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## **EVALUATION OF THE RADIOACTIVE AND NOXIOUS GAS PRODUCTION FOR THE DAΦNE COMPLEX**

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### **INTRODUCTION**

One of the radiation protection problems usually considered around the accelerators is the radioactive and noxious gas production. In this note such a problem will be examined for the DAΦNE complex in order to establish the precautions to take for ensuring safe working conditions to the staff of the machines and to the other employees of the centre.

Air inside the halls housing the machines will be activated by bremsstrahlung and by neutron radiation generated during the operation of the machines. The air irradiation also causes the formation of noxious gases such as ozone and nitrogen oxides and consequently an evaluation of the production of these gases has been also made.

The concentration estimations inside the halls of the DAΦNE complex have been made on the basis of the realistic operation conditions of the machines, but conservative assumptions have been adopted in calculations, particularly regarding the values of the saturation activities.

The radioactive gases produced inside the buildings housing the machines will be released in the external environment owing to the natural air-change inside the halls or to the forced ventilation. The atmospheric dispersal could expose the workers outside the buildings and the population, up to large distances from the centre, to radiological risks. Therefore, on the basis of a model of atmospheric dispersion the risks due to the released activities from the DAΦNE complex have been evaluated.

### **RADIOACTIVE GAS PRODUCTION**

The interaction of bremsstrahlung and of neutron radiation with air around the machines of the DAΦNE complex, will give rise to the production of radionuclides like  $^3\text{H}$ ,  $^7\text{Be}$ ,  $^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$ ,  $^{16}\text{N}$ ,  $^{38}\text{Cl}$ ,  $^{39}\text{Cl}$  and  $^{41}\text{Ar}$ . The activity due to these radionuclides is in general short-lived, thereby the concentrations of activity that will be reached inside the halls housing the machines only through the radioactive decay can reduce a lot in a short time. Inside the halls there will be either natural or forced air-change that contribute to reduce the concentration levels.

The activity  $A(t)$  has been therefore calculated, according to the different operation conditions of the machines, by the following equation:

$$\frac{dA(t)}{dt} = \lambda A_s - \left(\lambda + \frac{kF}{V}\right)A(t) \quad (1)$$

where  $\lambda$  is the decay constant of the radionuclide of interest,  $F$  the ventilation rate,  $V$  the volume of the hall,  $k$  a factor assumed equal to 1/3 [Mo91] which takes into account the imperfect mixing of the radionuclides,  $A_s$  the saturation activity.

Among the radionuclides previously mentioned the most important ones from the radiation protection point of view for the DAΦNE installation are  $^{13}\text{N}$  ( $T_{1/2}=9.96$  min),  $^{15}\text{O}$  ( $T_{1/2}=123$  s) and  $^{41}\text{Ar}$  ( $T_{1/2}=1.83$  h).  $^{13}\text{N}$  and  $^{15}\text{O}$  are produced by bremsstrahlung through the  $(\gamma, n)$  nuclear reactions. The saturation activity,  $A_s$ , of  $^{13}\text{N}$  and  $^{15}\text{O}$ , used in the calculations, are obtained by the source terms proposed by Swanson [IAEA79] expressed per unit of bremsstrahlung pathlength in air and of electron beam power incident on a thick high-Z target.  $^{41}\text{Ar}$  is produced by thermal neutrons through the  $(n, \gamma)$  reaction and the saturation activity  $A_s$  is given by:

$$A_s = \phi_{th} n \sigma \quad (2)$$

where  $\phi_{th}$  is the fluence rate of thermal neutrons,  $n$  the number of  $^{40}\text{Ar}$  atoms into the hall volume and  $\sigma$  the cross section of the  $^{40}\text{Ar}(n,\gamma)^{41}\text{Ar}$  reaction. The fluence rate of thermal neutrons  $\phi_{th}$  has been evaluated according to the Moyer's formula [Pa73]:

$$\phi_{th} = 1.25 \frac{Y}{S} \quad (3)$$

where  $Y$  is the number of neutrons per second produced into the hall and  $S$  is the internal surface of such a hall. The neutron yield  $Y$  has been obtained using the data on neutron yield from thick targets per unit electron beam power according to [IAEA79].

The released activity in the environment has been calculated integrating over the time the product of the concentration of activity inside a hall by the air exchange rate. The evaluation of the radiation doses due to activity released in the environment has been made using the AQUILA code (Air quality inferred from long term average meteorological data)[De86, ENEA87a, ENEA87b].

In the following the radioactive gas production inside the halls housing Linac, Damping Ring and Main Rings will be separately analyzed.

### Linac hall

The production of radioactive gases inside the Linac hall has been evaluated considering the beam losses during the acceleration of electrons or positrons by the Linac up to 510 MeV. According to the running conditions of the machine, on which are based the shielding calculations [Es92], the major beam losses will occur during the positron run owing to the electron-positron conversion. In calculations, both the beam losses during the positron run and the ones during the electron run have been taken into account.

Table 1: Concentrations inside the Linac hall

Radionuclides	Maximum concentration (Bq/cm <sup>3</sup> )	Saturation concentration (Bq/cm <sup>3</sup> )
<sup>13</sup> N	$2.20 \times 10^{-1}$	$16.20 \times 10^{-1}$
<sup>15</sup> O	$1.03 \times 10^{-1}$	$2.04 \times 10^{-1}$
<sup>41</sup> Ar	$1.65 \times 10^{-4}$	$4.38 \times 10^{-3}$

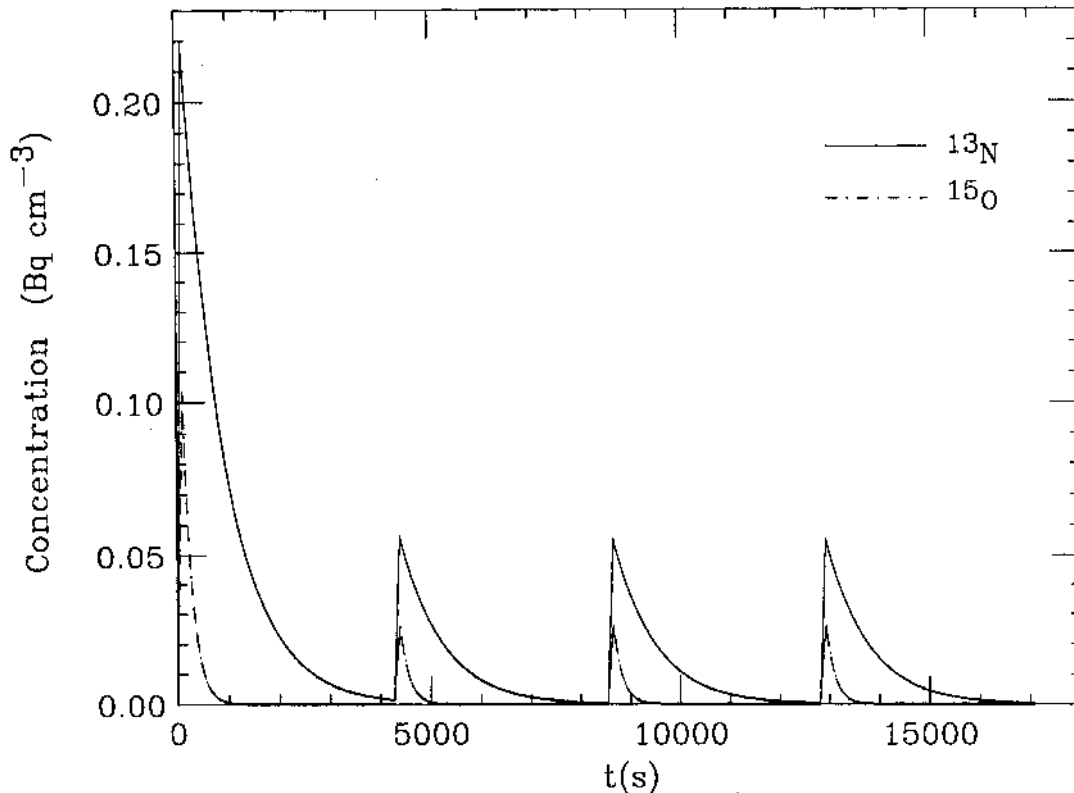


Figure 1: <sup>13</sup>N and <sup>15</sup>O concentrations inside the Linac hall as a function of time.

The gas concentration inside the Linac hall has been evaluated considering that in the normal operation the Linac will perform a first beam injection, lasting 120 s, which leads the current inside the Main Ring to a maximum value of about 5 A (complete injection). The subsequent injections, lasting 120 s, will be topping-up injections occurring everytime the current is reduced of 25% of its maximum value and this will happen about every 1.15 h in the normal working conditions.

The <sup>13</sup>N and <sup>15</sup>O concentrations into the Linac hall, where a ventilation system will perform 2.33 complete air changes per hour ( $0.6 \text{ m}^3 \text{ s}^{-1}$ ), are shown as a function of time in Fig. 1. The first peak corresponds to the end of a complete injection whereas the subsequent peaks correspond to the end of the topping-up injections. As shown in Fig. 1, during the normal operation of

Table 2: Released activities in one year from the Linac

Radionuclides	Released activity (Bq/y)
$^{13}\text{N}$	$1.79 \times 10^{11}$
$^{15}\text{O}$	$2.26 \times 10^{10}$
$^{41}\text{Ar}$	$4.83 \times 10^8$

Table 3: Doses in one year at different distances from the release point.

Distance (m)	Dose	
	to the whole body (Sv/y)	to the skin (Sv/y)
10	$2.6 \times 10^{-5}$	$4.0 \times 10^{-5}$
50	$2.7 \times 10^{-6}$	$4.2 \times 10^{-6}$
100	$9.1 \times 10^{-7}$	$1.4 \times 10^{-6}$
500	$4.8 \times 10^{-8}$	$7.4 \times 10^{-8}$
1000	$1.1 \times 10^{-8}$	$1.6 \times 10^{-8}$
5000	$7.9 \times 10^{-11}$	$1.2 \times 10^{-10}$

the Linac the maximum gas concentration is reached at the end of a complete injection. For the gases of interest the maximum gas concentrations reached during the normal operation of the Linac are reported in Table 1.

Both the  $^{13}\text{N}$  and  $^{15}\text{O}$  concentrations exceed the maximum permissible one, equal to  $7.4 \times 10^{-2}$  Bq/cm<sup>3</sup> (for an exposure of 40 hours per week)[IAEA79]. The time required for the  $^{13}\text{N}$  concentration to fall, after shutdown of the machine, from the maximum value to the permissible value is estimated to be about 11 minutes. In this interval of time the  $^{15}\text{O}$  concentration also drops below the maximum permissible concentration since its half-time is smaller than that of the  $^{13}\text{N}$ . In order to guarantee safety working conditions a waiting time of 15 minutes for the entry to the Linac hall seems reasonable.

The activity released in the environment has been evaluated considering the operation conditions foreseen in one year for the Linac, corresponding to 50 hours of complete injection per beam. The obtained results are shown in Table 2 where the released activities per year from the Linac are reported for the radionuclides of interest.

Italian legislation provides that the release of an amount of  $^{41}\text{Ar}$  activity over  $3.7 \times 10^8$  Bq per year must be authorized. But no indication exists for the other radionuclides here considered. According to European Directive, that will be soon issued and then incorporated into Italian legislation, the release of activity both for  $^{15}\text{O}$  and  $^{41}\text{Ar}$  over  $10^9$  Bq per year must be submitted to authorization. Anyway, the amounts of activity of the radionuclides that should be released from the DAΦNE installation have been authorized.

The evaluation of the radiation doses due to the released activity from the Linac has been made through the AQUILA code which estimates the activity

concentration at different distances and directions inside circular sectors centered on the release point. The meteorological conditions of Frascati site have been assumed equal to the ones of Ciampino site, 6 km far from Frascati, where a weather-station of the Air Force is operating in the airport. According to the plan, the expulsion will happen at a height between 4 and 10 meter from the ground. In the calculations an expulsion at a height of 4 m has been considered because the doses are the maximum ones in this condition. Moreover for each distance the maximum dose which results among the different directions has been cautiously considered. The dose values to the whole body and to the skin, estimated at various distances from the release point using the dose-rate conversion factor according to [Ko80, Pi85], are reported in Table 3. The doses obtained are below the dose annual limits recommended by ICRP [ICRP90] both for occupational exposure (20mSv/y for the effective dose and 500 mSv/y for the equivalent dose to the skin) and for public exposure (1 mSv/y for the effective dose and 50 mSv/y for the equivalent dose to the skin).

In the commissioning, the duration of an injection could be longer than one performed in the normal operation and then the values of gas concentrations would be higher than ones here presented. The gas concentration could achieve values corresponding to saturation conditions if the duration of the injection was at least two or three times the half-period. If the saturation conditions are reached with a complete injection, the gas concentrations will have the values reported in Table 1. In this case the time required for the  $^{13}\text{N}$  concentration to fall from the saturation value to the permissible value is estimated to be about 31 minutes. However, it should be mentioned that the commissioning will not be carried out under working conditions at full power of the Linac.

The dose received by an individual that entered into the Linac hall where saturation conditions were established with a complete injection, only 15 minutes after the shut-down of the machine, would be about of  $1.4 \times 10^{-2}$  mSv to the whole body and  $2.2 \times 10^{-2}$  mSv to the skin, according to the dose-rate conversion factors [Ko80, Pi85] for immersion in cloud containing the radionuclides of interest. Anyway, the entry of the machine staff members to the Linac hall, shortly after the shut-down of the machine, represents an occurrence that rarely will happen and no problem will arise from the safety point of view.

### Damping Ring hall

Electrons and positrons injected into the Damping Ring at 510 MeV will circulate for a time of about 100 ms before being transferred into one of the Main Rings. The radioactive gas production inside the Damping Ring hall will be mainly due to the beam power losses during the injections since the very short duration of the beam circulation into the machine makes the radioactive production in this phase negligible. The beam power losses have been evaluated on the basis of the running conditions used for shielding calculations[Es92].

The maximum of the radioactive concentration is reached at the end of a complete injection as it can be seen in Fig.3, where the  $^{13}\text{N}$  and  $^{15}\text{O}$  concentrations are shown as a function of time. The  $^{41}\text{Ar}$  concentration is not shown

because it is so low that can be disregarded. The maximum values of  $^{13}\text{N}$  and  $^{15}\text{O}$  concentrations, corresponding to the first peaks in Fig.2, is estimated to be  $2.93 \times 10^{-4} \text{ Bq/cm}^3$  for the  $^{13}\text{N}$  and  $1.4 \times 10^{-4} \text{ Bq/cm}^3$  for the  $^{15}\text{O}$ .

At the end of the topping-up injections, corresponding to the peaks following the first maximum peaks in Fig.2, the  $^{13}\text{N}$  concentration is estimated to be about  $7.5 \times 10^{-5} \text{ Bq/cm}^3$ , whereas the  $^{15}\text{O}$  concentration is about  $3.5 \times 10^{-5} \text{ Bq/cm}^3$ . No waiting time is necessary for the entry to the Damping Ring hall since the concentrations would be well below the maximum permissible ones.

The total released activity in one year from the Damping Ring, assuming one total air exchange per hour inside the hall, is estimated to be  $1.45 \times 10^8 \text{ Bq}$  for the  $^{13}\text{N}$  and  $1.68 \times 10^7 \text{ Bq}$  for the  $^{15}\text{O}$  in the same running conditions previously considered.

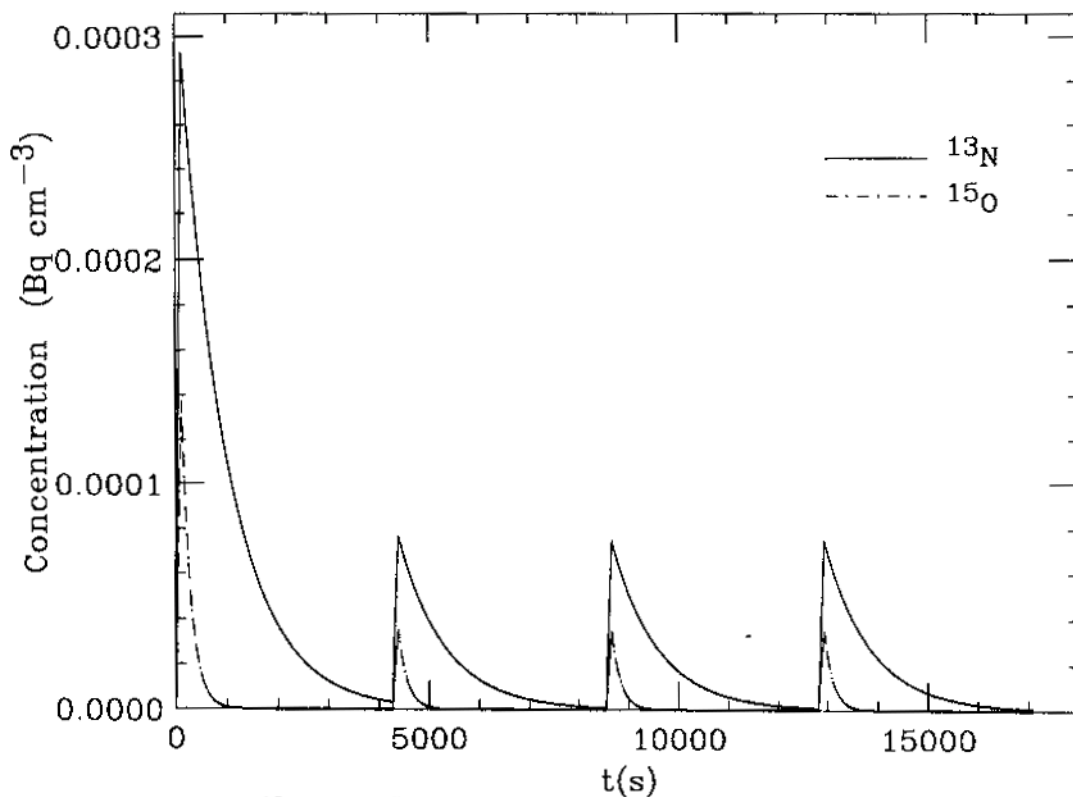


Figure 2:  $^{13}\text{N}$  and  $^{15}\text{O}$  concentrations inside the Damping Ring hall as a function of time.

The radiation doses due to the released activity from the Damping Ring result much lower than the ones due to the released activity from the Linac. Since these latter ones are quite low, the former ones can be disregarded from the safety point of view.

### Main Rings hall

During the normal operation of DAΦNE two electron/positron beams, with mean life estimated to be about 4 hours, will circulate in opposite directions

inside the Main Rings. The radioactive gas production has been evaluated considering both the beam losses during beam injection and the beam losses during the beam circulation according to the running conditions used for shielding calculations[Es92].

In Fig. 3 the  $^{13}\text{N}$  and  $^{15}\text{O}$  concentrations inside the Main Rings hall as a function of time, during a normal operation, are shown. The  $^{41}\text{Ar}$  concentration is not shown because it is so low that can be disregarded. The maximum concentrations of  $^{13}\text{N}$  and  $^{15}\text{O}$  are reached after the complete injection, corresponding to the first peak of each curve in Fig. 3, and they are equal to  $3.7 \times 10^{-5} \text{ Bq/cm}^3$  and  $1.7 \times 10^{-5} \text{ Bq/cm}^3$  respectively.

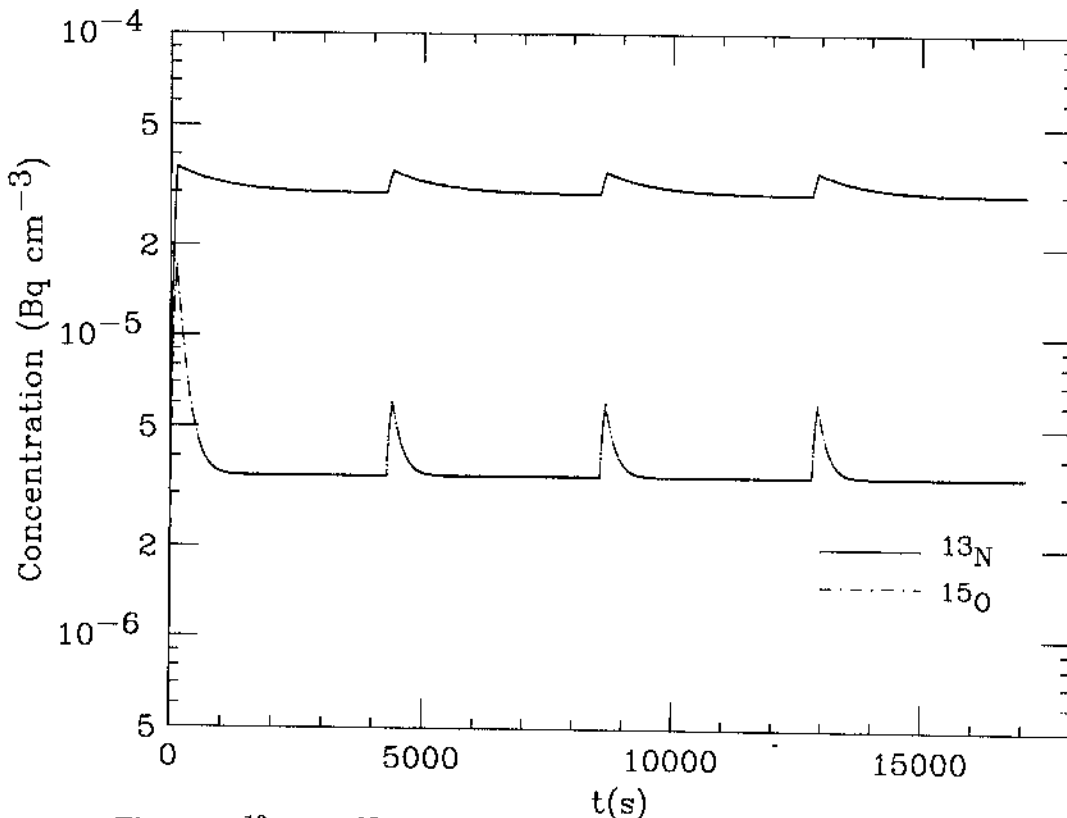


Figure 3:  $^{13}\text{N}$  and  $^{15}\text{O}$  concentrations inside the Main Rings hall as a function of time.

These values are below the maximum permissible concentration and no waiting time is required.

For the  $^{13}\text{N}$  the released activity in the topping-up injections ( $2.4 \times 10^4 \text{ Bq}$ ) would be greater than one in the complete injections ( $1.3 \times 10^4 \text{ Bq}$ ). On the contrary, for the  $^{15}\text{O}$  the major released activity would be in the complete injections ( $6.6 \times 10^3 \text{ Bq}$ ). The released activity per year, according to the operation conditions previously considered, is estimated to be  $4.6 \times 10^9 \text{ Bq}$  for the  $^{13}\text{N}$  and  $5.4 \times 10^8 \text{ Bq}$  for the  $^{15}\text{O}$ .

The radiation doses due to the released activity from the Main Rings will be negligible from the safety point of view for the same reasons explained for the Damping Ring.

## NOXIOUS GAS PRODUCTION

The radiation escaping in air causes the dissociation of the oxygen molecules leading to monoatomic oxygen. The attachment of monoatomic oxygen to the oxygen molecules forms ozone. Ozone can react with other compounds formed in air giving rise to the formation of other noxious gases such as nitrogen dioxide  $\text{NO}_2$  and nitric acid  $\text{HNO}_3$ . Among the noxious products, ozone is the most important one to be considered because of its high production rate and low threshold limit value, TVL=0.1 parts per million (ppm).

The estimation of the ozone concentration inside an hall has been carried out using the following equation which describes the change of the number  $N$  of ozone molecules per unit time:

$$\frac{dN(t)}{dt} = PG - \alpha N(t) - \frac{KP}{V} N(t) - \frac{kF}{V} N(t) \quad (4)$$

where  $P$  is the power deposited in air,  $G$  is the number of ozone molecules formed per eV (0.1 mol/eV) [IAEA79],  $K$  a constant related to destructive effect of radiation on ozone ( $5 \times 10^{-18}$  cm<sup>3</sup>/eV [We87]),  $\alpha$  is the decomposition constant ( $2.31 \times 10^{-4}$  s<sup>-1</sup>) [Mo91],  $k$  a factor which takes into account the imperfect mixing of the ozone molecules assumed equal to 1/3 [Mo91].

The ozone production is essentially due to bremsstrahlung since the beam will never travel in air, so that the power deposited in air can be expressed as [Mo91]:

$$P = \frac{Wxf}{\lambda_\gamma} \quad (5)$$

where  $W$  is the beam power loss,  $f$  is the fraction of electron beam energy which converts to bremsstrahlung radiation and enters the air,  $x$  is the air path and  $\lambda_\gamma$  is the attenuation length of photons in air. In the literature the  $f$  values can be found ranging from 0.1 to 0.5 [Fa84, CE87, Mo91], here it has been chosen the value 0.5 in order to consider the major production of ozone.

In the Linac hall the ozone concentration as a function of time, during the normal operation of the machine, is shown in the Fig. 4 where the first peak ( $1.43 \times 10^{-2}$  ppm) corresponds to the concentration at the end of the first complete injection. This value is lower than TVL and it is the maximum one reached during the normal operation.

The concentrations of  $\text{NO}_2$  and  $\text{HNO}_3$  can be approximately estimated on the basis of ozone concentration opportunely scaled by the G value ratios. By using the G value of 0.048 mol/eV for the  $\text{NO}_2$  and 0.015 mol/eV for the  $\text{HNO}_3$  [IAEA79, Fa84] the concentrations are of  $6.9 \times 10^{-3}$  ppm and  $2.2 \times 10^{-3}$  ppm respectively. These values are lower than the TVLs of  $\text{NO}_2$  and  $\text{HNO}_3$  which are 3 ppm and 2 ppm respectively.

Ozone concentration, during the beam tests, could be higher than one estimated in the normal Linac operation. In the case of a complete injection, lasting for a time sufficient to reach the saturation condition, the ozone concentration is estimated to be 0.274 ppm. This value exceeds the TVL and the time required for the  $\text{O}_3$  concentration to fall from the saturation value to the TVL results in 19 minutes.



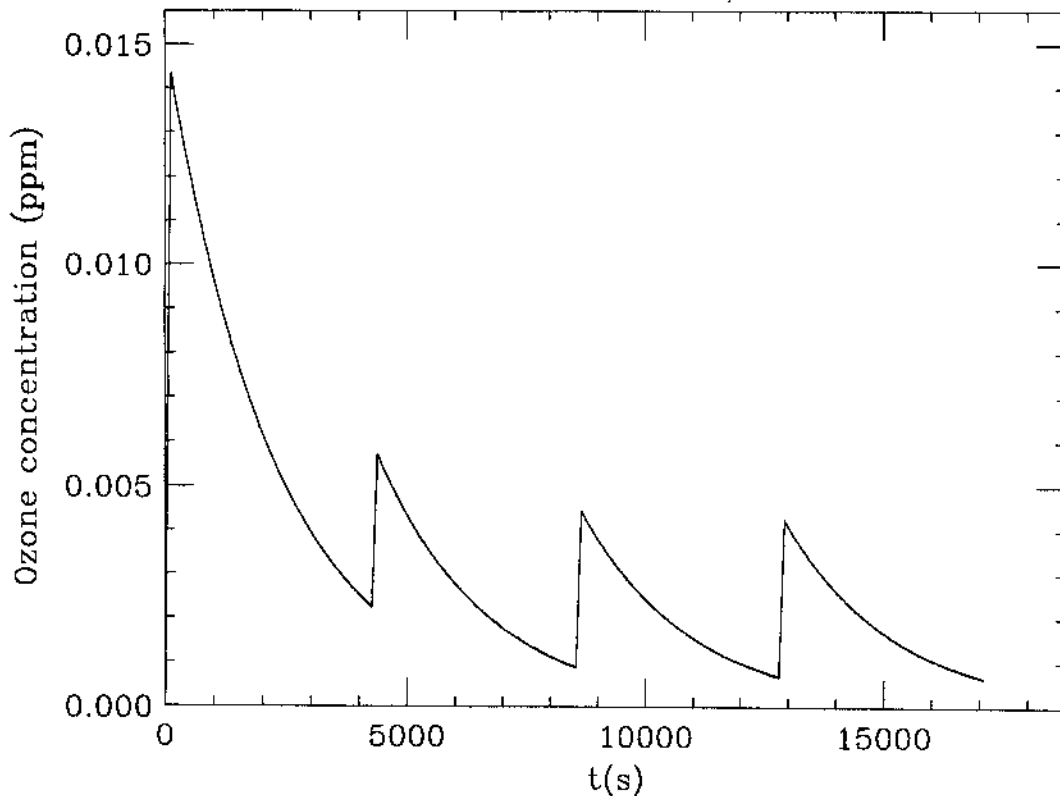


Figure 4: Ozone concentration as a function of time inside the Linac hall.

Anyway, if saturation conditions are reached there will be no hazard entering the Linac hall 15 minutes after the shutdown of the machine. The ozone concentration at this time (0.12 ppm) would be such that the effects of the ozone exposure on man would be at most symptomatic, that is irritation of the throat and tickling in the nose would be felt [Ho85].

The ozone production inside the Damping Ring hall and the Main Rings hall is due essentially to the bremsstrahlung since the synchrotron radiation is negligible. In fact, the Damping Ring and the Main Rings are operating at energies such that the major part of the synchrotron radiation is stopped into the wall of the vacuum chamber.

Both inside the Damping Ring hall and the Main Rings hall the maximum ozone concentration will be achieved after a complete injection and it is estimated to be  $2.0 \times 10^{-5}$  ppm and  $2.4 \times 10^{-6}$  ppm respectively. The ozone production does not pose safety problems during the normal operation of the Main Rings and the Damping Ring since the concentrations inside the halls will be well lower than the TVL.

The  $\text{NO}_2$  and  $\text{HNO}_3$  production inside the Main Rings hall and the Damping Ring hall can be disregarded from the safety point of view.

## References

- [CE87] Report of the CEBAF Radiation Control Review Panel, TN-0061, Newport News, Virginia 23606, April 16-18, 1987.
- [De86] F. Desiato, M. Pellegrino, Modello previsionale di dispersione in atmosfera di sostanze rilasciate da impianti convenzionali, ENEA 1986.
- [ENEA87a] ENEA-DISP, Valutazione della compatibilità ambientale della Centrale ENEL di Tavazzano-Montanaso, Doc. DISP(87)4,1987.
- [ENEA87b] ENEA-DISP, Valutazione della compatibilità ambientale della Centrale ENEL di Brindisi Sud, Doc. DISP(87)7,1987.
- [Es92] A. Esposito, M. Pelliccioni, DAΦNE Shielding, LNF-92/044(IR).
- [Fa84] A. Fasso, K. Goebel, M. Hoefert, G.Rau, H. Schonbacher, G.R. Stevenson, A.H. Sullivan, W.P. Swanson, J.W.N. Tuyn, Radiation Problems in the Design of the Large Electron-Positron Collider (LEP), CERN 84-02, March 5, 1984.
- [We87] C. Weilandics et al., Ozone Production at the National Synchrotron Light Source, BNL 39351, Brookhaven National Laboratory, January 1987.
- [Ho85] M. Horvath, L. Bilitzky, J. Huttner, Ozone, Monograph 20, Topics in Inorganic and General Chemistry, Edited by R.J.H. Clark, Elsevier, 1985.
- [IAEA 79] W.P.Swanson, Radiological safety aspects of the operation of electron linear accelerators, IAEA, Technical Report Series, No.188(1979).
- [ICRP90] Recommendations of the International Commission on Radiological Protection. Publication 60 (Oxford: Pergamon Press) (1990).
- [Ko80] D.C. Kocher, Dose-rate conversion factors for external exposure to photon and electron radiation from the radionuclides occurring in routine releases from nuclear fuel cycle facilities, Health Physics, Vol.38, 543(1980).
- [Mo91] H.J. Moe, Advanced Photon Source, Argonne National Laboratory, July 1991.
- [Pa73] H.W. Patterson and R.H. Thomas, Accelerator Health Physics, Academic Press (1973).
- [Pi85] H.V. Piltingsrud and G.L. Gels, An evaluation of the external radiation exposures dosimetry and calculation of maximum permissible concentration values for airborne materials containing  $^{18}\text{F}$ ,  $^{15}\text{O}$ ,  $^{13}\text{N}$ ,  $^{11}\text{C}$  and  $^{133}\text{Xe}$ , Health Physics, Vol. 49, No.5, 805(1985).