 2}\right)^{-1}$.

The number most relevant to our calculations in Section 3 is the number K = 0.05 ^{3}H per star in their rock. It is to be noted that the CERN papers are using the volume of water present in the Molasse and that all of the tritium produced in the rock is assumed to be present in that water; there is no concept of a "90% leaching" volume of water being used.

4.2 NuMI TDR

The radiation safety chapter (Chapter 4) of the NuMI TDR [1] makes reference to TM-2009 [2] and a paper by Cossairt and Cupps [19]. The result of the considerations in Chapter 4 is a K value of 0.0076 ^{3}H atoms per star in rock. It is to be noted that this value is a factor of 10 below the equivalent number for Fermilab soil (Reference [10]) and a factor of 6.6 below the equivalent value in the CERN papers (References [17] & [18]). TDR Chapter 4 continues the assumption[22] that the tritium must be leached out of the rock. It places the tritium produced in a "90% volume" of water; this is a volume based upon a combination of leaching measurements found in References [15] and [6]. Table 4-6 in the TDR gives the weight factor ω_3 as 0.325. TDR Chapter 4 boosts the "leached" tritium numbers by a factor of 1.3, in order to account for direct production of tritium in groundwater.

Starting from equation 2, we now calculate the peak saturated tritium concentration that would correspond to the configuration in the NuMI TDR [1]. Reading from Figures 4-12 and 4-15 a working limit for star density at

[22]This assumption is consistent with current Fermilab methodology, which is given in Reference [20]. This methodology is also incorporated into the Fermilab ES&H Manual [21]. This methodology was developed for the case where irradiation takes place in unconsolidated media (e.g. soil) above an underlying aquifer, and the water carrying radionuclides must migrate to the underlying aquifer to be subject to regulatory consideration. We question the validity of this methodology for the very different NuMI environment. Also, we believe that it is not fully consistent with the data of Reference [3].

the tunnel wall is the value[23] of 6×10^{-11} stars cm^{-3} p^{-1}. We then have

$$(861)\left[\frac{6 \times 10^{-11}}{1.3 \times 10^{-11}}\right] = 3,974\frac{pCi}{g} \tag{5}$$

Proceeding with a comparison based upon utilizing a K value, we make the argument that the tritium concentration numbers in Chapter 4 of the NuMI TDR are low—first, by having K low, and second, by diluting the tritium in too much water. These two factors are[24]

$$\frac{.035}{.0076} = 4.6 \quad \text{(ratio, K factor, atoms of } ^3\text{H per star)}$$

$$\frac{.325}{.071} = 4.6 \quad \text{(ratio, volume of water used for } ^3\text{H concentration)}$$

In the second of these equations 0.325 comes from Table 4-6 of the NuMI TDR & 0.071 was first seen in equation 3 of this paper. The product of these two factors is 21. This number should be divided by 1.3 because of the contribution that was added for direct production of tritium in water; the resultant factor[25] is 16.

[23]TM-2009 [2] used 1.3×10^{-11} stars cm^{-3} p^{-1} for the star density at the decay tunnel wall. Appendix A.1 used this same number for star density, to calculate the flux of hadrons to use in equation 1. TDR Figures 4-12 and 4-15 indicate a band of limiting star density, ranging from 6×10^{-11} stars cm^{-3} p^{-1} to 1.2×10^{-11}. Figure 4-12 indicates that this band corresponds to a range of G_{decay} from 0.10 to 0.19 (where G_{decay} is the factor relating average star density to peak star density). The star densities were calculated in the NuMI TDR using MARS [8]. Those calculated for TM-2009 [2] were calculated with CASIM [7]. MARS and CASIM are believed to agree to within a factor of two, when calculating star densities.

[24]For the first of these factors we are using the K value of .035 from equation 17.

[25]A general concern for any calculation employing a K factor based upon the Borak et. al. results is that the value for $\sum_i n_i \sigma_{ij}$ that goes into the numerator for K has a flux threshold of 30 MeV in its determination, yet the prescription for obtaining the number of radionuclides is to multiply K by a star density from CASIM or MARS which has a threshold of 48 MeV in its determination. As discussed in Section 3.2 the effect of lowering the star density energy threshold can be calculated to be a factor of 1.46. Application of this factor would boost the number 16 to 23.

5 Impact for Shield Design

It is important to note that ground water in the Dolomite will not be static. Water motion will reduce radionuclide concentrations—compared to the static values presented in the previous section[26]. Such motion includes regional flow and inflow into unlined NuMI tunnels[27].

The predominant water flow through the Dolomitic rock is expected to be through interlocking joints (fractures) in the rock structure. These fractures are largely vertical in orientation, with significant open structure in the upper Dolomite formations (Joliet, Kankakee and Elwood). The joints become less frequently spaced, and also largely closed, in progression through the underlying Maquoketa formations—where significant Dolomitic shale resides. The result is a vertically confined aquifer system, where the predominant regional water flow is horizontal. For saturated regions there is also water seepage through the rock matrix—with much lower hydraulic conductivity than found through the fractures. Reference [23] has calculated an average horizontal flow velocity of 5.9 m/yr (regional) in a southeasterly direction, with significant large localized variations through regions of different hydraulic conductivity. A general gradient in hydraulic conductivity ranges also exists, as a function of depth, with value decreasing for increasing depth below the top of the rock interface (with glacial till).

The presence of the unlined NuMI tunnel results in water inflow to this tunnel. A realistic calculation, including inflow effects together with the effects of a nearby large well (FNAL Well-1), would be complex. To be credible such a calculation should be done utilizing appropriate professional rock hydrogeology modeling resources[28]. This paper is meant to provide information on which to base a discussion of the need for either more comprehensive modeling of groundwater flow around the NuMI tunnels and caverns, or a re-evaluaton of the NuMI shielding design.

The Close Out Report ([24]) of the Lehman panel recommended (under their comments on Facility Construction, WBS 1.2) that: "Groundwater systems in rock need further study including transport mechanisms. Existing

[26]The reduction mechanisms are decay and limited exposure time.

[27]Reference [22] discusses quantitatively the effects of uniform inflow.

[28]In the time interval since 1/15/99 EarthTech has been retained as a consultant to do such modeling for the NuMI tunnels. Their first report should be ready by the end of May, 1999.

groundwater data should be compiled and analyzed to establish existing conditions for groundwater elevation and gradients. This data should be input into computer models to estimate groundwater flows into and around the tunnels and caverns to assist in predicting transport of radionuclides." A study like that recommended should help to quantify the benefit of water inflow into the tunnels (for the reduction of tritium concentration in the groundwater resource outside the tunnels).

6 Summary and Recommendations

We have addressed with a new model expectations for tritium production in the Dolomitic rock adjacent to the NuMI beam tunnels.

As part of this process, we have revisited calculations for expected tritium activity under static groundwater conditions. Utilizing directly the results of Borak et. al. [3] for the irradiation of soil samples near proton synchrotrons, we have provided an interpretation which is consistent with the full body of this data, and which enables simple extrapolation to expectations for groundwater activation in rock, instead of soil.

We find that the Borak et. al. data shows that a subsequent leaching process after the beam irradiation period does not remove a substantial amount of tritium from soil (or from rock). Water which becomes tritiated has to be present during the irradiation. In Appendix C we discuss other sources of data that can be interpreted in a similar manner.

We also find expected tritium activation levels adjacent to the NuMI beam tunnels under static conditions which are a significant factor higher than those projected in the NuMI TDR [1] , and illustrate the reasons for this discrepancy.

We believe that water inflow into the NuMI tunnels provides significant reduction of groundwater activation, but have not been able to quantify this in a manner that accounts for local variations in rock jointing. In Reference [22] J. D. Cossairt has shown a dramatic reduction, in calculations averaging effects over the tunnel length[29]. Considerable motivation should exist for

[29]In fact, the velocity values in Reference [22] neglect to include the effect of the formation porosity, and would therefore actually be higher–by a factor of about five. This would have the effect of enhancing the effect of inflow above what is shown in that paper.

more comprehensive modeling of groundwater flow around the NuMI tunnels and caverns utilizing professional hydrogeology modeling resources[30].

We recommend that beam activation testing of Dolomitic rock samples be carried out during upcoming accelerator running, with care taken to preserve water in the samples during irradiation; such measurements should also take care to understand the effect of varying sample water percentage.

Finally, the understanding that water presence during the irradiation is essential for significant tritium activation allows possible consideration of more favorable shielding requirements for a large downstream portion of the NuMI beam tunnel in the Maquoketa formations, where saturated conditions may not be prevalent. It also enables in more straight-forward manner the utilization of solutions based on groundwater inflow into the NuMI tunnels, as transferrable tritium does not build up in the Dolomitic rock. Under these conditions, however, the consideration of ^{22}Na production and residual activation in the tunnel may determine shield requirements.

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A separate paper [25] considers the effects of our new view of tritium production on the sump water inflow calculation that was presented in TM-2009 [2]. This paper has some considerations of the effect of fractures as well.

[30]In the time interval since 1/15/99 EarthTech has been retained as a consultant to do such modeling for the NuMI tunnels. Their first report should be ready by the end of May, 1999.

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Appendix A Hadron Flux and Threshold Energy Dependence

A.1 Flux Calculation

We will attempt to calculate the flux of hadrons for NuMI, using the relationship between star density and hadron flux that is given by the expression

$$\phi = \lambda \times S \tag{6}$$

where ϕ is hadron flux, λ is the interaction length, and S is star density. Star density S just outside the decay pipe tunnel wall is taken as[31]

$$S = (0.7) \times (1.86 \times 10^{-11}) \times (9.4 \times 10^{12}) = 122.4 \quad \frac{stars}{cm^3\ sec}.$$

The interaction lengths for both nucleons and pions are given in the output of CASIM [7] runs that have Dolomite in the 9 available materials in the input deck. The values for Dolomite are given in the third column of Table 1. For flux, equation (6) gives

$$\phi = 34.5 \times 122.4 = 4223 \quad \frac{particles}{cm^2\ sec}. \tag{7}$$

Since CASIM uses a momentum threshold of 300 MeV/c, this flux is for hadrons with the same momentum threshold. One can easily calculate that a 300 MeV/c momentum threshold corresponds to a 48 MeV kinetic energy threshold for neutrons (and protons).

To use the activities in Table 4 of Borak et. al. we need fluxes with a 30 MeV threshold. Fig. 1 and Table 2 of Borak et. al. indicate that it is reasonable to assume[32] that the neutron energy spectrum falls like $E^{-1.8}$. Using this we can estimate how the flux with a threshold of 48 MeV relates to a flux with a threshold of 30 MeV.

[31] Each of these numbers is taken from TM2009 [2].

[32] Reference [26] uses the same energy dependence for the neutron spectrum in soil. It makes a similar adjustment, to translate from a threshold of 20 MeV for ^{11}C production to a threshold of 30 MeV for ^{22}Na production. Section A.2 of this appendix discusses the consequence of variation in the energy dependence of this spectrum.

We have that

$$\phi_{48} \;=\; \int_{48}^{\infty} \frac{dN}{dE}\, dE \quad \text{equals} \quad \lambda \times S$$

$$\int_{30}^{48} \frac{dN}{dE}\, dE \quad \text{is what is missing}$$

$$\text{Let } \frac{dN}{dE} \;=\; E^{-1.8}\, \xi$$

$$\text{Then} \quad \phi_{48} \;=\; \int_{48}^{\infty} E^{-1.8}\, \xi\, dE \;=\; \frac{E^{-0.8}}{-0.8}\bigg|_{48}^{\infty} \xi \;=\; \frac{(48)^{-.8}}{.8}\, \xi$$

$$\int_{30}^{48} \frac{dN}{dE}\, dE \;=\; \left[\frac{(30)^{-.8}}{.8} - \frac{(48)^{-.8}}{.8}\right] \xi$$

Add together $\int_{30}^{48} \frac{dN}{dE}\, dE$ and $\int_{48}^{\infty} \frac{dN}{dE}\, dE$ and get

$$\phi_{30} \;=\; \int_{30}^{\infty} \frac{dN}{dE}\, dE \;=\; \frac{(30)^{-.8}}{.8}\, \xi$$

Noting that ξ can be expressed as

$$\xi \;=\; \left[\frac{.8}{(48)^{-.8}}\right] \phi_{48}$$

the hadron flux (ϕ_{30}) with a threshold of 30 MeV is

$$\phi_{30} \;=\; \left[\frac{(30)^{-.8}}{.8}\right] \left[\frac{.8}{(48)^{-.8}}\right]\, \phi_{48} \tag{8}$$

$$\;=\; 1.456 \tag{9}$$

The flux adjustment factor calculated[33] is 1.456.

[33] Alex Elwyn kindly made available to us References [28] and [29], which contain graphs of neutron spectra. Using a couple of straight lines fit by eye on the spectrum of Reference [29] we did a similar calculation and obtained a flux correction factor of 1.17. Cat James has used the MARS [8] program to obtain this flux correction factor, and she determined its value to be 1.3.

Input	Units	Dolomite	Moist Soil I	Moist Soil II
Atomic Number		10.9	10.7	10.7
Atomic Weight	amu	21.8	21.6	21.6
Density	g/cm^3	2.8	2.25	2.15
Ionization Potential	eV	137.2	135.0	135.0
Radiation Length	cm	9.68	12.3	12.8
Nuclear Radius	Fermi	3.63	3.62	3.62
Elastic Cross Section	barn	0.164	0.162	0.162
Output				
interaction length for nucleons	cm	34.5	42.9	44.9
interaction length for pions	cm	42.7	53.0	55.5

Table 1: This table shows interaction lengths for nucleons and pions calculated by CASIM for three sets of input parameters—for Dolomite and two versions of "moist soil". Moist soil I is defined in TM-1898 [27] and has a density of 2.25 g/cm^3. Moist soil II has a density of 2.15 g/cm^3–meant to agree with the average soil density of the four samples in Borak et. al. [3]

A.2 Study of Sensitivity to Spectrum Shape

We can next explore some other choices for the energy dependence of the neutron spectrum, and we can explore the effect of introducing a physical cutoff to the spectrum at an energy E_{max}. We can write

$$\frac{dN}{dE} = E^{-1.8+\lambda} \tag{10}$$

$$\phi_\alpha = \int_\alpha^{E_{max}} \xi\, E^{-1.8+\lambda}\, dE \tag{11}$$

$$\tag{12}$$

where α is either 30 or 48 MeV. We can continue as follows

$$\phi_\alpha = \frac{\xi}{-.8+\lambda}\left[E_{max}^{-.8+\lambda} - \alpha^{-.8+\lambda}\right] \tag{13}$$

$$= \xi\,\frac{\alpha^{-.8+\lambda}}{-\lambda+.8}\left[1-\left(\frac{E_{max}}{\alpha}\right)^{.8-\lambda}\right] \tag{14}$$

This last expression diverges for $(-\lambda+.8) \Longrightarrow 0$. We will restrict ourselves to asking the effect of letting $\lambda = \pm.2$. From the expressions above we can determine

$$\phi_{30} = \phi_{48}\left(\frac{48}{30}\right)^{.8-\lambda}\frac{\left[1-\left(\frac{48}{E_{max}}\right)^{.8-\lambda}\right]}{\left[1-\left(\frac{30}{E_{max}}\right)^{.8-\lambda}\right]} \tag{15}$$

For the choices E_{max} = 20,000 MeV and $\lambda = \pm.2$ we can make a small table

E_{max}	λ	term 1	term 2
20,000	0	1.456	.99748
	-.2	1.6	.99910
	.2	1.32	.99328

In the table "term 1" is the term $(\frac{48}{30})^{.8-\lambda}$ and "term 2" is the term that follows it in equation 15 (which includes the effect of E_{max}).

Appendix B Tritium cross section from Borak et. al.

In their Table 4 Borak et. al. give a value of $\sum n_i \sigma_{ij}$ for tritium equal to $5.9 \times 10^{-3}(\frac{cm^2}{g})$ for the A-1 sample, in the last line of the table—where activity and cross sections[34] are per gram of water in the soil sample. In the expression $\sum n_i \sigma_{ij}$, n_i is the number of the i th type of target nuclei per gram and σ_{ij} is the excitation function for element i and production of radioisotope j (j=3 for much of this paper). The number 5.9×10^{-3} $(\frac{cm^2}{g})$ in the last line of Table 4 in Borak et. al is normalized to the amount of water (14% by weight) in the A-1 sample. The number just above it in the Table 4 is $8.2 \times 10^{-4}(\frac{cm^2}{g})$, and is normalized to one gram of soil[35].

The situation is complex, since tritium is being produced from molecules in the soil and from the oxygen atom in the water molecules. The ionized tritium that is produced has a range that typically is a fraction of a centimeter. If the ionized tritium is produced from soil molecules near enough to water molecules, it can displace a hydrogen atom in a water molecule. Regarding the production of tritium as separate from the process whereby it becomes incorporated into a water molecule is a way of understanding the last two lines of Table 4 of Borak et. al.

Table 1 of Borak et. al. gives the elemental composition of soil[36]. This information is reproduced here in Table 2. Looking at Figure 1 of Reference [14] leads us to believe that it is reasonable to assume[37] $\frac{\sigma_{i3}}{\sigma_{16,3}} = \left(\frac{A}{16}\right)^{\frac{2}{3}}$—for the purpose of unfolding a value for $\sigma_{16,3}$ from the data in Tables 1 and 4 of Borak et. al. The factor $(\mathrm{A}_i/16)^{\frac{2}{3}}$ ranges from 0.83 for Carbon to 2.31 for Iron[38]. Doing this allows us to extract the value of 23.2 millibarns for the

[34]Note that the activity (1.6×10^{-1}) and number of radionuclide (5.9×10^{-3}) values are related by the factor $\frac{1}{.037}$; this is the conversion factor between disintegrations per second and picoCuries.

[35]The ratio $\frac{8.2 \times 10^{-4}}{5.9 \times 10^{-3}}$ is 0.14

[36]It doesn't indicate if this is for one particular sample or is an average over the four samples.

[37]In Table 8 other values for this exponent are also used; the effect of changing the exponent is not strong.

[38]Assuming $(A/16)^{\frac{2}{3}}$ scaling of cross section is equivalent to assuming that the spallation is occurring off of the nucleons at the surface of the atom

value of $\sigma_{16,3}$ for sample A-1, using the equation

$$\sum_i n_i \sigma_{ij} \;=\; \left[\sum_i n_i \left(\frac{A_i}{16} \right)^{\frac{2}{3}} \right] \sigma_{16,3} \tag{16}$$

When we do this calculation for all four samples, we arrive at the values for

Element	A	Weight Fraction
Silicon	28	0.1447
Aluminum	27	0.0244
Iron	56	0.011
Calcium	40	0.07
Magnesium	25	0.0379
Carbon	12	0.0512
Sodium	23	0.0034
Potassium	39	0.00814
Oxygen	16	0.64

Table 2: Elemental Composition of Soil, taken from Reference [3].

cross section ($\sigma_{16,3}$) shown in Table 3. The variation in the values for $\sigma_{16,3}$ in Table 3 shows the effect of the varying amounts of water in each sample.

Sample	% H_2O (by weight)	$\sum n_i \sigma_{i,3}$ (cm^2/g)	Extracted $\sigma_{16,3}$
A-1	14	8.2×10^{-4}	23.2 mb
B-1	18.8	1.1×10^{-3}	31.2 mb
B-2	8.2	3.3×10^{-4}	9.3 mb
B-3	11.6	5.2×10^{-4}	14.7 mb
Average	13.2	6.9×10^{-4}	19.6 mb

Table 3: Values for $\sigma_{16,3}$ obtained from a calculation like that—for all four samples in Borak et. al. Also included are the water % by weight values from Table 1 of Borak et. al. and the values for $\sum n_i \sigma_{i,3}$ from their Table 4.

The average value of 19.6 mb is not an unreasonable value for $\sigma_{16,3}$, when compared with the value mentioned in the paper by D. Cossairt and V. Cupps

[19]. They give there a value of σ_3 =33 mb for the production cross section for producing ^{3}H by means of the ^{16}O(p,^{3}H)X spallation reaction. When the value 19.6 mb is compared with the values in Figures 1-3, 8, & 9 in Reference [14] it could be said that it is perhaps somewhat high.

Tables 4 & 5 contain information similar to that in table 2, with the material being Dolomite and CERN Molasse (see Reference [17]). The information they contain can be used to recalculate K_3 (the probability that a ^{3}H nucleus will be produced at each "star"), using the formula[39]

$$K_3 \;=\; \left[\frac{\sum_i n_i\sigma_{ij}}{\sum_i n_i\sigma_{inel}}\right]\left[\frac{f_{new\_material}}{f_{soil\_average}}\right]$$

where the numerator in the first factor can be re-expressed using equation 16 and we use the average value for $\sigma_{3,16} = 19.6$ from Table 3. The second factor makes an adjustment for the percentage by weight of water in the material. From Table 6 we have that $f_{soil\_average} = 0.1315$. For Dolomite $f_{Dolomite} = 0.071$, from equation 3. For Molasse $f_{Molasse} = 0.10$, from Reference [18]. Using this information we arrive at the values[40]

$$K_3, Dolomite \;=\; .035 \tag{17}$$

$$K_3, Molasse \;=\; .048 \tag{18}$$

[39]See Reference [11] for a similar use of this formula.

[40]The value for $\sum_i n_i\sigma_{inel}$ computed from Table 4 was 1.07×10^{-2}; for Molasse Table 5 was used and the value computed for $\sum_i n_i\sigma_{inel}$ was 1.09×10^{-2}.

Element	A	Weight Fraction	σ_{inel} (barns)
Calcium	40	0.216	0.62
Carbon	12	0.130	0.195
Oxygen	16	0.519	0.31
Magnesium	25	0.135	0.43

Table 4: Elemental Composition of Dolomite. This uses for Dolomite the molecular formula $(CaCO_3)(MgCO_3$, where one of the Calcium atoms in Calcium Carbonate is replaced by a magnesium atom. Also included are the inelastic cross sections from Table I of Reference [13].

Element	A	Weight Fraction	σ_{inel} (barns)
Oxygen	16	0.54	0.31
Silicon	28	0.294	0.47
Calcium	40	0.123	0.62
Carbon	12	0.037	0.195
Hydrogen	1	0.0073	0.025

Table 5: Elemental Composition of CERN Molasse. Also included are the inelastic cross sections from Table I of Reference [13].

Sample	Description	% H_2O (by weight)	depth (feet)	$\sum_i n_i \sigma_{i3}$ cm^2/g	K_3 (atoms/star)
A-1	Glacial till	14	≈ 20	8.2×10^{-04}	0.075
B-1	Gray sandy clay	18.8	3-6	1.1×10^{-03}	0.100
B-2	Red sandy clay	8.2	6-12	3.3×10^{-04}	0.030
B-3	Gray clay	11.6	15-22	5.2×10^{-04}	0.047
	Average (soil)	13.2		6.9×10^{-04}	0.063
	Dolomite (eq. 17)	7.1			0.035
	Molasse (eq. 18)	10			0.048

Table 6: Compendium of K values for Tritium (in water) used in this paper. The first four are a result of computing K values for each sample from Borak et. al. [3]. The next is an average of those four values. Below that we have included the values from equations 17, 18, for comparison. For the first four rows we arrive at the value for K by dividing column 5 by 1.1×10^{-2}, which we take from Reference [11]. As noted earlier the K values are quite dependent on the amount of water contained in the samples used.

Description	K_3 (atoms/star)
Dolomite [2]	0.03
† Dolomite [1], [19]	0.0076
Molasse [17]	0.05
Limestone [6]	0.0023
Austin Chalk [6]	0.012
Taylor Marl [6]	0.027
Eagle Ford Shale [6]	0.033
Ellis County soil [6]	0.076
Fermilab soil [6]	0.044
Bentonite [6]	0.18

Table 7: Compendium of additional K values for Tritium (in water). The source of the K_3 values is the reference indicated next to the description. As noted earlier the K values are quite dependent on the amount of water contained in the samples used. The % water in the samples was not given in Reference [6] (as it was in reference [3]). [† The value of K in the second row is the value used in the NuMI TDR [1]. This value was calculated in reference [19], based upon a cross section for direct production of tritium in water & using rock porosity to adjust to a volume of rock.]

α in A^α	$K_{3,Dolomite}$	$\sigma_{3,16}$ (barns)
$\frac{2}{3}$	0.035	1.963×10^{-26}
0.8	0.034	1.922×10^{-26}
1.0	0.033	1.857×10^{-26}

Table 8: Table showing the effect on $K_{3,Dolomite}$ and $\sigma_{3,16}$ of varying the exponent α in $\left[\frac{A_i}{16}\right]^\alpha$ in equation 16.

Appendix C Independent Validation of Results

Even although the Borak et. al. [3] data was accumulated in a careful set of measurements, and the transition from data using soil to expectations for Dolomite appears not to have large uncertainties, it remains very important to obtain independent validation of the projections presented here.

One measurement parameter which may provide additional validation of the Borak et. al. results is that of the measured ratio of tritium to ^{22}Na activation in different samples. The expectation is that for a given target material ^{22}Na activation is proportional to particle flux or star density. Besides this dependence, tritium activation is also dependent on the amount of water present in the sample during beam exposure. Indicative of this is, for example, data reported by Baker [15] for soil activation measurements near the Main Ring Abort.

In soil boring measurements outside the tunnel wall adjacent to the Abort (Fig. 2 & 3 of reference [15]), the Baker data shows ^{22}Na activation which falls off in the exponential manner expected with increasing distance from the abort target source. The observed tritium distribution is very different, being relatively much reduced, compared to the ^{22}Na, in the elevation region along the tunnel, where the presence of tunnel under-drains implies greater removal of soil moisture. The ratio of activity of tritium and ^{22}Na changes by about a factor of eight, when comparing levels away from the tunnel wall and those in proximity to the tunnel wall.

Additional data plots which are also suggestive of greatly reduced tritium levels when less water is present during beam activation are seen in Fig. 3 and 4 of reference [16]. Figure 3 from reference [16] shows relative tritium and ^{22}Na activation concentrations obtained by a boring through the sand and gravel surrounding the Neutrino Area target tube. This porous material is capped by a thick clay cover. Underneath it is an impervious membrane. Just above the impervious membrane there is a drain line to remove water that collects there. Reference [16] reports that prior to making the boring into this region, water was not collected from the drain above the membrane. Hence, this sand & gravel region surrounding the target tube was relatively dry. It can be seen in Figure 3 of reference [16] that ^{22}Na levels are typically a factor of several greater than those observed for tritium.

Borings were also made near the downstream wall of a secondary target in the Neutrino Area. Results from these are plotted in Figure 4 of reference [16]. Near to the wall, in a medium of sand and gravel, we again see elevated ^{22}Na levels (relative to tritium). A second boring, in clay two meters further from the wall, shows the opposite effect, with relatively much higher tritium levels.

The behavior of the ratio of activity of tritium and ^{22}Na in the data of references [16] and [15] corroborates the interpretation of the Borak et. al. results—i.e. that tritium activity is dependent on water being present in the medium during exposure. However, to demonstrate this conclusively and more quantitatively for NuMI it is important that new, careful accelerator exposures be done in the near future, measuring activations for Dolomite (and soil) samples containing different percentages of water.