

CERN 68-42
9 December 1968
Health Physics Group

ORGANISATION EUROPÉENNE POUR LA RECHERCHE NUCLÉAIRE
CERN EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

INDUCED RADIOACTIVITY IN THE EARTH SHIELDING ON TOP OF
HIGH-ENERGY PARTICLE ACCELERATORS

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G E N E V A
1968

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ABSTRACT

A study was made of the activity of radionuclides having half-lives longer than two days, that are produced in the earth shielding of high-energy particle accelerators; the CERN 28 GeV Proton Synchrotron was taken as a representative example. Measurements were restricted to ^{22}Na and ^{45}Ca for reasons of counting techniques, chemical treatment, and relative hazard. Up to 2 pCi/g ^{45}Ca and 6.9 pCi/g ^{22}Na were measured. The concentrations determined at different locations were compared with calculations using experimental neutron flux data, and a good agreement of both values was found.

It can be concluded from the measurements that no hazard exists as far as activity concentration of the effluent drainage water or manipulation of the earth from the shielding during shut-down periods is concerned, at least when working conditions such as those of the CERN Proton Synchrotron at present are maintained.

However, secondary particle fluxes increase considerably for improvements of proton intensity and target efficiency at 28 GeV/c proton energy or for a larger 300 GeV/c proton accelerator. For that case it can be shown from the data presented that a certain hazard due to ^{22}Na and ^{45}Ca concentration even for the effluent drainage water cannot be ruled out.

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1. INTRODUCTION

Hazards arising from the induced radioactivity in the vicinity of high-energy particle accelerators have often been discussed¹⁻³⁾ and estimations of the activity level have been made⁴⁻⁸⁾. In view of the increasing energies and intensities of future accelerators, and improved facilities at existing ones, it seemed worth while to compare the results of an elaborate determination of these induced activities with estimations presented up to now and with revised calculations.

An attempt was made, therefore, to evaluate the degree of activation of some elements in the earth shielding on top of the CERN Proton Synchrotron tunnel. The study was facilitated by the survey results of the CERN Health Physics Group on neutron fluxes near the 28 GeV Proton Synchrotron and by the many observations and figures collected during the CERN - Lawrence Radiation Laboratory - Rutherford Laboratory (CLR) shielding experiment in 1966⁹⁻¹¹⁾. Using these values and the chemical composition of soil as a basis, the activation near the target regions has been calculated for comparison with measurements of the activity of some of the long half-life isotopes formed.

It was assumed that only neutrons ranging from thermal energy to very high energy leak through the concrete shielding of the tunnel construction. A list was made of all radioactive isotopes generated by the nuclear reactions possible [(n, γ); (n,2n); (n, α); (n,p); (n,spall.)] between these neutrons and the elements present in the soil. This list was shortened by eliminating all isotopes with half-lives shorter than several days and radioisotopes produced at a very low rate by spallation. As regards the composition of the soil used for the CERN Proton Synchrotron shielding - which without much variation is similar for many accelerators - only four radionuclides seem to be produced in measurable quantities, namely ⁴⁵Ca, ²²Na, ⁷Be, and ⁵⁴Mn. Of these ⁴⁵Ca and ²²Na reach concentrations near the maximum permitted levels.

Therefore, a method was worked out to measure ⁴⁵Ca and ²²Na, simultaneously. Because of the low β -energy of ⁴⁵Ca ($E_{\max} = 0.25$ MeV), a liquid scintillation technique was applied to obtain appreciable counting efficiencies for the low count rates expected. This technique necessitated a chemical separation of alkaline earth and alkali elements from all other elements in the soil to avoid too large salt concentrations and their quenching effects, especially the nearly 100% colour quench due to Fe^{III} ions in solution. The separation processes were combined with a concentration process to get better count rates. The liquid counter results were controlled by measuring the γ -activity of the treated samples with a 3 \times 3 in. NaI crystal and comparison of the ²²Na concentrations found. The chemical treatment chosen had the inconvenience that ⁷Be and ⁵⁴Mn were at least partly lost during the process and, therefore, could not be measured for comparison with the calculation of their concentration. However, the good agreement of calculations and measurements done for ⁴⁵Ca and ²²Na implies that the real concentration of ⁷Be and ⁵⁴Mn, respectively, is of the order of magnitude calculated.

2. SAMPLE POSITIONS AND SAMPLING

The direction of emission of secondary particles and their approximate intensity distribution near target regions is known from measurements made by the Health Physics Group^{10,12}). The highest intensities will often be found downstream from the target at an angle of about 60° with the beam. To meet these conditions most samples were taken 3 m downstream from the end of the last upstream magnet above the axis of the tunnel and in some cases above the inner tunnel wall.

A hole was dug with a small excavator and the first soil sample was taken at 1 m depth from the top level of the shielding, the second one at 2 m depth. All samples had a weight of about 1 kg and were put in tightly closed plastic bottles to avoid water evaporation. The samples were collected on 25 March, 1968, two days after the shut-down of the Proton Synchrotron, after a long working period of little more than one year (maintenance shut-downs included). For comparison, samples at a "quiet section" (between magnets 32 and 33) of the accelerator were taken as well. Table 1 lists the samples and their respective position. The shielding thickness from beam level to the sample location is introduced into the sample symbol.

Table 1
Position of samples taken from the top of the
earth shielding (M for magnet)

Sample No. *)	Position
S(1;400;100)	3 m downstream target 1 (M100) above the axis; 1 m depth from top
S(1;200;100)	As above, but 2 m depth from top
S(1;400;98)	3 m downstream target 1 (M100) above inner diameter; 1 m depth from top
S(1;200;98)	As above, but 2 m depth from top
S(19;600;98)	At target 19 (M18), above inner diameter; 1 m depth from top
S(19;400;98)	As above, but 2 m depth from top
S(58;600;100)	3 m after SS58 (M57-M58) above the axis; 1 m depth from top
S(58;400;100)	As above, but 2 m depth from top
S(32;600;100)	Between M32 and M33, above the axis; 1 m depth from top
S(32;400;100)	As above, but 2 m depth from top

*) S(x;y;z): x : number of nearest target or magnet
y : shielding thickness from beam in g/cm²
z : distance from accelerator centre in m,
beam orbit at 100 m

3. CHEMICAL TREATMENT OF THE EARTH SAMPLES¹³⁻¹⁵⁾

For the counting technique chosen (liquid scintillation counting) samples must be in the form of a solution. To increase the reliability of the results this solution should only contain the radioelements sought.

The activity calculations (see Section 6, Table 4) have shown that ^{45}Ca , ^{22}Na , ^{54}Mn , and ^7Be could be built in measurable quantities. But only for ^{45}Ca and ^{22}Na the calculated concentrations are not too far from maximum permissible levels¹⁶⁾ and would increase considerably for larger secondary-particle fluxes. Therefore, these elements were chosen for determination and comparison with the calculated activities. To avoid contamination of the samples with other radioisotopes present in the soil a chemical treatment is necessary.

A separation of alkali and alkaline earth elements - except beryllium - from all other elements is relatively simple¹⁷⁾, if the loss of all radioelements - beryllium included - different from alkali and alkaline earth elements during the single steps of analysis is accepted. Nevertheless, it is a rather time consuming process to work with large quantities of material without incurring substantial losses of the elements to be separated.

As a first step the water content of the earth samples was analysed by drying a weighed quantity of approximately 50 g of each in an oven at 120°C up to constant weight. The water content was practically identical for all samples and amounted to about 11% of weight.

After preliminary tests on the chemical behaviour of the sort of soil examined the following method was chosen for the separation process, taking into account the soil composition^{9,18)} (in per cent of weight, corrected for water content: SiO_2 : 53.2%; Fe_2O_3 : 3.8%; Al_2O_3 : 9.65%; CaO : 9.35%; MgO : 2.5%; Na_2O : 0.64%; K_2O : 0.88%; besides: CO_2 , Mn_3O_4 , SO_3).

- a) 80 g of wet earth are weighed into an 800 ml beaker and slowly dissolved (CO_2 -development) in aqua regia (concentrated HCl + HNO_3 , 3 : 1).
- b) The mixture is stirred and heated until the non-silicate material is dissolved and then evaporated to dryness. After addition of concentrated HCl , this treatment has to be repeated several times till HNO_3 has been totally decomposed and evaporated.
- c) The dry substance is dissolved in dilute HCl , heated to boiling and filtered from the insoluble SiO_2 precipitate. The filter has to be washed with dilute HCl .
- d) The filtrate is evaporated to dryness, dissolved in very dilute HCl , and Fe^{III} and Al are precipitated as hydroxides by dropwise addition of NH_4OH to the hot solution.

To avoid co-precipitation of alkali and alkaline earth metals (adsorption on the surface of the gelic hydroxides), pH-conditions have to be very well maintained and controlled¹⁷⁾. The precipitate has to be washed well with a slightly ammoniakalic solution of 2% NH_4Cl in water in decanting the clear liquid off several times.

Note: Be, Mn, Sc, and phosphate are precipitated in this step as well!

- e) The hydroxide precipitate is filtered and well washed with the hot NH_4Cl solution mentioned above.

- f) The filtrate is heated and kept boiling after addition of concentrated HNO_3 to destroy the large quantities of ammonia and then slowly evaporated to dryness.
- g) The dry substance, containing only alkali and alkaline earth chlorides (except BeCl_2) is dissolved in 0.01 NHCl , the volume measured and an aliquot taken for counting.

The time needed for the whole process, treating three samples in parallel, amounts to 9-10 working days plus some nights for slow evaporation to dryness without being watched.

4. COUNTING PROCEDURE AND CALIBRATION

High counting efficiencies even for low energy β -emitters can only be obtained by liquid scintillation counting techniques¹⁹⁾. For aqueous samples, containing up to certain quantities of salts and acids, dioxane based liquid scintillators are very convenient.

For the present work, the Nuclear Enterprises' NE 240 Liquid Scintillator was chosen, which takes up to 20% of its volume of water and still shows relatively little quenching. Because of quenching effects, salt concentrations of samples should not vary too much, and the same volume of sample should always be taken for a given volume of scintillator. If the conditions are kept constant, all quenching effects cancel in respect to the calibration standard²⁰⁻²²⁾.

Counting was performed with a Nuclear Chicago Model 4534 Educational Liquid Scintillation System. This apparatus has three independent window amplifiers to count in three channels of chosen amplification and window width. Samples are changed by means of a manual device which is placed between two photomultipliers in coincidence.

For calibration the balance point amplification for a given radioactive isotope is determined at a chosen window width. The detecting efficiency is calculated from the count rate of a standard at this point and its disintegration rate.

The three independent channels have the advantage that doubly-labelled samples can be counted without changing amplification or window width of one channel at each measurement, but maintaining fixed positions at each channel. Doubly-labelled samples are samples in which two different β -emitting isotopes are present. In counting samples of this type, both isotopes are simultaneously counted in one period of time. It is not possible to choose two analyser channels such that only pulses from one isotope are counted in each channel, because the continuous β -spectra of any two isotopes will overlap to some extent. If the spectral overlapping is not too great - i.e., if a major fraction of the spectrum of the higher energy appears above the maximum energy of the lower energy isotope - it may be possible to choose parameters of amplification such that the counting efficiency of the lower energy isotope is practically zero in the channel for the higher energy.

If the energy separation between two isotopes is not great enough, both isotopes will be counted in both channels at a certain efficiency, i.e., four efficiencies have to be determined to calculate the activities of each isotope. In a given channel, the two efficiencies are not independent; if the channel is chosen to give a desired efficiency for one isotope, the value of efficiency for the other will be fixed. Therefore, the analyser may be set up by choosing an appropriate efficiency in each channel. This can be done

either by different amplification in both channels or by different window width and lower level discriminator settings, or by a combination of both.

The present problem - counting ^{45}Ca and ^{22}Na - can be solved by application of the doubly-labelled sample-counting method, i.e., a chemical separation of the two elements is not necessary (^{45}Ca : β^- , $E_{\text{max}} = 0.25$ MeV; ^{22}Na : β^+ , $E_{\text{max}} = 0.54$ MeV). To get high efficiencies, a series of tests was performed. Finally, the amplification of both channels was put to a value slightly lower than the balance point position of ^{22}Na for maximal window width (9.5 V). Window width and lower level discriminators were adjusted such that each radioelement reached approximately 50% efficiency in its characteristic channel and about 15% in the other channel. The four efficiencies were determined by means of calibrated standards and the results controlled by counting doubly-labelled samples of standard activities. The following efficiencies^{*)} were found:

^{45}Ca :	channel A	$\epsilon_{11} = 48.4\%$
	channel B	$\epsilon_{21} = 12.6\%$
^{22}Na :	channel A	$\epsilon_{12} = 17.3\%$
	channel B	$\epsilon_{22} = 57.8\%$

For comparison the efficiencies of the pure radioelements at their respective balance point and maximum window width are given as well:

^{45}Ca :	$\epsilon = 84.8\%$
^{22}Na :	$\epsilon = 83.2\%$

For doubly-labelled samples containing unknown quantities of two radionuclides the respective activities can be calculated from the count rates in the two different channels and from the four efficiencies. The formula for this calculation is easily derived and given below:

$$A_{\text{Ca}} = \frac{\epsilon_{22}\text{CR}_A - \epsilon_{12}\text{CR}_B}{\epsilon_{11}\epsilon_{22} - \epsilon_{21}\epsilon_{12}} \quad (1a)$$

$$A_{\text{Na}} = \frac{\epsilon_{11}\text{CR}_B - \epsilon_{21}\text{CR}_A}{\epsilon_{11}\epsilon_{22} - \epsilon_{21}\epsilon_{12}} \quad (1b)$$

where

- $A_{\text{Na, Ca}}$: activity of the respective isotope (dpm)
- $\text{CR}_{A, B}$: count rate in channel A or B, respectively (cpm)
- ϵ_{ij} : counting efficiency for Ca ($j = 1$) and for Na ($j = 2$) in channel A ($i = 1$) and channel B ($i = 2$) as given numerically above.

The quantity $(\epsilon_{11}\epsilon_{22} - \epsilon_{21}\epsilon_{12})$ amounts to 0.258 for the numerical values given above.

*) Note: a) Efficiencies for ^{22}Na are given in respect to the total activity. Because of the branched decay (β^+ : 89.8%; K: 10.2%) the efficiency in respect to β^+ -decay would be higher, for instance 92.5% at balance point.
 b) Salt concentrations (CaCl_2 and NaCl) of the calibration standards were matched to be similar to those of the earth samples to obtain equal quenching.

After the chemical treatment described in Section 3 the samples consisted of a known volume of a solution, which contained alkali and alkaline earth chlorides in 0.01N HCl. 1.5 ml of this volume were pipetted into a plastic counting bottle, 15 ml of scintillator added, and this was shaken for good mixture and counted for 100 minutes in each of the two analyser channels. Counting was repeated for better statistics and two preparations of every sample were measured in the same way.

A background sample was prepared by dissolving certain quantities of CaCl_2 , KCl and NaCl - corresponding to the chemical composition of the soil and the concentration in the samples - in a known volume of 0.01N HCl and counted several times in the same way as the soil samples. This was done to eliminate errors due to ^{40}K content of the soil samples. Background count rates amounted to 2035 counts per 100 minutes in channel A and to 1580 counts per 100 minutes in channel B.

The ^{40}K spectrum was found to have its maximum above the upper discriminator level of channel B ($E_{\text{max}} = 1.33 \text{ MeV}$) and did not influence the count rates in channels A and B very much. The count rate from upper discriminator level to indefinite for channel B was 1790 counts per 100 minutes.

A cross-check of the results was performed in measuring the γ -activity of the chemically-treated liquid samples with a 3×3 in. NaI crystal, which had been calibrated by means of a ^{22}Na standard solution of the same volume under identical geometry conditions.

5. RESULTS FOR THE EARTH SAMPLES

In the liquid scintillation counter, sample count rates in the range of 2065 to 2600 counts per 100 minutes for channel A and of 1620 to 2800 counts per 100 minutes for channel B were measured. With Eqs. (1a) and (1b) - see Section 4 - the respective activities of ^{45}Ca and of ^{22}Na have been calculated, taking into account the concentration factor known from the volume of the sample after chemical treatment and its initial weight. The results are given in picocuries per gram of earth (water content included) and corrected for decay from the date of the shut-down of the Proton Synchrotron to the day of measurement.

In Table 2 the measured activity concentrations of ^{45}Ca are given for every sample, while Table 3 contains the results obtained for ^{22}Na for both counting techniques applied. For comparison, the activity concentrations calculated from the flux data available (see Section 6) are listed in the second line of the tables together with the flux values used for calculation in the third and fourth line.

The errors mentioned in Tables 2 and 3 have been calculated from the combined statistical errors of sample and background count rates. Errors due to losses during the chemical treatment - especially Ca could have been adsorbed on the surface of hydroxide precipitates - can only be estimated. If the conditions are carefully maintained in the single steps of separation, the "chemical error" should not be larger than -2% to -5% of the concentration found. Larger errors may be caused by different quenching of the scintillation process, if the salt concentrations in the samples vary considerably. If the efficiencies ϵ_{ij} decrease by 10%, the calculated activities of ^{45}Ca and of ^{22}Na will increase by about 11%, but this order of magnitude is still much smaller than the limits given by the statistical errors and can be disregarded in practice because of the nearly constant composition of the treated samples.

Table 2.
⁴⁵Ca activity measured and calculated.
 Fluxes used for the calculation are corrected as described in Section 6.
 Efficiency assumed for target 1: 40%.

Sample No.	⁴⁴ Ca(n,γ) ⁴⁵ Ca									
	S(1;400;100)	S(1;200;100)	S(1;400;98)	S(1;200;98)	S(19;600;98)	S(19;400;98)	S(58;600;100)	S(58;400;100)	S(32;600;100)	S(32;400;100)
Specific activity measured pCi/g	0.600 (±0.186)	2.062 (±0.412)	0 (±0.302)	1.436 (±0.356)	0 (±0.242)	0 (±0.196)	0 (±0.214)	0.570 (±0.382)	0 (±0.104)	0 (±0.163)
Specific activity calculated pCi/g	0.89	5.94	< 0.89 *)	< 5.94 *)	0.015	0.09	0.042	0.25	0.03	0.06
Thermal neutron fluxes used n/cm ² sec	1.43 × 10 ³	9.57 × 10 ³	1.43 × 10 ³	9.57 × 10 ²	26	156	69	410	~ 50	~ 100

*) Calculated as for S(1;400;100) and S(1;200;100), but slightly smaller because of larger shielding thickness from the beam.

Table 3

^{22}Na activity concentration, measured and calculated.
Fluxes used for calculation corrected as described in Section 6.
Efficiency assumed for target 1: 40%.

Sample No.	$^{23}\text{Na}(n,2n) \begin{matrix} > \\ > \end{matrix} ^{22}\text{Na}$ $^{27}\text{Al}(n,\text{spall.})$									
	S(1;400;100)	S(1;200;100)	S(1;400;98)	S(1;200;98)	S(19;600;98)	S(19;400;98)	S(58;600;100)	S(58;400;100)	S(32;600;100)	S(32;400;100)
Specific activity measured pCi/g										
Liquid scintillation counter (β)	0.413 (± 0.117)	6.932 (± 0.260)	0.505 (± 0.252)	1.513 (± 0.210)	0 (± 0.242)	0.557 (± 0.395)	0 (± 0.263)	0.207 (± 0.207)	0.158 (± 0.226)	0 (± 0.163)
NaI-crystal (γ)	0.46	6.50	0.52	1.73	-	0.23	-	0.12	-	-
Specific activity calculated pCi/g	2.26	18.17	< 2.26 *)	< 18.17 *)	0.02	0.17	0.05	0.45	< 0.01	0.05
Fast neutron flux used n/cm ² sec	3.35×10^3	3.35×10^4	3.35×10^3	3.35×10^4	36.5	365	95.8	958	12.3	123
High-energy neutron flux used n/cm ² sec	2.57×10^3	1.8×10^4	2.57×10^3	1.8×10^4	41.4	280	108.7	735	14	95

*) Calculated as for S(1;400;100) and S(1;200;100), but slightly smaller because of larger shielding thickness in lateral position.

6. CALCULATION OF INDUCED RADIOACTIVITY IN SOIL

For a radionuclide of half-life τ the build-up of activity during irradiation over a time t is given by

$$A(t) = A_0(1 - \exp[-\frac{0.693 t}{\tau}]), \quad (2)$$

where $A(t)$ is the activity at time t and A_0 the saturation activity. $A(t)$ and A_0 are expressed in disintegrations per second. The saturation activity is defined to be that activity where as many radioactive atoms decay per unit time as are formed. It can be calculated by the following expression:

$$A_0 = \phi \cdot \sigma \cdot N \cdot a \quad (3)$$

where

ϕ : neutron flux per $\text{cm}^2 \text{ sec}$

σ : cross-section for the respective nuclear reaction in cm^2

N : number of atoms of the respective element (per gram, if $A(t)$ is the activity per gram)

a : isotopic abundance of the isotope reacting.

For the period considered, the Proton Synchrotron was running for 375 days, shut-down intervals included (17 May 1967 to 23 May 1968). The shut-down intervals amounted to 80 days. But these figures will only be considered in respect to average neutron flux calculations, because the CERN Proton Synchrotron has been running regularly for several years and thus saturation can be assumed for the radioisotopes concerned, not taking into account possible losses by rain water erosion.

Cross-sections for the different reactions were taken from up-to-date compilations^{2,3,24}). As the spectral distribution of neutrons is not well known, the chosen cross-sections are a compromise for the energy range concerned. Therefore, the respective results of activity calculation should not be regarded as absolute but as approximated values. Fast neutron (25 keV - 14 MeV) fluxes had been measured by means of moderated indium detectors and by the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reaction [practical threshold: 7 MeV ¹¹], while high-energy (above 20 MeV) neutron fluxes had been determined by the $^{12}\text{C}(n,2n)^{11}\text{C}$ reaction [practical threshold: 20 MeV ¹¹].

The number of atoms per gram of substance followed from the soil analysis performed for the CLR-shielding experiment by the Laboratoire d'Essai des Matériaux de l'Université de Lausanne¹⁸). These values were corrected by comparing the water content at the time of sampling (11%, see Section 3) with that of the samples analysed during the CLR-shielding experiment.

The main difficulty consisted in finding the appropriate values for the neutron fluxes of different neutron energies. The figures given in the CLR-shielding experiment results⁹) are normalized to 10^{12} p/sec circulating in the accelerator and have been measured at 13 GeV and at 25.6 GeV particle energy. For use in the following calculations they have

Table 4

Calculated saturation activities for all long half-life radioelements formed in the earth shielding above target 1 at 200 g/cm² shielding thickness from beam.

Radioisotope formed	Nuclear reaction	Cross-section (mb) at respective neutron energy	Half-life	Atoms of parent element per gram of soil (corrected for water content)	Isotopic abundance	ϕ (n/cm ² sec) corrected, at 200 g/cm ² shielding thickness	Saturation activity pCi/g	mpc ^{16,27} 168 h/week occup. exposure pCi/ml
⁵⁹ Fe	⁵⁸ Fe(n, γ) ⁵⁹ Fe	1200 (th.n.)	45 d	2.821×10^{20}	⁵⁸ Fe: 0.31	9.57×10^3	0.27	600
⁴⁵ Ca	⁴⁴ Ca(n, γ) ⁴⁵ Ca	1100 (th.n.)	153 d	1.000×10^{21}	⁴⁴ Ca: 2.07	9.57×10^3	5.94	90
³⁵ S	³⁴ S(n, γ) ³⁵ S	260 (th.n.)	87 d	3.357×10^{18}	³⁴ S : 4.20	9.57×10^3	< 0.01	600
³² P	³² S(n,p) ³² P	380 (11 MeV)	14.2 d	3.357×10^{18}	³² S :95.04	3.35×10^4	1.10	200
⁵⁴ Mn	⁵⁴ Fe(n,p) ⁵⁴ Mn	500 (6 MeV)	278 d	2.821×10^{21}	⁵⁴ Fe: 5.84	3.35×10^4	7.46	1000
	⁵⁵ Mn(n,2n) ⁵⁴ Mn	1100 (18 MeV)		3.430×10^{18}	⁵⁵ Mn: 100		<u>3.57</u> \sum 11.03	
²² Na	²² Na(n,2n) ²² Na	70 (20 MeV)	2.6 y	1.233×10^{20}	²³ Na: 100	3.35×10^4	7.81	400
	²⁷ Al(n,spall.) ²² Na	20 (100 MeV)		1.134×10^{21}	²⁷ Al: 100	1.8×10^4	<u>10.36</u> \sum 18.17	
⁴⁶ Sc	⁵⁵ Mn(n,spall.) ⁴⁶ Sc	5.7 (170 MeV)	84 d	3.430×10^{18}	⁵⁵ Mn: 100	1.8×10^4	< 0.01	400
⁷ Be	¹⁶ O(n,spall.) ⁷ Be	10 (1 GeV)	53 d	1.705×10^{22}	¹⁶ O : 99.76	$<1.8 \times 10^4$	< 77.9	20,000

to be corrected for the average proton flux over long periods (0.38×10^{12} p/sec) and interpolated to 18.4 GeV proton energy, the mean energy of protons for long working periods of the accelerator. Furthermore, the samples are taken at different depths measured from the top of the earth shield, thus the respective neutron fluxes have to be interpolated from data for different shielding thickness^{18,25}). And, besides, the target efficiency has to be taken into account to calculate the fraction of neutrons of different energies released from the target in respect to the normalized values²⁶). (A target efficiency of 40% was assumed for target 1 and the fluxes at the other positions calculated from the number of high-energy particles measured at every magnet of the Proton Synchrotron ring relative to that number at target 1.) After all, the resulting flux over the period considered is averaged by applying a correction factor known from the length of shut-down intervals and from the total working period of the machine. This factor amounts here to $(375-80)/375 = 0.787$. The final corrected flux is assumed to be characteristic for several working years of the Proton Synchrotron and used for the calculation of saturation activities (the corrected flux values can be found in lines 3 and 4 of Tables 2 and 3).

The total shielding thickness from beam level to the sample location was calculated from the known cross-sections of the CERN Proton Synchrotron concrete tunnel plus earth shielding and from the geometry as in the CLR-shielding experiment^{18,25}). For samples taken at 1 m depth from the top of the shielding 400 g/cm² shielding thickness were taken for target 1, 600 g/cm² for the other positions (less earth on target 1!). The second sample at 2 m depth corresponds to 200 g/cm² at target 1 and to 400 g/cm² at the other points. The shielding thickness of the concrete roof of the tunnel is included in these figures.

In Table 4 (column 8) the calculated saturation activities of the radioisotopes formed (considering the restriction to half-lives longer than 10 days) are listed for the case of maximum activation, which takes place near target 1 and for the samples examined at 200 g/cm² shielding thickness. The cross-sections of the respective nuclear reactions (column 3), half-lives (column 4), concentration of parent element per gram of soil (column 5), isotopic abundance (column 6), and the corrected flux values used (column 7) are given as well as the ICRP values of maximum permissible concentration (mpc) per millilitre of water¹⁶) (column 9).

7. DISCUSSION

The ⁴⁵Ca and ²²Na activities measured in soil samples of the CERN 28 GeV Proton Synchrotron tunnel earth shielding agree very well with the induced activities calculated (see Tables 2 and 3).

For the region of highest activation (near target 1) approximately the same relation between measured and calculated activities has been found for ⁴⁵Ca in samples S(1;200;100) and S(1;200;98), and for ²²Na in samples S(1;200;100), S(1;400;100), and S(1;400;98). Discrepancies as in sample S(1;400;100) for ⁴⁵Ca might be related to larger thermal neutron fluxes than assumed. Flux values might as well need correction in the calculation of ²²Na concentration for sample S(1;200;98), because the lateral positions have thicker shielding while the same fast and high-energy neutron flux values have been applied as for positions above the axis of the beam.

For samples S(58;400;100) for ^{45}Ca and S(19;400;98) as well as S(58;400;100) for ^{22}Na the poor counting statistics exclude a definite discussion. For all other samples the calculation explains that practically no activity could be measured.

The measured activities near target 1 are on an average about a factor of three smaller than the calculated activities. This had been expected because of rain water erosion of the whole shielding.

If it is assumed that rain water runs very slowly through the shielding such that equilibrium between calcium and sodium ions in solid phase and in solution is always maintained, the relation of radioactive isotopes to inactive isotopes of the same element should be the same for liquid and solid phase. This kind of equilibrium seems very probable in respect to the quantities of soil and rain water and their relation: about 1000 l of rain ^{*)} fall per year on 1 m², but 1 m² corresponds for the shielding to an earth column of about 3.5 m length, i.e., about 7 tons of material of density 2 g/cm³.

In flowing slowly through the earth column the rain water will take up sodium and calcium salts (mainly silicates, carbonates, sulphates) from the soil up to the respective solubilities for salt mixtures. It can be assumed that equilibrium between ions in liquid and solid phase is already reached when the water arrives at the more active zones near the concrete tunnel, where the second equilibrium of isotopic exchange is adjusted.

If concentrations of about 45 ppm of Na⁺ and 80 ppm of Ca⁺⁺ are taken as representative for the water at equilibrium with soil, the relation between the respective specific concentrations (for soil: see Table 4) will be 1 : 100 for sodium and 1 : 1000 for calcium. Under the assumption of equilibrium distribution of activity between both phases, drainage water coming from "hot places" in the shielding will contain at least 180 pCi/l of ^{22}Na and 6 pCi/l of ^{45}Ca , taking the soil activity concentrations at 200 g/cm² shielding thickness near target 1.

A number of samples has been taken directly from the drainage water at inspection openings (survey pillars 1-8) near magnets 1, 14, 26, 39, 51, 64, 76, and 89. At these places the drainage of water collected from the inner side of the accelerator ring and that of water collected from the outside environments are interconnected. It should be pointed out here that the surface area from which the water is flowing into the drainage system is much greater than the surface of the tunnel, and that water from an active area might be found at a different place due to a certain inclination of the drainage tubes. Besides, the specific activity of the soil will surely be higher the nearer is the concrete tunnel, and a considerable activity could be washed out from the soil at the outer surface of the concrete wall, which has a much higher specific activity than the earth shielding above⁶⁾. Therefore, it is not in contradiction to the activity concentration found in soil that most of the drainage water samples showed only background activity, while two of them contained measurable quantities of ^{22}Na : 550 pCi/l near magnet 26 (survey pillar 3), and 380 pCi/l near magnet 76 (survey pillar 7).

^{*)} Average over 30 years for the Geneva region: 93.3 cm/m². Communication of the Meteorological Centre of the Geneva Airport, Genève-Cointrin.

If the proton intensities of the CERN Proton Synchrotron are increased by a factor of 20 - without changing the percentage of protons allotted to target 1 - the specific activities induced will also be higher by about the same factor. For a 300 GeV proton accelerator this factor of increase may be around 400 (a factor of about 25 for intensity increase multiplied by a factor of about 15 for proton energy increase) compared with the present conditions of the CERN Proton Synchrotron. Using as a basis the values measured for the drainage water, specific ^{22}Na concentrations of about 10 pCi/ml could be reached near hot regions for improved proton intensities at the CERN Proton Synchrotron. This is still a factor of 40 below the maximum permissible concentrations and well on the safe side. For a 300 GeV accelerator, the ^{22}Na concentration would be around 200 pCi/ml, i.e., near the maximum permissible concentration of 400 pCi/ml. For bad meteorological conditions, for instance, heavy rain after a long period of dry weather, another factor of 5-10 might have to be applied to the estimated concentration.

Because of the relation assumed between activity concentration in solid and liquid phase, the soil of the shielding will become rather radioactive at low shielding thicknesses near hot places, and specific activities induced will already reach maximum permissible concentrations for a secondary particle flux increase by a factor of 20.

From the data measured and calculated for ^{45}Ca and for ^{22}Na , and their agreement, it may as well be assumed that ^{32}P and ^{54}Mn should show about the same distribution between drainage water and soil as the examined ions. In that case, taking a surely pessimistic distribution factor of 1 : 100 for the relation of ions in liquid and solid phase, the activities at present conditions would reach 11 pCi/l for ^{32}P and 110 pCi/l for ^{54}Mn . These concentrations are factors of 2×10^4 for ^{32}P and of 10^4 for ^{54}Mn below maximum permissible concentrations and would even not reach these limits in the drainage water of a 300 GeV accelerator, while the activity concentration in the soil could already be 2-4 times larger than maximum permissible concentrations.

For ^7Be , which is formed by spallation reactions, the build-up of an equilibrium between ions in solution and ions in solid phase is not possible because of the lack of inactive beryllium in soil. On the other hand - looking at the chemistry of beryllium, especially in neutral solutions - it seems quite improbable that the ^7Be built should be totally dissolved in water penetrating the soil. It is even most probable that the greatest part of ^7Be formed directly in the water contained in the soil by spallation of oxygen will be absorbed and filtered off by the earth. Therefore, a relation of 1 : 100 for the distribution of ^7Be between water and soil may even be exaggerated. But taking this distribution factor, the activity of ^7Be in drainage water derived from the calculated production in soil at 200 g/cm² shielding thickness would be 800 pCi/l, i.e., a factor of 2.5×10^4 below maximum permissible concentrations, and these limits would also not be reached for a 300 GeV accelerator.

For the present paper, ground water activation has not been taken into account. Middelkoop⁴⁾ has estimated ^7Be concentrations in ground water near a 300 GeV proton accelerator. He assumed a continuous 20% beam loss of the 300 GeV protons (10^{13} p/sec) at one place, which is in turn taken as point source. After this estimation, at 2m distance from this source of fast neutrons (10-100 MeV energy) 1.6 μCi of ^7Be could be built per cm³ of rock and soil containing 15% of water. The reaction cross-section for fast neutrons in this energy interval was assumed to be 1 mb.

Inside the accelerator tunnel - without shielding - the highest fast neutron flux measured (sulphur as threshold detector) during the CLR-shielding experiment at 2m distance from a source of radiation was 10^7 particles per cm^2 and second. From this figure, also taking a cross-section of 1 mb and a water content of 15%, the maximum ^7Be saturation activity would be $0.0014 \mu\text{Ci}$ per cm^3 of soil at present working conditions of the CERN Proton Synchrotron. For a 300 GeV accelerator, assuming as above a 400 times higher flux of secondaries than at present, the ^7Be saturation activity in ground water would increase to $0.55 \mu\text{Ci}$ per cm^3 in certain hot regions. This value is in nearly perfect agreement with that proposed by Middelkoop taking into account that he could only estimate fluxes, while the flux assumed in this paper is partly based on measurements.

If dilution processes from the hydrology of the site, water flow rate, distribution of secondary particle sources, and chemistry of beryllium, are respected for the discussion, the ^7Be concentration in the ground water around a 300 GeV proton accelerator as given above might considerably decrease. But it has to be pointed out as well that the activities could be more important than estimated near beam dumps of internal or external proton beams.

After all, it can be concluded that at working conditions as those of the CERN Proton Synchrotron at present - average proton flux 3.8×10^{11} , average energy 18.4 GeV - no hazard exists from the effluent drainage water or even from a manipulation of the soil near target regions. But it cannot be ruled out that radioactive isotopes, especially ^{22}Na and ^7Be , near or above maximum permissible concentrations may be washed out from the soil surrounding future high-energy accelerators or be directly built in the ground water, and a regular control of this water is recommended, while soil or rock near target regions or other points of beam losses should only be manipulated with the necessary precautions.

Acknowledgements

The author gratefully acknowledges the helpful and stimulating discussions he had with Drs. K. Goebel and W.C. Middelkoop.

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