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AIRBORNE RADIOACTIVITY PRODUCED AT
HIGH-ENERGY ACCELERATORS

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1. INTRODUCTION

During accelerator operation radioactive gases are produced by the interaction between the primary and secondary particles from the machine and the atmospheric air in the accelerator halls. Spallation reactions in solid machine parts can also lead to the formation of radioactive gases.

If the air is confined in the accelerator hall there will be no release of radioactivity into the outside zones during operation of the machine. When the machine is stopped an unacceptable concentration of radioactivity in the air may be present which requires decay or dilution to reduce it to acceptable levels before people enter the hall.

In the present work we indicate the most important radioactive isotopes which can be formed from the air of a high-energy proton accelerator and we calculate the relative amount of them produced for different conditions at an accelerator like the CERN 28 GeV Proton Synchrotron. These calculations are checked by comparing them with the measurements carried out at the CERN accelerators.

Maximum permissible concentrations of these radioactive gases (which are not in the ICRP Tables) are calculated and allow us to evaluate the risk for different operations in the accelerator halls.

2. GASEOUS RADIOACTIVE NUCLIDES PRODUCED IN THE ATMOSPHERIC AIR

Table 1 lists all the radioactive isotopes with a half-life > 1 sec (in decreasing half-life order) which can be produced by irradiation of the atmospheric air in a proton accelerator hall, from spallation, (γ , n) reaction and thermal neutron capture. The contribution from other possible reactions with high-energy neutrons is negligible.

In columns 6 and 7 of Table 1 are indicated the type of the reactions and their relative average cross-sections for the production of the radio-isotopes considered; the cross-sections for the spallation and (γ , n) reaction are the average of the values quoted by Bruninx¹⁾.

3. CALCULATION OF THE SPECIFIC RADIOACTIVITY - THE CERN PS CASE

The following equation is used to calculate the total specific radioactivity ($\mu\text{Ci/litre}$) in the air as a function of the exposure time and activating fluxes:

$$\alpha = \beta \sum_k \left[\sum_i \bar{\Phi}_1 N_i \sigma_{i,k} + \sum_j \bar{\Phi}_2 N_j \sigma_{j,k} \right] \left(1 - e^{-\lambda_k T} \right) e^{-\lambda_k t} \quad (1)$$

In Appendix 1 the explanation of the symbols used and the method of derivation are indicated.

From the particle flux measurements made in the CERN PS^{*)} hall we extrapolated (Appendix 2) average fluxes of very high energy particles and thermal neutrons which were introduced in Eq. (1). The results of the calculation are summarized in Table 2. On the basis of this table, the following deductions can be made.

- i) The saturation of all the radionuclides is very rarely reached since the normal running period seldom exceeds 15 days and the air is renewed; consequently, the values indicated in column 1 are excessive.

*) A 28 GeV proton synchrotron of an intensity of the order of 10^{12} p/sec.

- ii) A renewing cycle of 30 min is a good approximation for the CERN PS; it is interesting to see that the total specific activity for such an irradiation time is only about a factor of 2 lower than the activity at saturation.
- iii) After 5 min shutdown the specific activity is reduced by a factor of 2. The contribution of the different radio-isotopes changes slightly from the contribution at machine stop: ^{13}N remains the main contributor whereas the contribution of ^{11}C and ^{41}A increases.

4. MEASUREMENTS AT THE CERN ACCELERATORS

The instrument used for the measurements was a gas-flow monitor consisting of an ionization chamber through which the air was circulating continuously^{*)}. The air was drawn through tubing connecting the location of measurements in the accelerator hall to the monitoring instrumentation placed in a low-radiation area outside the accelerator shielding; this causes a delay of 3 to 5 minutes from the moment of sampling to the time of measurement, while the air is traversing the pipe. The decay of the mixture of the gases was followed by stopping the gas flow, and from the decomposition of the decay curves the identification of the main components could be made. Figure 1 shows the decay measured at the **two machines**. In the figure is also given a computed curve for the CERN PS assuming a 30 minute exposure time, a 5 minute decay and radioactivity due to ^{41}A : 14%; ^{11}C : 31%; ^{13}N : 47% and ^{15}O : 8%.

In Table 3 the results of the measurements are summarized. The slightly higher values of the specific activity measured at the CERN PS machine as compared to the calculated ones, may be due partly to the presence of aerosols and dust in the air, which have not been stopped by the filter and the de-ionizing chamber of the measuring instrument and which have not been taken into account in the calculation, and partly to the uncertainty in evaluating the particle fluxes and the air-renewing cycle in the machine.

*) The instrument is a EIL 53C portable gas monitor - ^{85}Kr gas has been used for calibration.

The ^{41}A contribution which is quite visible in the curve of Fig. 1 does not seem to be more than 10% of the total activity.

The measurements made in the hall of the CERN Synchro-cyclotron*) at different locations also reported in Table 3 and Fig. 1, showed a slightly higher concentration with a higher ^{41}A contribution. At the point where the maximum concentration, reported in Table 3, was measured, an average flux of 1.5×10^6 part. $\text{cm}^{-2} \text{ sec}^{-1}$ of very high energy was found with an ^{11}C -detector. Measurements made in a tight box placed in the path of the 600 MeV extracted proton beam and at 1 m from a target placed in the extracted beam, showed the presence of a nuclide of about 4h half-life which is assumed to be $^{85\text{m}}\text{K}$ probably produced by spallation with elements other than air gases. No ^{41}A was found in these cases, as expected, owing to the fact that only high-energy particles were present.

5. MAXIMUM PERMISSIBLE CONCENTRATIONS

ICRP (International Commission on Radiological Protection) states²⁾ that submersion is the limiting mode of exposure for inert β or γ emitting gases. In using the formulae recommended by ICRP we calculated the $(\text{MPC})_a$ for the radioactive gases found at the accelerators and which are not indicated in the ICRP tables.

In Table 4 the calculated values for a dose-rate of 0.1 rem/week and an exposure time of 40 hours/week are indicated.

If we weigh the contribution of the different radioactive gases to the total air radioactivity (^{13}N : 47%; ^{11}C : 31%; ^{41}A : 14%; ^{15}O : 8%) an average value of about $5 \times 10^{-3} \mu\text{Ci/l}$ for the MPC is found. This value is 10 times higher than the value quoted first by R. Thomas³⁾ but it is in quite good accordance with the value ($2 \times 10^{-3} \mu\text{Ci/l}$) proposed by A. George et al.⁴⁾ for a LINAC, and reported lately by R. Thomas⁵⁾.

An advisable value of $10^{-3} \mu\text{Ci/l}$ can be accepted for unknown mixtures of such radioactive gases.

*) A 600 MeV proton synchro-cyclotron of 1.5 μA .

6. CONCLUSIONS

Values 10^3 times higher than the MPC have been measured under exceptional conditions. Under normal machine operation values ranging from 10 to 30 times the MPC can easily be found in the machine halls, 5 minutes after the machine stop. Even though an average half-life of about 20 min for the total concentration is admitted, the risk for persons involved in frequent maintenance work during short shutdown periods can become important if swift renewal of the air is not provided. At the Synchro-cyclotron, for instance, 10 exposures of 6 min after a cooling time of 5 min may cause a total exposure equivalent to ~ 50 mrem.

Under the present conditions of the CERN accelerators and with the understanding that the ventilation apparatus will continue functioning during the shut-down periods, it is possible to consider the radioactive gases as a second degree danger as compared to the risks of external irradiation from the machine parts.

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Table 1

Radio-nuclide	Half-life	Type of decay (MeV - %)	Parent element	Fraction by weight of parent element in air	Production reaction	Cross-section mb
${}^3_1\text{H}$	12.2 y	β^- (0.018 - 100%)	C	1.6×10^{-4}	spallation	
			N	0.755	spallation	30
			O	0.23	spallation	30
${}^7_3\text{Be}$	53 d	γ (0.48 - 12%) E.C.	C	see above	spallation	
			N	" "	spallation	10
			O	" "	spallation	10
${}^{41}_{18}\text{A}$	1.8 h	β^- (1.2 - 99%) γ (1.3 - 99%)	${}^{40}_{18}\text{A}$	0.013	thermal capture	610
${}^{11}_6\text{C}$	20.5 min	β^+ (0.9 - 100%)	C	see above		
			N	" "	spallation	20
			O	" "	spallation	20
${}^{13}_7\text{N}$	10 min	β^+ (1.25 - 100%)	N	" "	spallation and (γ , n)	30 ^{*)}
			O	" "	spallation	10

*) This is the total cross-section. The spallation cross-section is about 10 mb and the (γ , n) reaction cross-section is assumed to be 10 mb. We suppose that the high-energy γ -ray flux is double that produced by high-energy particles; consequently, a total cross-section of 30 mb can be used.

Table 1 (continued)

Radio-nuclide	Half-life	Type of decay (MeV - %)	Parent element	Fraction by weight of parent element in air	Production reaction	Cross-section mb
$^{15}_8\text{O}$	2.1 min	β^+ (1.7 - 100%)	O	see above	(n, 2n) and (γ , n)	60 ^{**})
$^{14}_8\text{O}$	74 sec	β^+ (1.8 - 99%) γ (2.3 - 99%)	O	" "	spallation	5
$^{19}_3\text{O}$	29 sec	β^- (4.5 - 30%) (2.9 - 70%) γ : various	^{18}O	4.2×10^{-4}	thermal capture	
$^{10}_6\text{C}$	19 sec	β^+ (2.1 - 99%) γ (0.7 - 99%)	C	see above	spallation	
$^{16}_7\text{N}$	7.3 sec	β^- : various γ : various	^{15}N O	0.0028 see above	thermal capture (n,p) spallation	60
$^{17}_7\text{N}$	4.1 sec	β^- n			***)	
$^{15}_6\text{C}$	2.3 sec	β^- (4.5 - 80%) and (9.5 - 20%) γ (5.3 - 20%)			***)	

**) Total cross-section. The (n,2n) reaction cross-section is of 40 mb. On the same assumption as above a total cross-section of 60 mb can be used.

***) The production of these isotopes is very improbable.

Table 2

Specific activities calculated for the CERN Proton Synchrotron

At saturation			After 30 min of exposure			After 30 min of exposure and 5 min of decay		
Total ($\mu\text{Ci/l}$)	Contribution of parent elements*) (%)	Contribution from different radio- isotopes**) (%)	Total ($\mu\text{Ci/l}$)	Contribution of parent elements*) (%)	Contribution from different radio- isotopes**) (%)	Total ($\mu\text{Ci/l}$)	Contribution of parent elements*) (%)	Contribution from different radio- isotopes**) (%)
40.5×10^{-3}	N (50%)	^3H (21.5%)	19×10^{-3}	N (47%)		9.8×10^{-3}	N (69%)	^{41}A (13.5%)
		^7Be (7.2%)			^{41}A (7%)			^{11}C (31%)
	O (30%)	^{41}A (20%)		O (46%)	^{11}C (19%)		O (17.5%)	^{13}N (47%)
		^{11}C (14.5%)			^{13}N (34%)			^{15}O (8%)
	A (20%)	^{13}N (18.8%)		A (7%)	^{15}O (19%)		A (13.5%)	
		^{15}O (9%)			^{14}O (1.6%)			
		^{14}O (0.7%)			^{16}N (19.4%)			
		^{16}N (9%)						

*) The contribution from C is excluded from the calculation as the C concentration in air and the cross-sections of the reactions with C are too small.

**) Only radio-isotopes which have a $> 0.5\%$ contribution and whose production is probable have been taken into account.

Table 3

Maximum values of specific radioactivity
of the air measured at the CERN accelerators

(5 min after machine stop)

Location	Total ($\mu\text{Ci/l}$)	Contribution from different radio-isotopes
In the ring of the CERN Proton Synchrotron (near target regions)	$\sim 10^{-2}$	^{13}N ($\sim 50\%$); ^{11}C ($\sim 30\%$); ^{41}A ($< 10\%$) other isotopes : $> 10\%$.
In the hall of the CERN Synchro-cyclotron (near target region)	$\sim 3.5 \times 10^{-2}$	^{11}C ($\sim 55\%$); ^{41}A ($\sim 22\%$); ^{13}N ($\sim 12\%$) other isotopes: $\sim 10\%$.
In the extracted 600 MeV proton beam of the CERN Synchro-cyclotron	8	^{11}C ($\sim 67\%$); isotopes with half-life < 20 min: ($\sim 32\%$); presence of $^{85\text{m}}\text{Kr}$; no ^{41}A .
Near a target placed in the path of the extracted 600 MeV proton beam of the CERN Synchro-cyclotron.	2×10^{-1}	^{11}C ($\sim 23\%$); isotopes with half-life < 20 min: ($\sim 76\%$); presence of $^{85\text{m}}\text{Kr}$; no ^{41}A .

Table 4

Maximum permissible concentrations in the air

Radio-nuclide	(MPC) _a ($\mu\text{Ci/l}$)
^3_1H	2
^7_4Be	6×10^{-3}
$^{41}_{18}\text{A}$	2×10^{-3}
$^{11}_6\text{C}$	8×10^{-3}
$^{13}_7\text{N}$	5×10^{-3}
$^{15}_8\text{O}$	3×10^{-3}
$^{16}_7\text{N}$	5×10^{-4}

CALCULATION OF THE SPECIFIC RADIOACTIVITY
OF THE AIR IN AN ACCELERATOR

The radioactive nuclides which can be formed by spallation and thermal neutron capture in the atmospheric air are listed in Table 1. In this table are also listed the production cross-sections, the abundance of the parent element or isotope in the atmospheric air and the half-life and emission of the product.

The calculation can be derived from the following equation

$$\alpha = \beta \sum_{i,k} N_i \bar{\Phi}_{i,k} \bar{\sigma}_{i,k} \left(1 - e^{-\lambda_k T}\right) e^{-\lambda_k t} \quad (1)$$

where

α = total specific radioactivity ($\mu\text{Ci/litre}$);
 N_i = the number of atoms of the parent target element or isotope "i" in a litre of atmospheric air. This can be expressed by:

$$N_i = 1.2 \times 6.02 \times 10^{23} \frac{F_i}{A_i}$$

where:

F_i = fraction of the parent target element or isotope "i" in the air;
 A_i = the mass number of parent target or isotope "i";
 $\bar{\Phi}_{i,k}$ = average flux of activating particles which interact with the parent element "i" for the production of the radioactive isotope "k";
 $\bar{\sigma}_{i,k}$ = average cross-section of the parent target element or isotope "i" for the production of the radioactive isotope "k";
 λ_k = the decay constant of the radioactive isotope "k";
 T = exposure time (see text);
 t = time after machine stop;
 β = constant.

The nuclear processes considered are: spallation [which also includes (γ, n) reactions] and thermal capture. Equation (1) can thus be simplified as follows:

$$\alpha = \beta \sum_k \left[\sum_i \bar{\Phi}_1 N_i \sigma_{i,k} + \sum_j \bar{\Phi}_2 N_j \sigma_{j,k} \right] \left(1 - e^{-\lambda_k^T} \right) e^{-\lambda_k t} \quad (2)$$

where:

$\bar{\Phi}_1$ = the average thermal neutron flux;

$\bar{\Phi}_2$ = the average very high energy particle flux.

Index i refers to isotopes which undergo thermal capture reactions and index j to isotopes involved in spallation and (γ, n) reactions for the production of a radioactive isotope k.

ESTIMATE OF THERMAL NEUTRON FLUX AND
VERY HIGH ENERGY FLUX IN THE PS RING

Measurements of particle fluxes in the CERN PS ring have been made^{6,7)}. For an internal circulating beam of 10^{12} p/sec at a height of 1.5 m above the vacuum chamber $\sim 90^\circ$ from beam direction at target position, very high energy particle fluxes between 10^6 to 4×10^6 part. $\text{cm}^{-2} \text{sec}^{-1}$ have been measured according to the target type. 10 m behind the target position, in areas which can be defined as quiet areas in relation to the target influence, very high energy particle fluxes between 4×10^3 and 4×10^4 have been measured for different types and positions of target. Thermal neutron flux measurements give values (at 3 m from target, 90° from beam direction) of about 3×10^7 n $\text{cm}^{-2} \text{sec}^{-1}$ and about 10^6 n $\text{cm}^{-2} \text{sec}^{-1}$ in quiet areas, ~ 10 m behind the target. Owing to the fact that several targets are often simultaneously used and that beam losses are located in several places along the whole vacuum pipe, we applied for our calculation an average value of 2×10^5 part. $\text{cm}^{-2} \text{sec}^{-1}$ of very high energy and 2×10^6 n $\text{cm}^{-2} \text{sec}^{-1}$ for the thermal region.

In fact, the tunnel length is about 600 m; if 4 targets are used and 6 permanent loss points exist we can suppose that 10 main radiation sources are distributed along the ring, which means that the same radiation situation is repeated every 60 m along the ring.

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Figure caption

Fig. 1 : Typical decay curves of radioactive air at the two CERN accelerators and computed decay calculated assuming that the contribution of isotopes is as follows:

^{11}A : 14%; ^{11}C : 31%; ^{13}N : 47% and ^{15}O : 8%.