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Coordinatore Prof. Vincenzo Guidi

Radiation Protection Issues for Cyclotron Produced Radionuclides

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Dottorando	Tutore
Lucia Sarchiapone	Prof. Mauro Gambaccini

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1. Introduction

Research in the field of nuclear physics has always moved on two courses, to all appearances distinct: what is called *basic research* on one side and *applied physics* on the other side. Progress on one of the two paths has always gone along with progress on the other, just to confirm that the distinction exists only at first glance. There are many examples of how theoretical studies can be translated into technologies and applications within everyone's means, providing undeniable improvement in living standards.

In this context progress in the field of healthcare and, more precisely, in nuclear medicine finds its place. Diagnostic and therapeutic techniques of common use nowadays – radiography, tomography, hadrontherapy, brachytherapy, just to cite a few of them – are built on the achievements of the study of the interaction between fundamental particles and matter, and on the investigation of the possible exploitation of radiation.

This scientific bipolarity has been recognized by the NuPECC, the Nuclear Physics European Collaboration Commettee, an expert committee of the European science foundation, that in 2014 acknowledged the remarkable impact of nuclear physics on society through its applications. The outcome of this statement is well expressed in a report entitled "Nuclear Physics for Medicine" [1].

What is important to underline is that nuclear physics laboratories, whose field of research deals with the development of detectors and with innovation technologies related to particles' accelerator, deeply contribute to the advancement of nuclear medicine. This contribution is anyway always indirect, thanks to technology transfer and to the application of science research findings, but very often it is direct, following the intentional choice of research institutions to commit their resources and beam-time to medical application, whether they are aimed at therapy, diagnosis (or the combination of the two, i.e. *theranostics*, word used referring to those treatments able to combine therapy and diagnosis in one treatment, taking the advantages of both and reducing the exposure to the patient) or radiopharmaceuticals.

On an international landscape, many examples of such laboratories can be found. TRIUMF, the Canada's national laboratory for particle and nuclear physics and accelerator-based science, where the non-reactor based production of isotopes tried to find a solution to the shortage of ^{99m}Tc [2]. CERN, the European Organization for Nuclear Research in Switzerland, where forefront research in particles accelerators has

provided the optimal substrate for a project purposely oriented to radiopharmaceuticals production, MEDICIS [3]. iThemba LABS, the South African laboratory for Accelerator Based Science, where the subatomic physics research, radiotherapy and production of radionuclides already share the same technology with the existing cyclotron [4].
At the national level, the National Institute of Nuclear Physics (INFN) fosters the engagement of the researchers and of the infrastructures of its laboratories in the field of nuclear medicine. The national laboratory of South (LNS) hosts the center of handrontherapy and advanced nuclear applications (CATANA) where the treatment of the shallow tumors is feasible with a proton beam delivered by a superconducting cyclotron [5].
The national laboratory of Legnaro (LNL) is going to create a research infrastructure, LARAMED, in the field of radiopharmaceuticals, thanks to the proton beam delivered by the recently installed cyclotron. The goals of LARAMED will be the research on innovative radionuclides to be used in tumors diagnosis and therapy and the consolidation of accelerator-based techniques for the production of existing substances.
All of the mentioned laboratory have a twofold mission: basic research and medical application. Being accelerator-based facilities, anyway, they share the same issues concerning radiological safety and, consequently, they also share the engineering solutions set up to tackle the risks associated to ionizing radiation

1.1 MOTIVATION

In this work, the LARAMED project has been taken as an example to outline the radiation protection issues of a laboratory dedicated to radiopharmaceuticals production, starting from the irradiation of a target with high power proton beams, to the analysis and treatment of the radioactive target in a radiochemical equipped laboratory. Radiation protection includes the identification of the radiation sources and the range of all the actions taken to guarantee the health protection of the workers and the general public. The treated matter is obviously shared with many applications, other than the irradiation of a target for medical radioisotopes, but the increasing interest in the use of cyclotrons of energy about 70 MeV for this task, is the rationale for the thesis.
The present work has been organized according to the following schematics:

- Chapter 1: a short, far-from-being-exhaustive introduction on the fundamental concepts of radiation protection, interaction mechanisms particle-matter, radioactivity and dosimetry (attention will be focused on the physical quantities used to evaluate the amount of exposure)
- Chapter 2: the methods used to perform the analysis will be shown. A short description of the Monte Carlo method, and why it is so widely used for this type of evaluations, is given. Some available codes for particular applications

will be shown. Some applications where Monte Carlo method and analytical investigation have been coupled to obtain reasonable results, are explained.

- Chapter 3: description of the LARAMED project, the laboratory framework and the cyclotron.
- Chapter 4: calculations and numerical simulation results. All the calculations with detailed reference to the source terms and the adopted approximations are shown in this chapter. The normal working conditions and the possible accidents will be reported, together with the illustration of possible consequences and safety solutions adopted.
- Chapter 5: Legal framework ruling the activities with a risk of exposure to ionizing radiation in Italy is shortly illustrated. Some important aspects concerning the licensing procedures will be explained.
- Chapter 6: measurements done during the commissioning of the cyclotron are shown, trying, when possible, to benchmark them with the calculations. A description of the instrumentation is also given.

2. Basic Concepts

The goal of this chapter is to give an introduction on the physical basis of the radiation protection. It will be, clearly, non-exhaustive for *all* the theories important for radiation protection evaluations, but it will explain the basic concepts on which the calculations have been based.
It will be recalled the mechanisms of interactions of particles with matter, the radioactive decay mechanism and some dosimetric concepts.

2.1 Radiation Fields

Radiation is the term widely and commonly used [6] to explain physical phenomena involving energy transfer from one place to a different one. Light coming from a lamp, radio-waves from an electrical circuit, X-rays from radiologic equipment, are few examples of energy transferred through space without displacement of any medium.
The general classification of radiation is according two categories: ionizing and non-ionizing. It is called ionizing that radiation capable of producing, directly or indirectly, ionization of atoms and molecules of crossed matter. If the energy of the radiation is high enough to remove the orbital electron from the atom during the interaction, then the atom is said to be ionized (otherwise it is only excited).
Ionizing radiation always transfers energy to any material with which it interacts. The energy it deposits in living tissues causes disruption of the atomic structure, and when the atoms thus affected are essential for the normal functioning a cell can be permanently damaged or killed. When ionizing radiation imparts energy to living tissue, damage is done: the larger the amount of energy deposited, the more extensive is the damage.
Being the harmful effect of radiation perceptible only through its consequences, it is of maximum importance to evaluate - through calculations and analysis - the entity of the energy imparted to the region of interest, and to monitor through suitable instrumentation, the emission from radiation sources.

2.1.1 Direct and Indirect ionization

In general, charged particles are capable of producing ionization directly, while uncharged particles can induce ionization through secondary mechanisms (indirectly ionizing radiation).

In this paragraph, a short description of the main mechanisms of interaction of particles with matter are presented, referring to the bibliography for a more complete explanation of the phenomena.
An electrically charged particle, with sufficient energy to release an electron from its orbit, undergoes an inelastic collision with an orbital electron, transferring to it energy at least equal to that of the binding energy of that electron. This transferred energy enables the release of the electron from its orbit. The path of light charged particles is not straight due to the continuous change of direction due to the interaction with almost same size target particles (and substantial energy transfer). The path of heavy charged particles, instead, is relatively straight, being the deflection negligible.
Photons and neutrons are the most commonly encountered indirect ionizing particles by radiation workers. Photons interact with matter with three most common interaction mechanisms: photoelectric effect, Compton effect and pair production. Which type of interaction a photon will undergo is determined by its energy and the material traversed.
In general, photons of lower energy (less than 100 keV) are more likely to interact through the photoelectric effect; photons of intermediate energy (above 100 keV), through the Compton effect. Pair production is not possible until photon energies of 1.02 MeV or greater are reached. The specific energy range of each interaction is also determined by the properties of the material though which the photon is travelling. For example, pair production becomes dominant in lead for photon energies in the range of 4 to 5 MeV, whereas, for tissue, photon energies of 20 MeV must be reached.
Neutron are produced primarily in two ways: as an end product in a nuclear interaction, or with fission. They are generally divided in two or more groups, as function of their energy: fast (neutron energy above 0.5 eV, where 0.5 eV is the so-called *Cadmium cut-off* below which the neutron is readily absorbed by Cadmium), slow (between 0.0025 eV and 0.5 eV) and thermal (below 0.0025 eV). Neutrons interact with the atomic nucleus, not with the orbital electrons, in a variety of ways.
Elastic scattering occurs when the neutron transfers part of its energy to a nucleus without leaving it in an excited state. For the energy and momentum conservation laws, the smaller the nucleus the greater the amount of energy transferred by the. Based on this principle low Z light materials find very wide application as moderators. The most commonly used are hydrogen-rich materials such as water, paraffin, polyethylene.
When the atomic nucleus is left in an excited state, an inelastic scattering has occurred. The nucleus might de-excite emitting a gamma-ray photon. This photon production has to be taken into account, for instance, when designing neutron shielding. When the neutron energy is above about 10 MeV, it is possible for a second neutron to be radiated by the nucleus so that inelastic scattering is often a reaction (n, nγ) type. If the de-excitation does not occur, the nucleus remains in a metastable state (typically this state lasts fractions of a second).
If no secondary neutrons are emitted by the nucleus, the reaction is known as neutron capture. In general, the probability for a capture to occur increases as the neutron energy decreases (it is called the *1/v* region of the material excitation function, where *v* is the

neutron velocity). Often when neutrons are captured, or absorbed, by the nucleus, charged particles are ejected instead of, or in addition to, gamma rays.
Fission takes place with heavy atomic nuclei. The neutron is absorbed and the resulting compound nucleus splits into two fission fragments and one or more fast neutrons.
Finally, the capture by a nucleus of neutron of energy of 100 MeV or higher may cause the emission, known as shower, of many different types of particles. [7]

2.2 RADIOACTIVITY

The most common source of alpha and beta particles and of gamma-ray photons is the natural or induced transformation or decay of the nucleus of an atom. Every chemical element has associated with it an atom that has a fixed number of protons in its nucleus and the same number of electrons orbiting around the nucleus. However, the number of neutrons present in the in the nucleus may vary (and often does). These are called isotopes of the chemical element, have the same chemical properties of the original element and they can be either stable or unstable, depending on the energetic status of the nucleus. When a nucleus is unstable it tries to rearrange in order to get a lower energetic status and to do this it emits a particle or a gamma ray. This nuclei transformation is called radioactive decay and its timescale ranges from fractions of a second to thousands of years. Each isotope has its own characteristic timescale, called mean lifetime and defined as the time requested to reduce the initial amount of radioactive material to a fraction *1/e* of it. The radioactivity of a certain nuclide always depends on the initially present amount of that isotope and the commonly used equation describing the evolution in time of the radioactivity is

$$A(t) = N\varphi\sigma \cdot (1 - e^{-\lambda t}) \tag{1}$$

where N is the number of target nuclei, φ is the flux of the projectile inducing the target transmutation and σ is the reaction cross section. The analytical explanation of the radioactivity evolution can be found in many reference books and will not be repeated here [6], [8].
In the following paragraph, an excursus on the radioactivity induced in air in presence of a ventilation system will be done, in order to understand the influence of the ventilation rate in the radioactivity evolution.

2.2.1 Induced Radioactivity in Air

The interaction of the beam protons with the target induces a secondary radiation field mainly composed of neutrons, photons and protons. Secondary particles interact with the air constituents and generate radioactive species.
The risks coming from the production of radioactive species in air are linked to the worker exposure during access after the accelerator shutdown, or to the eventual effluent dose to general public following radioactive air expulsion through the stack by the ventilation

system. An overview of the reactions and the generated radionuclides is given in Table 1.

Table 1 Production reactions for the radionuclides in air.

Reaction	T1/2	Threshold (MeV)	Cross section
^{16}O (n, p) ^{16}N	7.5 s	10	40 mb
^{16}O (n, 2n) ^{15}O	2 min	18	20 mb
^{14}N (n, p) ^{14}C	5730 years	0.5	0.10 b
^{14}N (n, p) ^{14}C	5730 years	Thermal capt.	1.81 b
^{14}N (n, t) ^{12}C	12 years	4.3	20 mb
^{14}N (n, 2n) ^{13}N	10 min	11.3	10 mb
^{40}Ar (n, p) ^{40}Cl	1,4 min	6.9	10 mb
^{40}Ar (n, np) ^{39}Cl	55 min	10.2	1 mb
^{40}Ar (n, d) ^{39}Cl	55 min	12.4	1 mb
^{40}Ar (n, α) ^{37}S	5.0 min	2.6	1 mb
^{40}Ar (n, γ) ^{41}Ar	1.83 h	Thermal	0.5 b

In order to take the ventilation into account in the radioactivity evaluation, some considerations have to be done. Supposing that the air changes inside the vault with a rate of R (air volume changes per second), one can express the following equilibrium condition:

$$\lambda n + Rn = N\Phi\sigma \tag{2}$$

where the radionuclides production is balanced by the losses through radioactive decay and removal by the ventilation system (n is the number of radioactive nuclides). Being $A=\lambda n$ the decay rate (radioactivity), the equation (2) can be expressed as:

$$A + \frac{R}{\lambda}A = N\Phi\sigma \Rightarrow A = N\Phi\sigma\frac{\lambda}{\lambda + R} \tag{3}$$

so that the released radioactivity per second is:

$$RA = R \cdot N\Phi\sigma \cdot \frac{\lambda}{\lambda + R} \tag{4}$$

In absence of a ventilation system radioactivity is defined as in equation (1) and for long irradiation periods radioactivity tends to a saturation value $A_S = N\Phi\sigma$, numerically equal to the production rate but expressed in Bequerel units.
For a better understanding of the saturation activity variation with the introduction of ventilation we can refer to figure 1: from eq. (2) one can see that saturation activity with

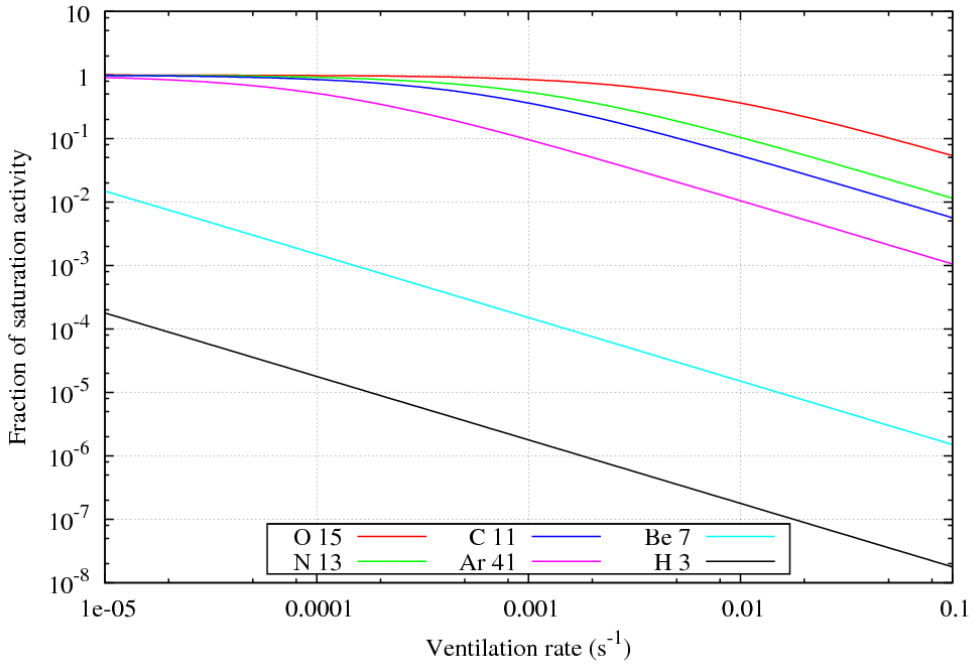


Figure 1 Radioactivity concentration as a fraction of the saturation activity without ventilation, in an enclosed volume, as a function of the ventilation rate.

ventilation is equal to the production rate $N\Phi\sigma$ scaled by the radionuclide decay constant divided by the effective decay constant $\lambda_{effective} = \lambda + R$ taking into account the ventilation rate. This quantity is the equilibrium activity:

$$A_{eq} = N\Phi\sigma\frac{\lambda}{\lambda + R}$$

So $\frac{\lambda}{\lambda + R}$ is the fraction of the equilibrium activity to the saturation activity in absence of ventilation (with no ventilation equilibrium activity and saturation activity coincide).

As the air change rate R increases, the amount of radioactivity inside the closed vault decreases. This is much more evident for short half-life nuclides.
In those areas where ventilation is not needed, for instance because the contamination levels are not important, it has been calculated the time needed to allow radioactivity to decay after the accelerator shutdown.

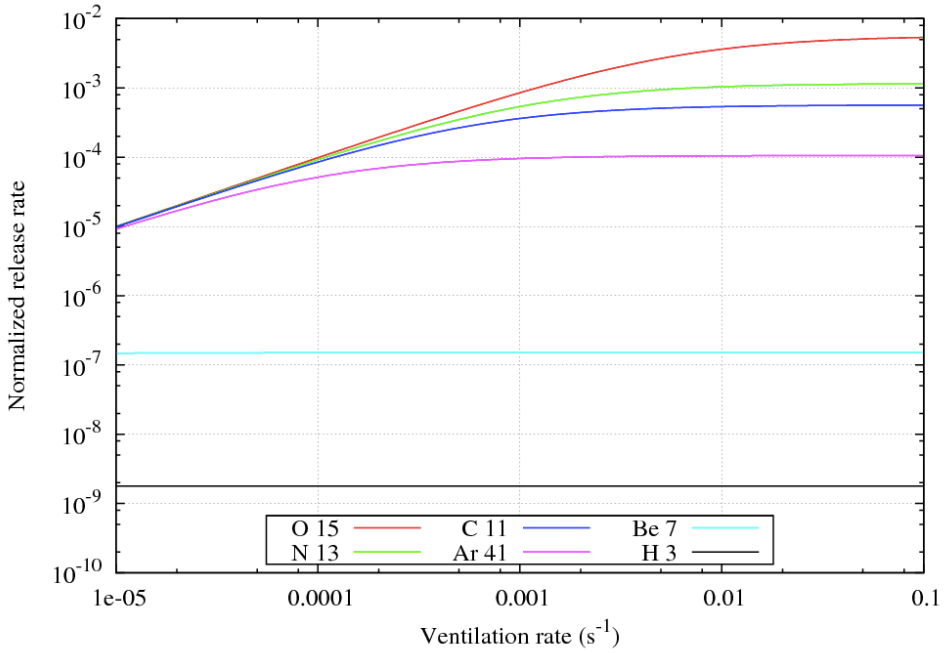


Figure 2 Radioactivity concentration as a fraction of the saturation activity without ventilation, released as a function of the ventilation rate.

Figure 2 shows how the radioactivity released at the stack changes as a function of the ventilation rate. The factor $R\lambda/\lambda + R$ is the fraction of the released radioactivity to the saturation activity in absence of ventilation (with no ventilation the released radioactivity is clearly null).

As the ventilation rate increases the released radioactivity increases as well.

In Table 2 some values concerning the quantities described have been explicitely reported, in reference to two ventilation rates: 3.5 air changes/hour (about 10^{-3} air changes per second) and 50 m^3/hour (about 5 10^{-5} air changes per second). The first one is a typical reference value for plants, while the second is an example of low rate value to be considered if the ventilation system is only required to balance the air losses due to the non-hermetic closure of the room.

What explained so far, is valid when the equilibrium between production and removal terms is already reached in the enclosed volume. What happens before the equilibrium is reached? How much conservative is the hypothesis of having, for each radionuclide, the saturation activity? The following is just to demonstrate that the hypothesis is very close to the real working conditions.

Table 2 Fraction of the saturation activity in an enclosed volume and at the extraction with two reference ventilation rates.

Nuclide	3,5 air changes/h		50 m^3/h	
	Enclosed volume	Extraction	Enclosed volume	Extraction
^{3}H	$1{,}8\ 10^{-6}$	$1{,}8\ 10^{-9}$	$3{,}8\ 10^{-5}$	$1{,}8\ 10^{-9}$
^{7}Be	$1{,}5\ 10^{-4}$	$1{,}5\ 10^{-7}$	$3{,}2\ 10^{-3}$	$1{,}5\ 10^{-7}$
^{11}C	$3{,}7\ 10^{-1}$	$3{,}6\ 10^{-4}$	$9{,}2\ 10^{-1}$	$4{,}3\ 10^{-5}$
^{13}N	$5{,}4\ 10^{-1}$	$5{,}3\ 10^{-4}$	$9{,}6\ 10^{-1}$	$4{,}4\ 10^{-5}$
^{15}O	$8{,}5\ 10^{-1}$	$8{,}3\ 10^{-4}$	$9{,}9\ 10^{-1}$	$4{,}6\ 10^{-5}$
^{41}Ar	$9{,}8\ 10^{-2}$	$9{,}5\ 10^{-5}$	$6{,}9\ 10^{-1}$	$3{,}2\ 10^{-5}$

The variation of radioactive nuclides depends on: production (P), radioactive decays ($-\lambda N$) and removal by the ventilation system ($-RN$):

$$dN = P\,dt\ -\ \lambda N\,dt\ -\ RN\,dt \tag{5}$$

$$\frac{dN}{dt} = P - \lambda N - RN \tag{6}$$

The solution to this first order differential equation is the function

$$N(t) = \frac{P}{\lambda+R}(1 - e^{-(\lambda+R)t}) \tag{7}$$

being the radioactivity defined as $A(t) = \lambda N(t)$ one can obtain:

$$A(t) = \frac{P\lambda}{\lambda+R}(1 - e^{-(\lambda+R)t}) \tag{8}$$

where the production term is, as usual, $P = N_0 \Phi \sigma$.
The graphical representation of the activity, as expressed in equation (8), normalized by the production term P in order to be more general, is the schematic in Figure 3 and the released fraction is represented in Figure 4.
The ventilation rate for this example is 3.5 air changes per hour. A time period lightly greater than 15 minutes the radioactivity of the analyzed species has reached the equilibrium value, that is the hypothetical value used for the calculations.

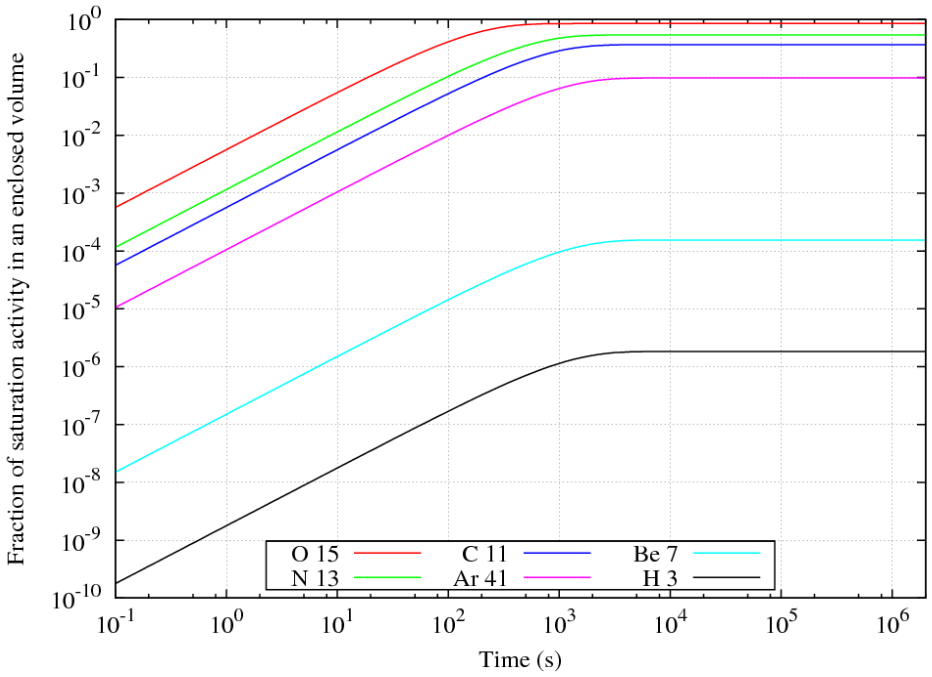


Figure 3 Radioactivity concentration evolution in time before reaching saturation in an enclosed volume.

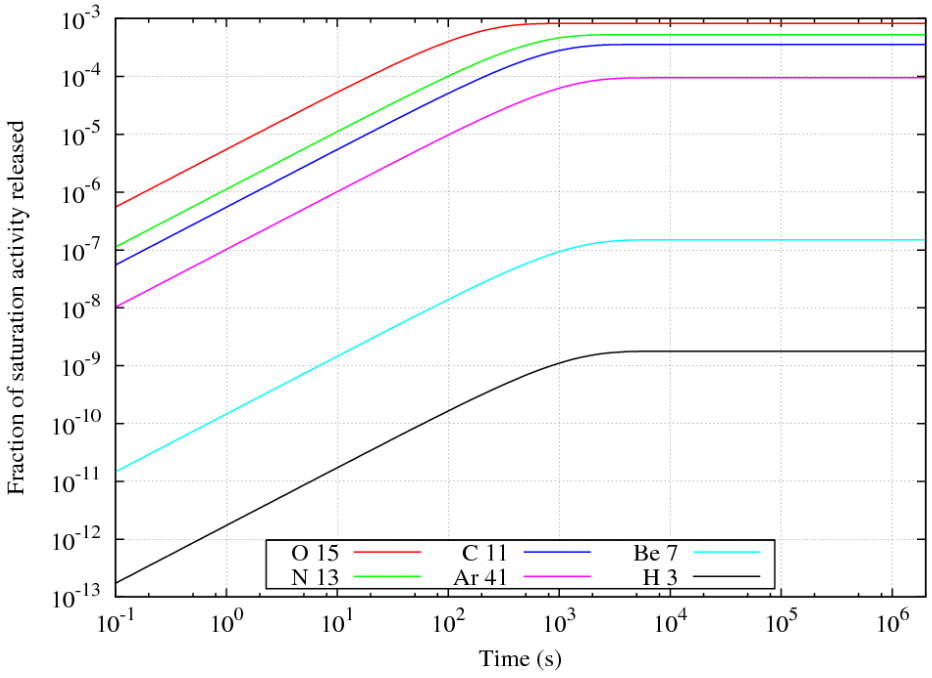


Figure 4 Radioactivity concentration evolution in time, before reaching saturation, released from an enclosed volume.

2.3 RADIATION PROTECTION PRINCIPLES

The quantities used in radiation protection are derived from physical quantities and weighted by the appropriate coefficients in order – on one side - to take into account the biological effects of ionizing radiation and – on the other side - to be able to quantify the exposure of the human body to ionizing radiation from both whole and partial body external irradiation and from intakes of radionuclides [10, 11]. In this sense, the protection quantities fully describe the harmful effect of radiation on the human body as a whole and for this reason they are used to define legal protection limits. The only drawback is that they are not directly measurable, thus the operational quantities have been introduced: they provide conservative estimates of the protection quantities and are often used to demonstrate compliance with dose limits. Radiation protection detectors for individual and area monitoring are calibrated in terms of operational quantities.
Definitions of the mentioned quantities are given in the following paragraphs.

2.3.1 Physical quantities

Fluence, Φ (unit: $1/m^2$) is the quotient of *dN* by *da*, where *dN* is the number of particles incident upon a small sphere of cross-sectional area *da*

$$\Phi = dN/da$$

In dosimetric calculations, fluence is frequently expressed in terms of the lengths of the particle trajectories. It can be shown that the fluence, Φ, is given by

$$\Phi = dl/dV$$

Where *dl* is the sum of the particle trajectory lengths in the volume *dV*. This is the reason why in the FLUKA code, detectors used to computationally evaluate the particles fluence are called *tracklength estimators.*
Absorbed dose, *D* (unit: gray, 1 Gy = 1 J/kg =100 rad) is the energy imparted by ionizing radiation in a volume element of a specified material divided by the mass of this volume element.
Kerma, *K* (unit: Gy) is the sum of the initial kinetic energies of all charged particles liberated by indirectly ionizing radiation in a volume element of the specified material divided by the mass of this volume element.
Linear Energy Transfer, *L* or *LET* (unit: J/m, often given in keV/μm) is the mean energy *dE*, lost by a charge particle owing to collisions with electrons in traversing a distance *dl* in matter. *Low-LET radiation*: X-rays and gamma-rays (accompanied by charged particles due to interactions with the surrounding medium) or light charged particles such as electrons that produce sparse ionizing events far apart at a molecular scale ($L < 10$ keV/ μm). *High LET radiation*: neutrons and heavy charged particles that produce ionizing events densely spaced at a molecular scale ($L > 10$ keV/ μm).

Activity, A (unit: bequerel, 1 Bq = 1 atom disintegration/s = 27 picoCurie) is the expectation value of the number of nuclear decays occurring in a given quantity of material per unit time.

2.3.2 Protection quantities

Organ Absorbed dose, D_T (unit: Gy) in an organ or tissue T of mass m_T is defined as the dose on average absorbed by the organ of interest

$$D_T = \frac{1}{m_T} \int_{m_T} D \, dm$$

Equivalent dose, H_T (unit: Sievert, 1 Sv=100 rem) in an organ or tissue T is equal to the sum of the absorbed doses $D_{T,R}$ in the organ or tissue caused by different radiation types R weighted with so-called radiation weighting factors w_R:

$$H_T = \sum_R w_R \times D_{T,R}$$

it expresses long term risks (primarily cancer and leukemia) from low-level chronic exposure. The values for w_R recommended by the International Commission on Radiological Protection (ICRP) [12] are unity for photons, electrons and muons, 2.0 for protons and charged pions, 20.0 for ions and a function of energy for neutrons

$E_n < 1$ MeV :
$2.5 + 18.2 \times \exp [-(\ln E_n)^2 / 6]$

$1 \text{ MeV} \leq E_n \leq 50$ MeV :
$5.0 + 17.0 \times \exp [-(\ln (2E_n))^2 / 6]$

$E_n > 50$ MeV :
$2.5 + 3.25 \times \exp [-(\ln (0.04E_n))^2 / 6]$

Effective dose, E (unit: Sv) is the sum of the equivalent doses, weighted by the tissue weighting factors w_T ($\sum_T w_T = 1$), of several organs and tissues T of the body that are considered to be most sensitive

$$E = \sum_T w_T \times H_T$$

2.3.3 Operational quantities

Ambient dose equivalent, $H^*(10)$ (unit: Sv) is the dose equivalent at a point in a radiation field that would be produced by the corresponding expanded and aligned filed in a 30 cm diameter sphere of unit density tissue at a depth of 10 mm on the radius vector opposing the direction of the aligned field. Ambient dose equivalent is the operational quantity for *area monitoring.*

Personal dose equivalent*,* $H_p(d)$ (unit: Sv) is the dose equivalent in standard tissue at an appropriate depth, d, below a specified point on the human body. The specified point is normally taken to be where the individual dosimeter is worn. For the assessment of the effective dose, $H_p(10)$ with a depth d = 10 mm is chosen, and for the assessment of the dose to the skin and to the hands and feet the personal dose equivalent, $H_p(0.07)$, with a depth d = 0.07 mm, is used. Personal dose equivalent is the operational quantity for *individual monitoring*.

2.3.4 Dose conversion coefficients

Dose conversion coefficients allow direct calculation of protection or operational quantities from particle fluence and are functions of particle type, energy and irradiation configuration. The most common coefficients are those for effective dose and ambient dose equivalent. The former are based on simulations in which the dose to organs of anthropomorphic phantoms is calculated for approximate actual conditions of exposure, such as irradiation of the front of the body (antero-posterior irradiation) or isotropic irradiation.

In Monte Carlo simulations, such coefficients allow multiplication with fluence at scoring time such that effective dose to a human body at the considered location is directly obtained. [10, 13].

3. Calculation Methods and Numerical Codes

In order to evaluate the radiological risk of a practice, the following schematic items should be covered:

- description of the physical process on which the practice is based: for the applications here described, the physical process is a particle/radiation transport problem and a short description of the methods to solve these problems is given below;
- identification of the path through which the individual or the environment is subject to potential exposure. For workers, it could be external irradiation due to the permanence in high dose rate environments or introduction of radioactivity through inhalation (for instance breathing radioactive air coming from an irradiation bunker), ingestion or through the skin (especially handling unsealed radioactive sources). For general public, it could be external irradiation due to submersion in a radioactive plume in case of an accidental release of radioactivity, or introduction
- evaluation of the exposure through those paths.

In general, problems dealing with the transport of particles and radiation through matter may be treated with deterministic or statistical methods. *Deterministic* are those methods based on the solution of the Boltzmann equation of transport. Often those are iterative algorithms where the number of iterations and the resolution of the computing grid strongly affect the accuracy of the solution [14].
Statistical methods, on the other hand, solve the problem through a stochastic convergence: increasing the number of histories brings a numerical solution closer to the exact solution.

3.1 Monte Carlo method

For radiation transport, the statistical approach known as Monte Carlo method, is recognized to be the most accurate and efficient choice. It is based on the use of variables randomly sampled from a known distribution, according to the following schematics, shared by all the Monte Carlo methods:

1. Mathematical formulation of the problem, in order to derive an equation to describe the behavior of the quantity of interest.
2. Formulation of the statistical interpretation of the problem, where the quantity of interest is expressed as a parameter of a distribution, for example the mean.
3. An algorithm for sampling the distribution is developed.
4. Estimators for the parameter and its statistical uncertainty are derived. These can be as simple as the sample average and the sample variance.
5. The algorithm and the estimators are optimized to reduce the computing time needed to achieve the desired level of statistical uncertainties. Methods for achieving this goal without introducing systematic error, or a bias, are referred to broadly as variance reduction methods.
6. A sample is generated, that is sufficiently large to achieve the desired level of statistical uncertainty of the estimate of the parameter.
7. The parameter and its uncertainty are estimated using the sample

Nowadays Monte Carlo codes have been developed in order to describe with unprecedented accuracy most physical processes. They can implement nuclear data from the most updated libraries and take advantage of the wide benchmarks of the last decades with experimental data. Moreover, many features are continuously improved by the developers and - together with the user communities' enlargement – also the request for new and challenging performances contributes to the growth of these techniques. Some of those features are the capability to easily describe in great detail complex geometries and even, in many cases, import them from commonly used CAD tools; the possibility to simulate in one and the same run the whole process concerning, as in the present application, the interaction of a proton beam with a target, the secondary interactions of generated particles and the calculation of radioactive decays (even with a runtime modified geometry).

For the present study, all the evaluations have been made using the Monte Carlo code FLUKA [15, 16], developed by INFN and CERN researchers purposely for radiation protection applications and hadron calorimeters. It is a general-purpose interaction and transport code, born in the 60s and continuously improved (the history of FLUKA is usually divided in three FLUKA generations). In the last generation of the code A. Ferrari (INFN) and A. Fassò set up a plan to transform FLUKA from a high-energy code, mostly devoted to radiation shielding and beam heating, into a code which could handle most particles of practical interest and their interactions over the widest possible energy range [17].

An accurate description of the code, of the physical models implemented therein and of the feasibilities can be found in literature or directly on the website. In the following paragraphs, it will be given a short description of the code features used in the calculations.

3.1.1 Scoring

Results of a Monte Carlo numerical simulation are obtained as the average of the occurrences of a quantity of interest, selected by the user. All the radiometric quantities can be scored either using the built-in estimators, equivalent to the measurements done during an experiment, or writing an own routine to be compiled to the main executable.
By running several independent calculations, it is possible to obtain the standard deviation.
Many options exist as built-in estimators, the great part of them relying on the geometrical description of the implemented system: there are region-dependent estimators (a certain quantity, as energy deposition for example, is averaged on an area with homogeneous characteristics, defined as region), geometry-independent estimators (a spatial mesh including different regions or parts of them superimposed on the geometry, the scored quantities are averaged on each volume of the mesh, called bin) that are often used to create colored maps of quantities distributions. Differential quantities, as for example the fluence of a particle as a function of its energy or angle (with respect to a specified reference system), can be scored inside one region or at the boundary between two confining regions.
There is a dedicated estimator for the evaluation of radionuclides production in a certain material (region dependent), extremely useful with problems related to induced activity and dose rate from activated components.

3.1.2 Radioactive decay

In order to handle the radioactive decay, it must be included in the simulation a beam-time irradiation profile and a defined interval for decay. The dedicated options in FLUKA allow to define not only the duration of the irradiation (as in the study case, where a target material is continuously irradiated with a proton beam) but also the intensity of the primary beam, in order to estimate correctly the induced radioactivity load as function of the proton current.
By the definition of decay intervals (time after the end of the irradiation) and the association of the appropriate estimators to these intervals it is possible to *take instantaneous pictures* after an irradiation, in order to follow the decay of the induced radioactive elements and to obtain the related dosimetric information at selected instants in time. One of the advantages of FLUKA with respect to other Monte Carlo codes is the capability of doing this in one and the same simulation run.
A very useful feature, recently introduced in the code, is the possibility to change the geometry runtime when a radioactive decay calculations is required. This capability has been used widely in the present study, whenever the removal of an irradiated object was foreseen and the residual dose rate coming from other sources needed to be evaluated.

3.1.3 Biasing techniques

When run in fully analogue mode, FLUKA allows the user to study fluctuations and correlations, and to set up a direct simulation of physical reality where all moments of phase space distributions are faithfully reproduced. On the other hand, in the many applications where only quantities averaged over many events are of interest, it is convenient to use calculation techniques converging to the correct expectation values but reducing the variance (or the CPU time, or both) by sampling from biased distributions. This is especially useful in deep penetration calculations (like thick shielding effectiveness), or when the results of interest are driven by rare physical interactions or cover a small domain of phase space.
The easiest biasing technique, and the most commonly applied in the shielding calculations presented here, is the importance biasing: it consists in assigning an importance value to each geometry region. The number of particles moving from a region to another will increase (by splitting) or decrease (via a technique called Russian Roulette) according to the ratio of *importances*, and the particle statistical weight will be modified inversely so that the total weight will remain unchanged. In this way, the user can strive to keep the particle population constant, making up for attenuation, or to make it decrease in regions far from the detectors where there is a lower probability to contribute to the score. In FLUKA, importance biasing can be done separately for hadrons/muons, electrons/positrons/photons and low-energy neutrons.
Not a biasing technique strictly speaking, is the use of black holes around areas of interest. In FLUKA blackhole is a material with infinite absorption cross section and zero density, conveniently introduced to avoid infinite transport of escaping particles through space. Sometimes is has been used to cut out sections of the facility to focus the radiation transport on a well-defined space, thus speeding up the simulation.

3.1.4 Radiation quantites

As expressed in paragraph 2.3, the quantities used to set the dose limits for the workers and the population are the most suitable to take into account the harmful capabilities of the different radiations and the radiosensitivity of the different organs. Nevertheless, these quantities are neither directly measurable nor directly inferred from calculations. In order to have them readily available running a FLUKA simulation, conversion coefficient have been calculated so that the physical quantities evaluated in the course of the simulation are translated into operational or protection quantities, in a very similar fashion to the instruments calibration procedures, [13]. The conversion coefficients for effective dose have been calculated taking into account various space-oriented irradiation. Ambient dose equivalent coefficients have been evaluated through the introduction of the ICRU sphere, as specified in [18, 19].

In the present study, unless otherwise specified, all the calculations concerning dose and dose rates have been made scoring the quantity ambient dose equivalent and ambient dose equivalent rate[1].

3.1.5 Two-step approach

Sometimes the use of a Monte Carlo simulation, even with the application of biasing techniques, might not be accurate and fast enough. For instance, in those applications where the interaction probability is low because of the low density of the target material, the convergence could not be reached in reasonable time. It is the case, for instance, of the evaluation of airborne activity in an enclosed volume where an irradiation occurs. The secondary radiation, mainly constituted of neutrons, generated during the target irradiation, causes the generation of radioactive isotopes of the air components. The evaluation of the induced radioactivity can be done in one run or in two-step. The two-step consists in: 1) the evaluation of secondary particle fluence as function of energy (spectrum) in the air volume of interest, 2) the folding of the particle spectrum with the cross section for the production of the main radioactive nuclides in air, [20, 21].
In the present work, the two-step approach has been followed for the calculations concerning airborne activation.

3.2 HOTSPOT

In order to evaluate the consequences of a possible accident, it has been used the HotSpot code, developed at the Lawrence Livermore National Laboratory to equip emergency response personnel and planners with a set of software tools for evaluating incidents involving radioactive material. The software is also used for safety-analysis of facilities handling radioactive material. [22]
The code includes atmospheric dispersion models like explosion, fire, resuspension or general release where the characteristics must be specified by the user. These models estimate the short range (less than 10 km), downwind radiological impact following the release of radioactive material resulting from a short-term release (less than a few hours), explosive release, fuel fire or an area contamination event.
The estimated radiation dose provided by the code is done following the radiation dosimetry methodologies recommended by the ICRP, including biokinetic and dosimetric models for estimation of dose equivalent following acute inhalation of radionuclides. [23-26].

[1] When the term *dose* is used throughout this work it is always meant ambient dose equivalent.

4. The LARAMED project: the cyclotron, the building and related projects

In this chapter it will be given a general description of the LARAMED project, starting from the cyclotron of the LNL, the projects arising around it (nuclear physics and possible applications) and the building layout.

4.1 THE SPES PROJECT

The overall frame of this work is the SPES project of the LNL [27]. The acronym SPES stands for Selective Production of Exotic Species. The project is aimed at the production of re-accelerated radioactive ion beams for fundamental physics research as well as at interdisciplinary applications, ranging from the production of radionuclides of medical interest to the generation of neutrons for material studies, nuclear technologies and medicine. All of these applications share the starting point, a proton beam of energy in the range 40 – 70 MeV delivered by the 70p Best Cyclotron [28].
Following the progress of the project and in an attempt to explain the SPES logo of Figure 5, we can say that the project is developed as a four-stage program:

α. The acquisition of the cyclotron by the laboratory, its commissioning and the production/re-acceleration of radioactive ion beams generated through the interaction of the proton beam with a non-fissile target
β. The generation and re-acceleration of radioactive ion beams from the interaction of the proton beam with a fissile target, uranium carbide
γ. The irradiation of target for the production of radionuclides of medical interest and related radiopharmaceuticals' production. This is the LARAMED project.
δ. Development of an intense neutron source, from the cyclotron and/or from a high intensity linear accelerator based on radio-frequency quadrupole (RFQ) technology. Applications of the neutron source range from nuclear astrophysics to test of electronics in space, characterization of nuclear waste or experimental tumor treatments.

Figure 5 The SPES logo.

4.2 LARAMED

LARAMED (acronym for LAboratory for the production of RAdionuclides for MEDicine), is a new interdisciplinary project promoted by the INFN in collaboration with other institutions, including the National research Council (CNR) and various Universities. The project has been substantially funded by the Italian Ministry of Education, University and Research (MIUR) in 2012 and 2016 within the scheme of Premium Programs.
A similar facility was proposed in [29], when – for the first time – it was thought about a beamline of the new incoming cyclotron, completely dedicated to radioisotopes for medical applications, and its feasibility was investigated.
Scope of the LARAMED project is to explore novel and highly efficient nuclear reactions for producing innovative radionuclides having nuclear properties potentially useful for clinical applications in diagnostic and therapeutic medicine. Specifically, the project is aimed at extensively investigating nuclear reactions induced by high-energy accelerated proton beams colliding onto solid targets. Development of automated procedures for the extraction and purification of radionuclides from irradiated targets is also another important topic covered by the experimental activities of the LARAMED project.
Essentially, the production chain of medical radionuclides involves the following three main steps: (1) target production and irradiation, (2) target chemical processing and (3) purification of the final radionuclide. The LARAMED framework is designed to cover all these different phases that will be accomplished inside dedicated infrastructures and laboratories. Typically, a preliminary evaluation for cross sections of nuclear reactions relevant to the production of the radionuclide of interest will commonly precede the subsequent attempt to irradiate a suitable target. Production of small amounts of radioactivity of the desired nuclide is another key step useful for disclosing the most appropriate chemical treatment necessary to separate and purify the final radionuclides to achieve the required quality standards. Finally, after establishing the most effective experimental setup, high-yield, radionuclide production is conveniently pursued to assess whether the selected nuclear process will be able to sustain a massive production of the

radioisotope commonly demanded for the preparation of radiolabeled diagnostic and therapeutic agents to be employed in routine clinical applications.
The entire chain, running from the basic experimental research on radionuclide production to the final clinical application on the patient, also includes other activities such as manufacturing of new radiopharmaceuticals following good manufacturing practice (GMP) and their preclinical and clinical evaluation will not be discussed in this work.

4.3 THE CYCLOTRON

The proton driver for the target irradiation within the LARAMED project and for the proton-fission induced in uranium carbide as requested by the SPES project, is a compact cyclotron built by the Best company, from Canada.
It has been delivered to LNL in 2015. The installation and commissioning phase has been terminated in June 2017.
The cyclotron has an external negative hydrogen ion source, four radial sectors with two separated dees in opposite valleys, cryogenic vacuum system and simultaneous beam extraction on opposite lines. The beam intensity is 700 microamperes with variable extraction energy between 35 and 70 MeV, [30, 31].
At present, the simultaneous beam delivery can only occur with the same beam energy extracted on both ports, but with a future upgrade two beams of different energies might be obtained.

4.4 THE FACILITY LAYOUT

The layout of the SPES facility is presented in Figure 6, with indication of the areas interested by the LARAMED project including the cyclotron.

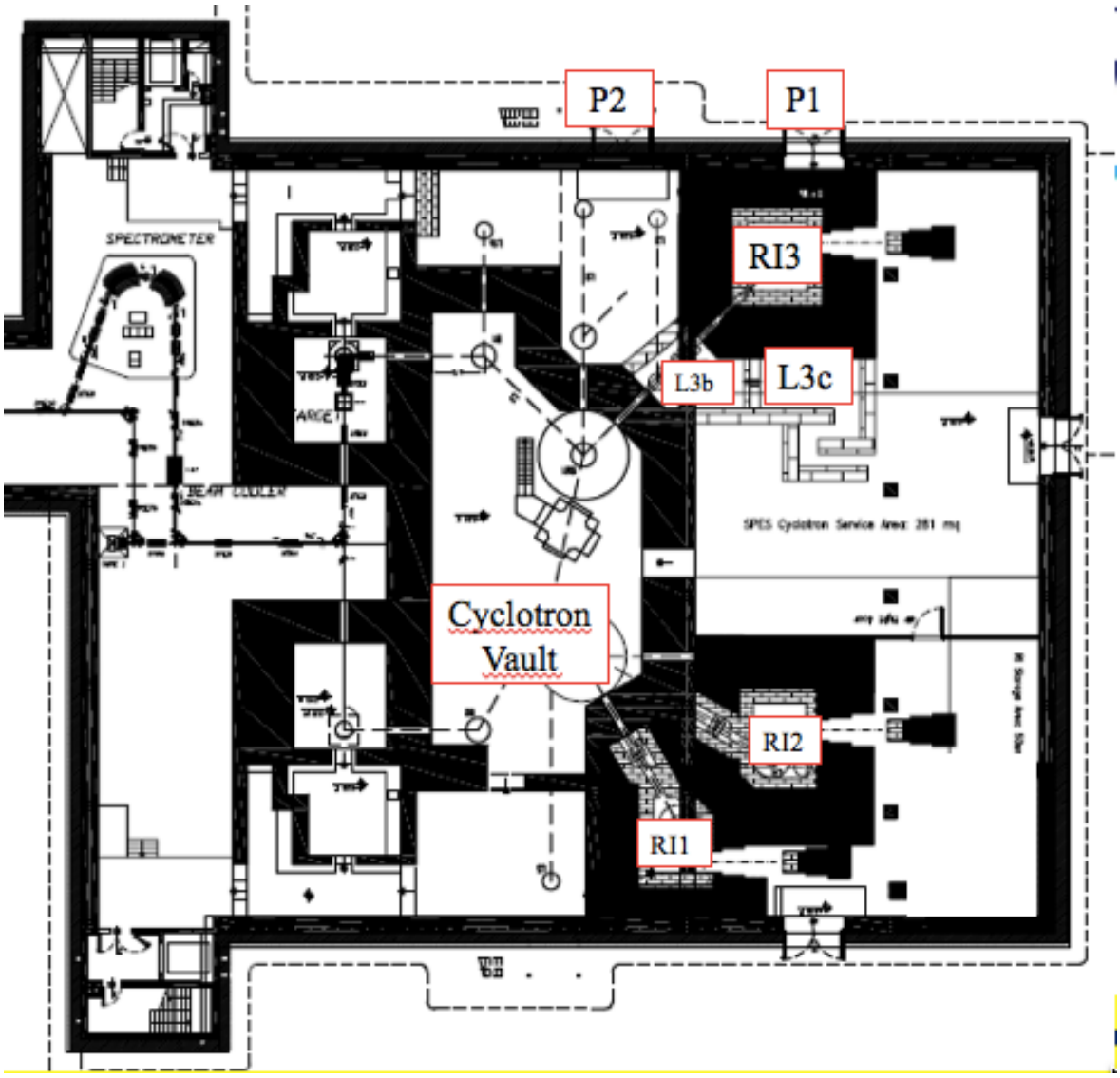


Figure 6 The SPES facility layout. The cyclotron vault and the irradiation bunkers of the LARAMED project are labelled.

5. Calculations and results

5.1 Source Term

The source term in the irradiation bunkers RI#1-2-3 has been evaluated taking into account the activities to be done in each area. In particular, they might be:

- Cross section studies
- Irradiation of a target for experimental tests on new radioisotopes (usually not requiring a long irradiation time)
- Irradiation of a target for massive production of a certain radioisotope (irradiation lasting for a period of 1 week, at last)

5.1.1 Cross section measurements

Usually cross section measurements need a dedicated and well characterized beam line in order to reduce the source of errors in the reaction parameters reconstruction. Moreover, they do not require high current beams, being the radioactivity induced in the target limited by the dead time of the detector.
As a general rule, one can assume that

a) the radioactivity A_S induced in a thin target with a proton beam of I_p (μA) can be obtained from the following:

$$A_S \approx 1.5\ 10^9\ I_p \qquad \text{Bq per g cm}^{-2}$$

b) the ambient equivalent dose rate R_{amb} is:

$$R_{amb} \approx 3\ 10^{-4}\ I_p \qquad \text{Sv m}^2\ \text{h}^{-1}\ \text{per g cm}^{-2}$$

The cross section study is done with thin foils enriched with the isotope suitable for the reaction of interest. Usually the foils' thickness is about 0.2 g/cm^2, so - considering that the induced radioactivity should be lower than approximately 10^7 Bq – the maximum proton current is 30 nA, 1.875 10^{11} protons per second (applying the rule expressed at the previous point a).

With the described parameters the dose rate[2] R_{amb} at 1 meter distance is about 0.2 μSv/h. Considering the limited amount of induced radioactivity and the low dose rate, it is clear that the irradiation of thin targets for cross section measurements is not a concern for the radiation protection of personnel. But it is, indeed, the interaction of the uncollided proton beam with the intercepting objects following the foils. Dedicated beam dumps are used to stop the beam completely, so that, in the context of this work, independently from the elemental composition of the thin target, the main source of radiation is due to the interaction of the proton beam with the beam dump.
For the studies on the production of radiopharmaceuticals, the required proton current depends first on the need of having a sufficient amount of radioactivity of the isotope of interest, second on the technological solutions adopted in the target realization, mainly the target cooling in order to efficiently absorb the power on the target.

5.1.2 Production target

The actual interest on radiopharmaceuticals covers a wide list of substances. The most original are those used for theranostics (^{64}Cu, ^{67}Cu) obtained from the irradiation of metallic Ni or Zn (either metallic or as oxide). Some of them are shown in Table 3 with the reaction for the production using proton beams and the required energy.

Table 3 Some possible nuclides of interest and the reactions for their generation (non-exhaustive list), [29] and [32]

Nuclide	$T_{1/2}$	Reaction	Required proton energy	σ_{max} (mbarn)
^{64}Cu	12,7 h	^{nat}Ni (p,n) ^{64}Cu	40	50
		^{64}Ni (p,n) ^{64}Cu	15	675
^{67}Cu	61,8 h	^{68}Zn (p,2p) ^{67}Cu	70 – 20	25
^{82}Sr/^{82}Rb	25,5 d	^{85}Rb (p,4n) ^{82}Sr	70 - 40	100
^{68}Ge/^{68}Ga	270,8 d	^{69}Ga (p,2p) ^{68}Ge	45	100
		^{69}Ga (p,2p) ^{68}Ge	20	550
^{123}Cs/^{123}I	13,2 h	^{124}Xe (p,2n) ^{123}Cs	44 – 16	700
^{124}I	4.17 d	^{nat}Te(p,n)^{124}I	53	150
		^{124}Te(p,n)^{124}I	12	590
^{186}Re	90 h	W(p,n)^{186}Re	10	17
^{103}Pd	17 d	^{103}Rh(p,n)^{103}Pd	10	500
^{228}Th	1.9 y	^{232}Th(p,X)^{228}Th	70	60
^{225}Ac	10 d	^{232}Th(p,X)^{225}Ac	60	3

[2] Where not otherwise specified, with *dose rate* it is meant *ambient equivalent dose rate*.

^{230}Pa	17.4 d	^{232}Th(p,3n)^{230}Pa	30	260

For few of them the proton energy obtainable from the cyclotron is required (40-70 MeV) while for the others, it might be foreseen the used of an energy degrader.
The neutron ambient equivalent dose rate as a function of the distance from the target is shown in Figure 7 for the targets listed above.

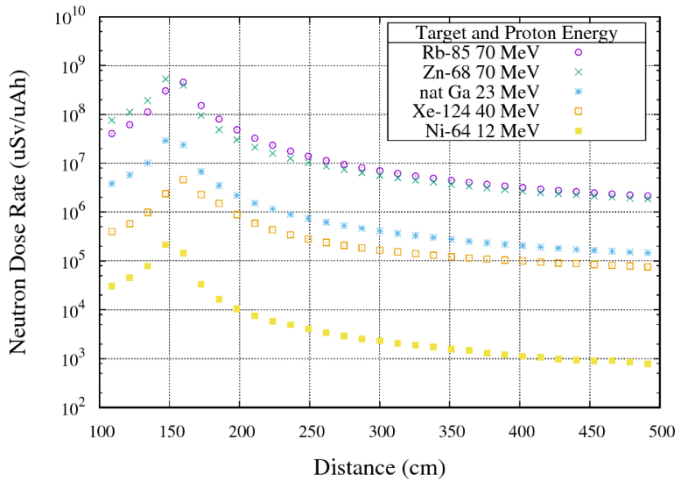


Figure 7 Neutron dose rate (μSv/h μA^{-1}) as a function of the distance from the irradiation target. The position of the target on the plot is 150 cm on the x-axis, the target's material and the proton energy are shown in the plot legend.

The reaction for the production ^{82}Sr for the ^{82}Sr/^{82}Rb generator, requiring proton energy of 40-70 MeV turns out to be the most worrisome from a radiation protection point of view. As indicated in the table, the reaction through which Rb (naturally composed of the only non-radioactive isotope ^{85}Rb (72.2%) and ^{87}Rb (27.8%), is converted into ^{82}Sr is very neutron-prolific.
^{85}Rb(p,4n)^{82}Sr is the reaction of specific interest for medical applications, but also ^{85}Rb(p,3n)^{83}Sr and ^{85}Rb(p,5n)^{81}Sr will occur, the cross sections are reported in Figure 8Figure 9 andFigure 10.
For this reason, all the calculations concerning shielding design, radioactivity build up and dose rates (prompt and residual) are based on the use of rubidium as target, among the different materials that can be anyway used in the installation within the same scope.

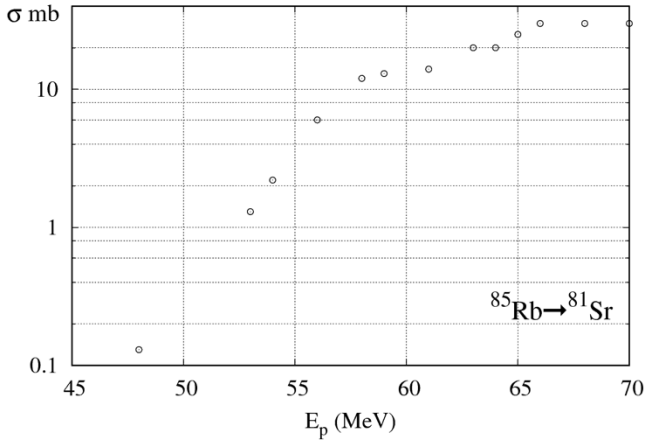


Figure 8 Excitation function for the reaction ^{85}Rb (p, 5n) ^{81}Sr. [32]

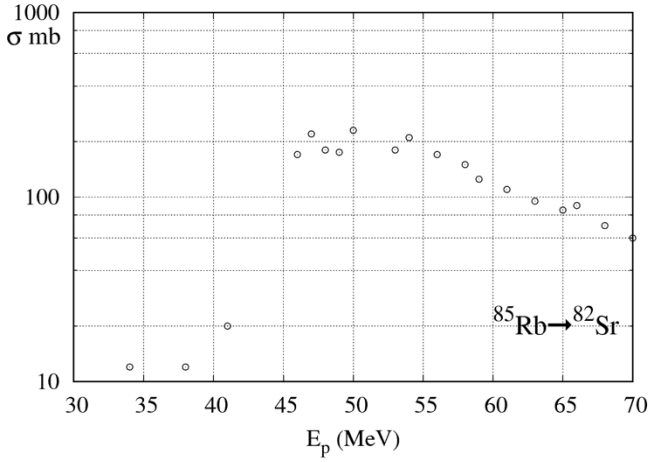


Figure 9 Excitation function for the reaction ^{85}Rb (p, 4n) ^{82}Sr [32]

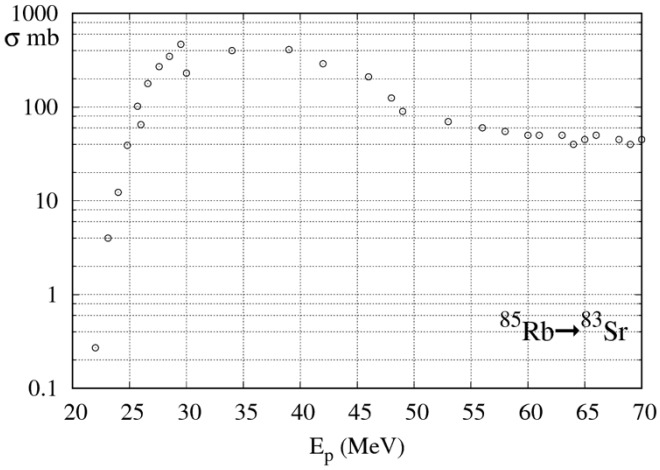


Figure 10 Excitation function for the reaction ^{85}Rb (p, 3n) ^{83}Sr. [32]

The fluence of neutrons emerging from the irradiation of a thick RbCl target with 70 MeV energy protons is represented in Figure 11, as a function of the neutron energy. The curves represent the energetic spectrum at different angles with respect to the direction of the incoming proton beam.
The energetic spectrum of the neutrons shows the evaporation peak at approximately 2 MeV. At the energy of 10 MeV and beyond the distribution is characterized by neutrons coming from direct collisions, mainly at 0° with respect to the proton beam direction. At larger angles the high energy peak decreases, to slowly disappear at 90°. As a general indication, there shouldn't be any high-energy neutron at angles greater than 90° but the multiple scattering events degrade and back-reflect some high neutron originally directed forward. The evaporation peak, that should be isotropic for thin targets, shows a slight difference in the angular distribution mainly due to the attenuation in the thick target used for this application.
Neutron emission as a function of the initial proton energy is also represented in Table 4.

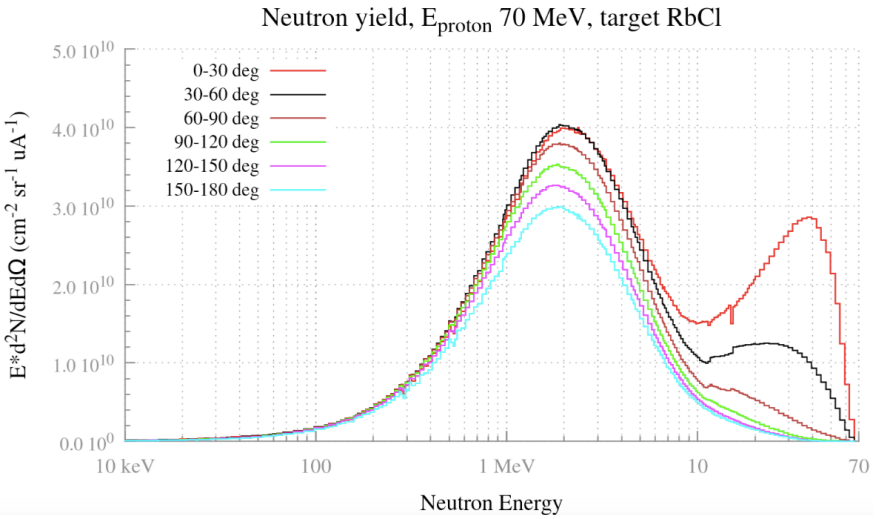


Figure 11 Energetic and angular distribution of the neutron emerging during the irradiation of a rubidium chloride target with 70 MeV protons. The intensity has been scaled to 1 μA proton current.

Table 4 Neutron yield as a function of the proton energy impinging on the RbCl target.

Energia (MeV)	Neutroni per protone su target
35	0,023
50	0,053
70	0,107

5.2 SHIELDING DESIGN

Based on the irradiation conditions - therefore on the source terms explained in the previous paragraph – the shielding must be optimized in order to avoid any overexposure of the personnel working in the same building (or in the laboratory in general) and of the public outside the facility. Suitable shielding should also provide adequate isolation from the environment, thus preventing spread of contamination during both normal operation and accidental situations.
The shielding of the irradiation bunkers in the SPES building is made of reinforced concrete, with density of 2.4 g/cm^3 (in the simulations the density has been kept 2.3 g/cm^3 in order to be slightly conservative and to take possible inhomogeneity into account).
It will be shown in detail the shielding calculations for the bunker dedicated to the radioisotope production using high intensity proton beam. An overview of the bunker for cross section measurements will also be given.

5.2.1 The high intensity bunker

It will be shown the evaluation of the shielding in the bunker RI3, taking into account the radiation protection constraints and the workload. The presence of non-exposed workers is foreseen at a distance of approximately 15 meters, where the door P1 is located.
In Figure 12,Figure 13 and Figure 14 the schematics of the bunker with the shielding walls thickness is shown. On the perimeter it has been foreseen 350 cm thickness, for the roof 450 cm and for the floor it is 300 cm. The same structure is adopted for all of the 3 bunkers, the one dedicated to experimental activities and those for the massive production.

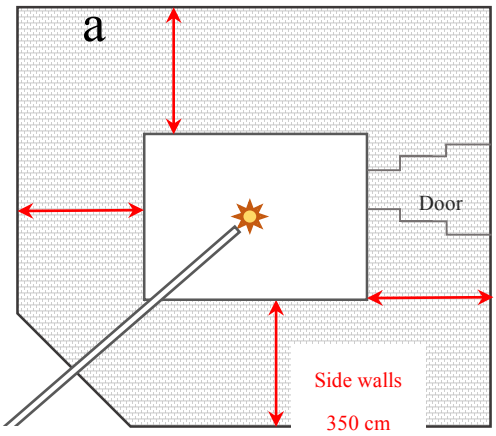


Figure 12 Irradiation bunker RI#3, plan view. The thickness of 350 cm for the perimeter shielding walls is shown. The star points at the position of the target inside the vault. The beam direction can also be deduced from the beam line shown. Letter a indicates the side of the bunker adjacent to the external building wall (100 cm additional shielding on that side).

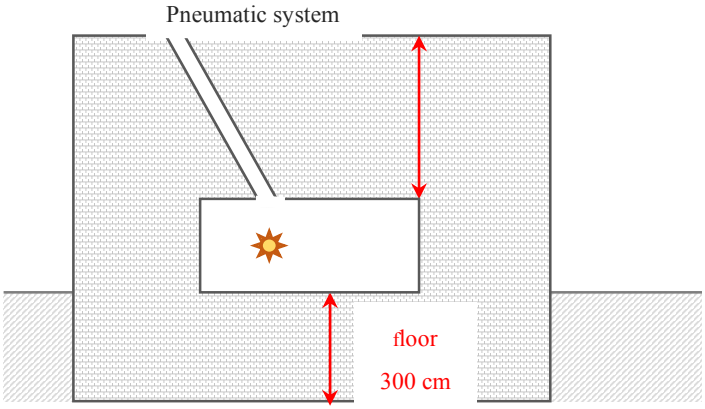


Figure 13 Front view of the irradiation bunker. The thickness of the roof (450 cm) and of the floor (300 cm) is highlighted. On the roof, besides the openings for technical services not shown in the sketch (water, air, cables) it has been drawn the penetration for the pneumatic system to transfer the target to the radiochemical labs at the 2nd floor.

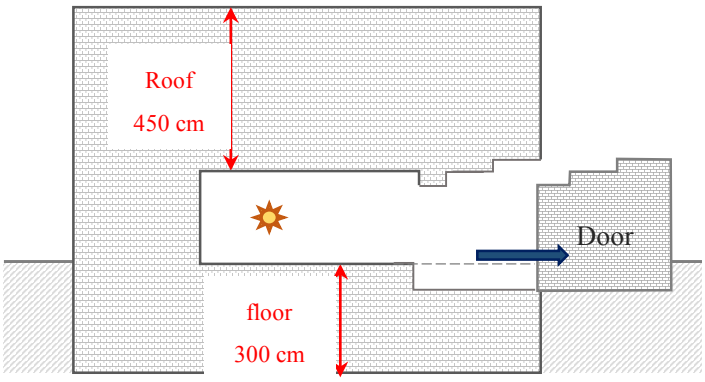


Figure 14 Front view with indication of the door movement. The steps (3 on top and 1 at the bottom) have been designed to prevent neutrons to travel along a straight path.

The attenuation of the neutron ambient equivalent dose rate is shown in Figure 15. It has been shown the transmission through concrete at 45° wrt the beam direction. (-45° is the direction parallel to the door moving axis while +45° is the direction perpendicular to the external wall, where the shielding is increased by 1 metere because of the external structure of the building). The source term for this calculation is the irradiation of a RbCl target with protons of 70 MeV energy and 1 μA current.

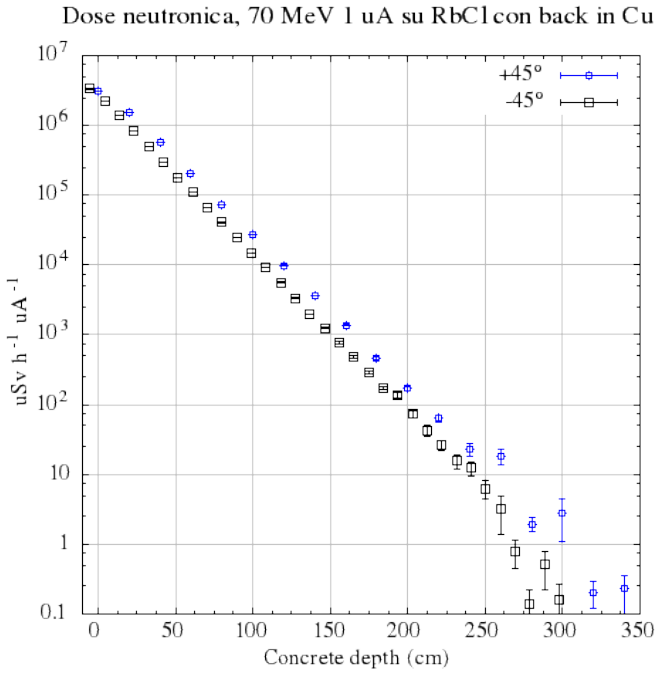


Figure 15 Neutron ambient equivalent dose rate through concrete during the irradiation of a RbCl target with protons of 70 MeV energy. Results are scaled for 1 μA proton current. The target is assumed to be 3 meters from the wall.

For personnel protection, the most critical point is the wall hosting the door. In that direction one can find the door to the outside of the building where transit of non-exposed workers is allowed. Moreover, at few meters distance there is the perimeter of the LNL, so this is the distance taken as reference for general public reference group.
The dose rate at 350 cm depth is 0.15 μSv/h/μA, so scaling to the workload considered as worst-case, that is a proton current of 500 μA, the dose rate out of the shielding door of the RI3 bunker is about 80 μSv/h. Considering that the door P1 is 15 meters from the shielding external door, and applying the inverse square law for dose attenuation it results a dose rate of 0.35 μSv/h outside. The area outside of the facility is a service road for the SPES building, thus an occupancy factor of 1/16 can be taken into account.
The workload is assumed to be in the order of 100 hours per year so that the dose outside of the facility is approximately 35 μSv per year, much lower than the non-exposed workers dose limit of 1 mSv/year. In case of changes in the working conditions one can either increase the shielding thickness or think about a local shielding for the target.

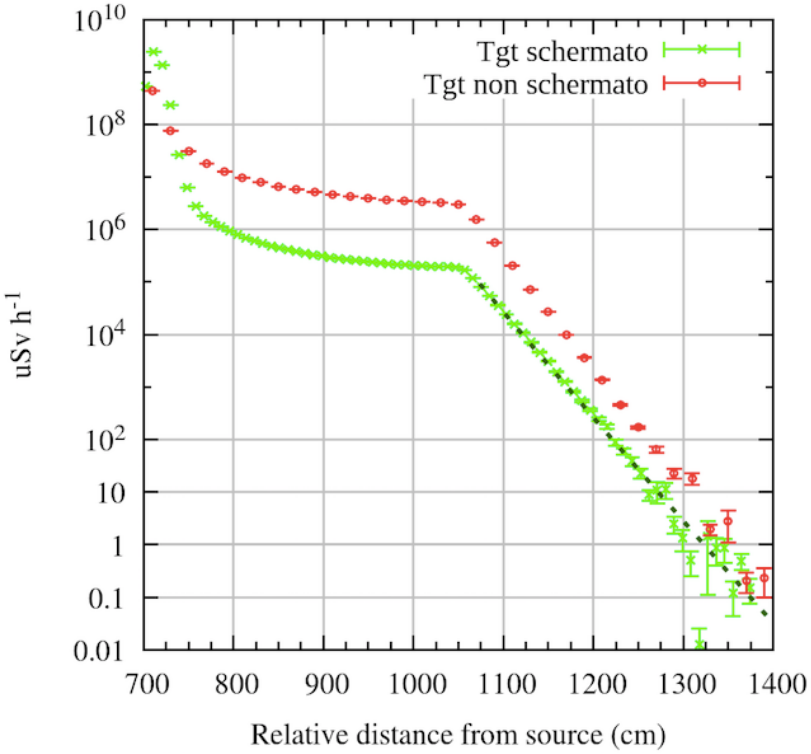


Figure 16 Transmission of the neutron dose rate in air (in the range x=700-1050 cm) and in concrete (x=1050-1400 cm) with and without local shielding. Results scaled per 1 μA protons on target.

A local shielding could have the advantage of reducing the radiological impact due to the target irradiation with high power protons, both

- during beamtime, reducing the dose rate inside and outside of the bunker (with a positive effect on the air activation, thanks to the possibility to contain neutrons)
- after the end of irradiation, mitigating the residual dose rate in presence of the hot target.

One of the possible configurations consists of using an internal layer of lead, an intermediate layer of polyethylene and an external layer of lead. The internal Pb layer can slow down neutrons through inelastic scattering so that polyethylene could more effectively absorb them, while the outer Pb layer could work as a shield against gamma radiation emitted from the target (prompt and residual) and also coming from the absorption of neutron by PE with consequent release of 2.0 MeV photons.
In Figure 16 it is shown the neutron dose rate as a function of the distance from the target. Between 0 and 300 cm the transmission is through air while between 300 and 650 cm it is through concrete. The dose rate in presence of the local shield is compared with dose rate without local shield. At the target position the dose rate with local shield is slightly greater because of the reflections inside the internal lead layer.
The neutron fluence out of the local shield as a function of the energy is shown in figure 17. The evaporation peak for backscattered neutrons (curve 150°-180°) is more important than the other directions because from the design – and from the implementation in the

Monte Carlo geometry – in the opposite direction with respect to the proton beam, the target is not shielded (because of the intrusion of the beam vacuum chamber) so there is a small light cone through which neutrons can be backscattered.
In Figure 18 the average (in angle) neutron spectrum is shown. The neutron distribution in each single layer is shown :

- tgt2vacuum: neutron produced in the target moving outwards
- vacuum2Pb: from the target to the internal lead layer (backreflected neutrons are not accounted for)
- Pb2PE: from the internal layer of lead to the intermediate polyethylene, evident the slowing down effect
- PE2Pb: from polyethylene to the external lead, on a separate plot to highlight the different scale
- Pb2vacuum: surviving neutrons moving toward the bunker.

While neutrons of about 1 MeV energy are very effectively absorbed by polyethylene, 10 MeV neutrons won't be completely stopped (10% of those arising from the internal lead do not undergo absorption).
Just one note concerning the units: the fluence in Figure 18 (a) and (b) are truly expressed as cm^{-2} sr^{-1} μA^{-1}, with the right normalization by the emission surface (otherwise it would have been impossible to appreciate the fluence attenuation from one layer to the other). In figure 17, the normalization by the emission surface has not been done (the unit cm^{-2} is meaningless in the unit expression), in order to show the total emission at arbitrary distance from the target. Baring this in mind, the two images are directly comparable.
So, just to conclude on the local shielding effectiveness, the neutron dose rate out of the bunker door is about 0.02 μSv/μAh, that is 10 μSv/h with the proton current of 500 μA. If the facility operates with this parameter for 100 hours per year the dose rate outside of the facility door is 4 μSv/h.

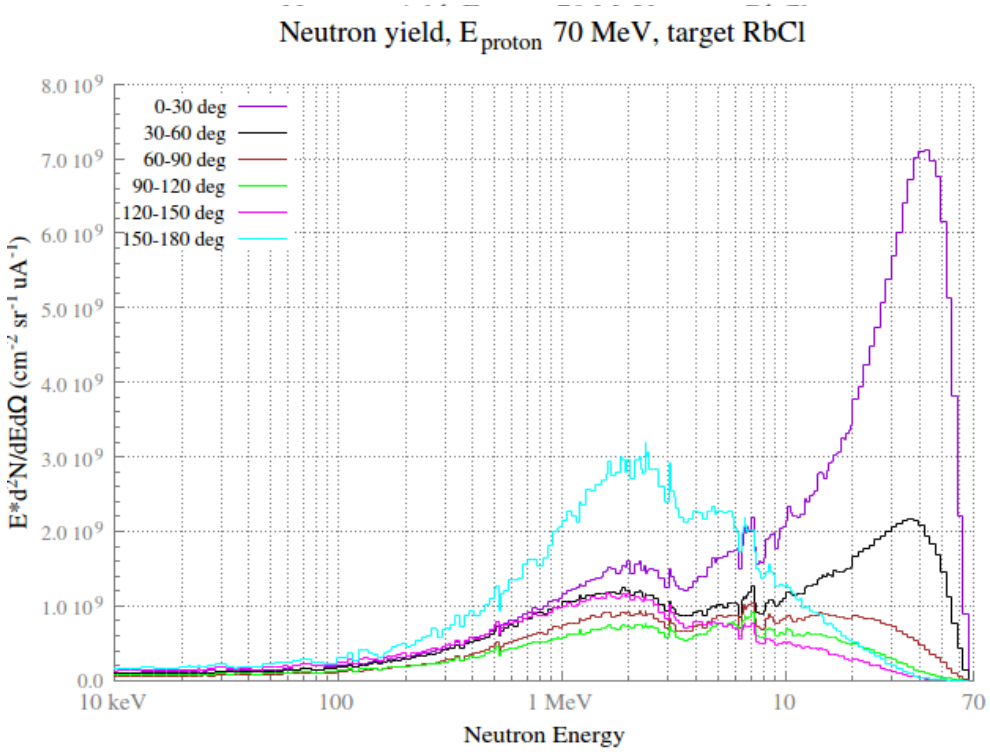
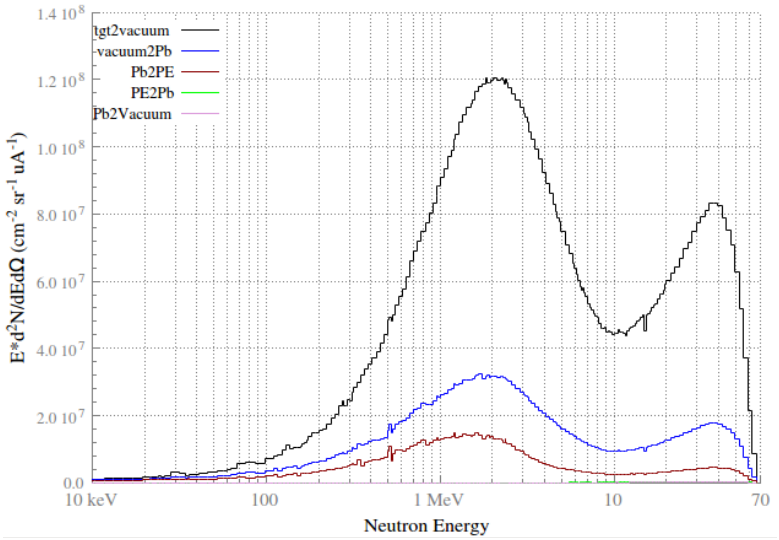
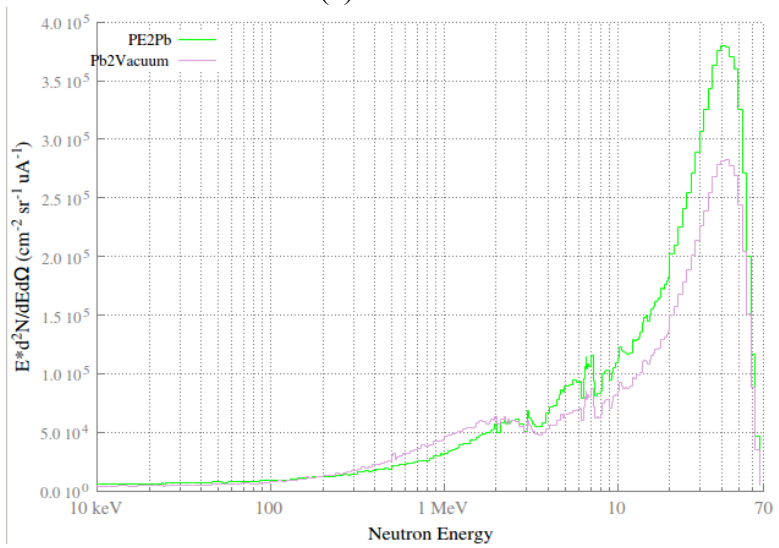


Figure 17 Energetic neutron spectrum outside of the local target shield.



(a)



(b)

Figure 18 Energetic distribution of neutrons (average on the whole solid angle) coming from the local shield layers.

As already mentioned, the dose rate can be mitigated increasing the shielding thickness, as represented in figure 19 where a proton beam current of 1 mA has been sent on a ^{85}Rb target in the bunker RI1, with a shielding thickness of 450 cm.
The dose rate outside of the shielding is about 10 µSv/h (where 350 cm and half the current provided 80 µSv/h).
What is interesting to notice in this calculation, is that a great contribution to the dose is given by neutrons channeling through the small spaces between the door and the floor or the side walls. In the simulation geometry, a small gap of 2 cm has been left to detect whether a rough sealing close to the irradiation source would constitute or not a problem. The results show that neutrons from that path contribute for 20 µSv/h to the total dose rate, thus reducing the efficiency of a big shielding wall. Engineering studies are under development to find a technical solution to this.

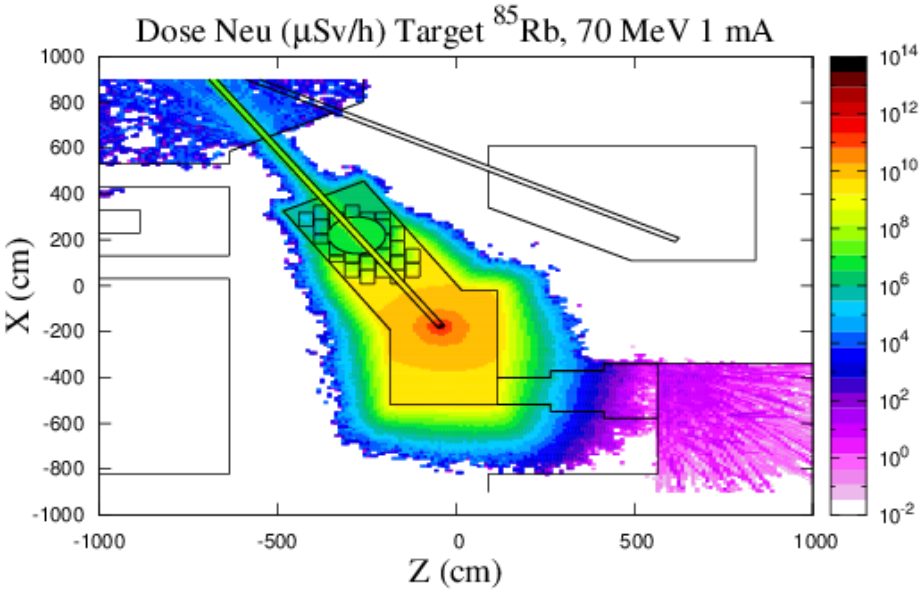


Figure 19 Neutron dose rate (µSv/h), plan view, during irradiation of rubidium target with 70 MeV energy proton of 1 mA current.

5.2.2 Cross section bunker L3C

The operation at low proton beam intensity for cross section measurements has been studied considering a proton beam of energy 70 MeV and maximum current 100 nA impinging on a graphite beam dump (in this application, the proton energy degradation passing through the thin foils has been neglected).

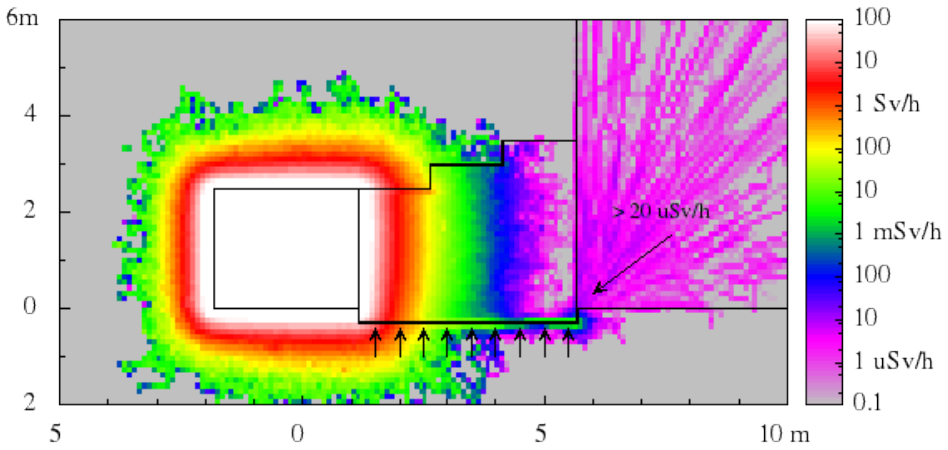


Figure 20 Neutron dose rate, front view: the arrows highlight the duct available for neutrons between the door and the floor.

In order to have a dedicated space for this practice, an area will be delimited in the A15 vault, with concrete blocks to shield it of 1 meter thickness each. Obviously 1 meter is not enough to guarantee a dose rate close to the environmental background on the external perimeter of the facility, as shown in Figure 21. As can be seen from the picture a dose rate of approximately 10 μSv/h is reached at the external door position (a dose limit of 1 mSv/year for that area implies less than 100 hours per year of operation, meaning a restriction for this experimental activity).

Similar to what has been calculated for the high intensity bunkers, the advantage of using a local shield for the beam dump has been evaluated, with the results shown in Figure 22: the dose rate at the external door is about 0.2 μSv/h, not constituting a limit for the practice (the shield included in this geometry is identical to the illustrated in the previous paragraph).

In this context, the other important advantage of using a local shield for the beam dump is that, when the operator must access to collect the stack foils for spectrometric analysis, he will not be exposed to the residual dose rate coming from the beam.

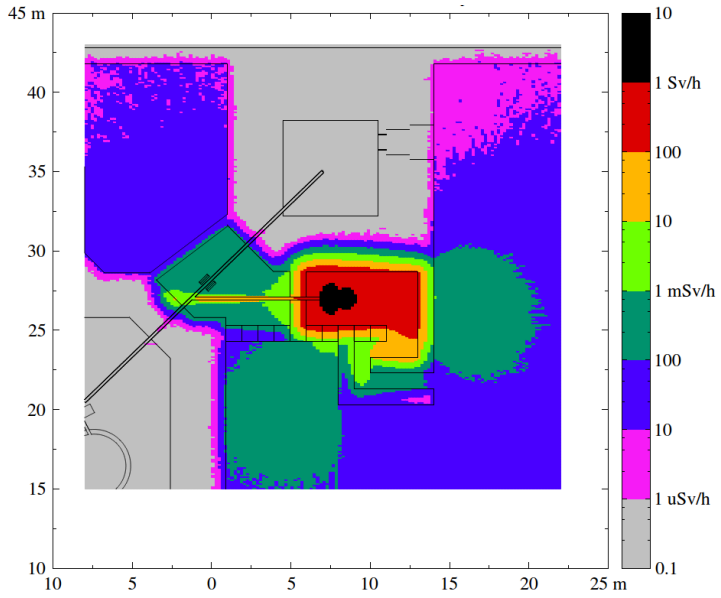


Figure 21 Neutron dose rate during irradiation in the bunker, for the cross section measurements.

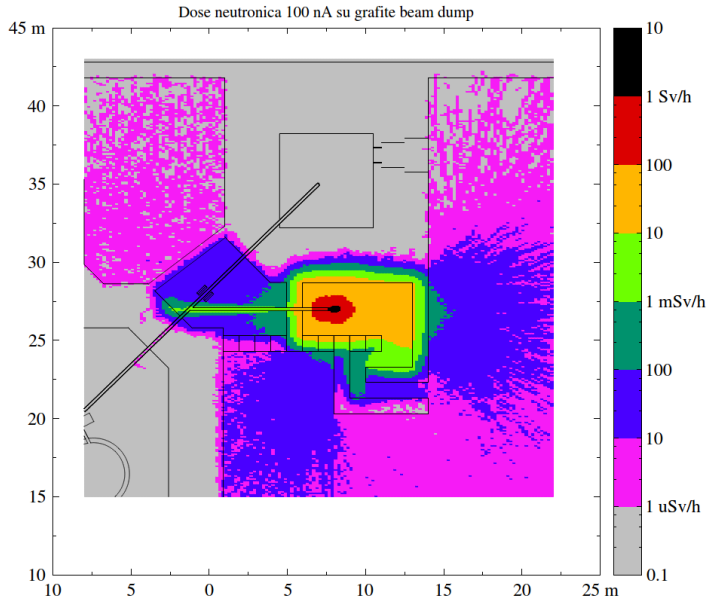


Figure 22 Neutron dose rate during irradiation in the bunker for the cross section measurements, including a local shield close to the beam dump.

5.2.3 Beam line Vault L3b

Based on the calculations and on the experience gained during the commissioning phase, it has been seen that proton beam losses might occur along the beam line from the

cyclotron to the target stations in RI3 and in L3C. Those losses are listed in Table 5 as specified in [30, 33]. They include the losses in the whole transport line from the cyclotron to the target.
Being the current lost a fraction of the current transported to the target, the secondary radiation field due to interaction with the beam line components (including vacuum chamber, quadrupoles, slits) might contribute to increase the ambient dose at the outside of the facility. This source of radiation requires a shielding in order to keep the dose for workers and public a slow as reasonably achievable. As a reference point it has been taken the door P2, actually it is just a point for goods' transfer for civil works and it is not used for personnel access.
The shielding has been optimized taking into account that the door is 15 meters distant from the irradiation point and when the proton beam is extracted from the cyclotron the access in area A9 in forbidden.
Taking as input a proton beam of 70 MeV energy, the neutron ambient equivalent dose rate at 1 meter from the interaction point is about 1.5 Sv/h m^2 for a beam current of 1 μA (see Figure 23, from [34]). If the project goal is to have 0.1 uSv/h at the reference point, a simple calculation can be done to infer the thickness needed.
Instead of rigorously applying the well-known formula

$$H(d,x)=H_0.\exp(-\lambda.x)/d^2$$

one can take advantage of the results obtained by [35] and resumed in Figure 24. The source term at 1 meter must be scaled to inverse of the square distance (1.5 Sv/h m^2 will be 6.7 mSv/h at 15 m distance) and the new dose rate reduced by a factor *x* to obtain the project goal value (6.7 mSv/h divided by 0.1 uSv/h results in $x = 6.7\ 10^4$). The factor x, directly compared with the curves of Figure 24, will give the shielding thickness needed in concrete.
The results are shown in Table 6, they do not take into account the occupancy factor and have been obtained using the curve at 0°, that is a strong approximation considering the beam optics along a straight line. Moreover, as already mentioned, the evaluated losses must be reduced at least of 1/3 for the part on the line under investigation.
The results of the Monte Carlo simulations have also been represented.

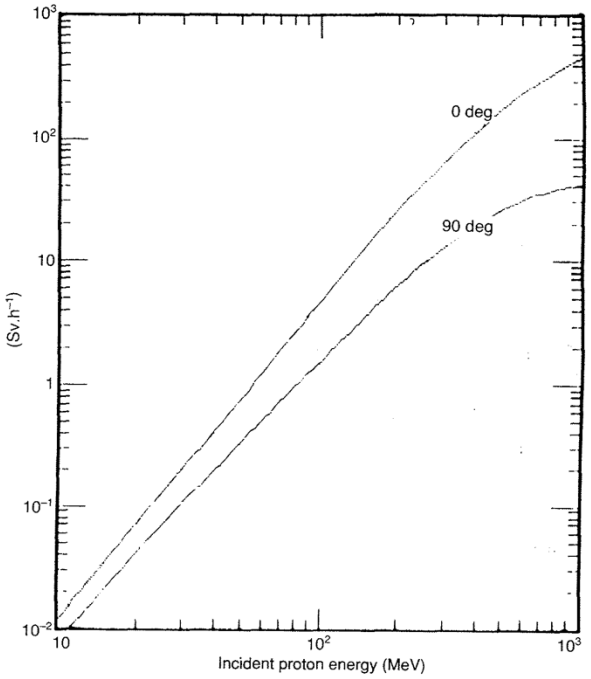


Figure 23 Source term for shielding calculations expressed as dose rate at 1 m and 0 and 90 deg for a beam of 10^{12} protons per second incident on a thick target. [34]

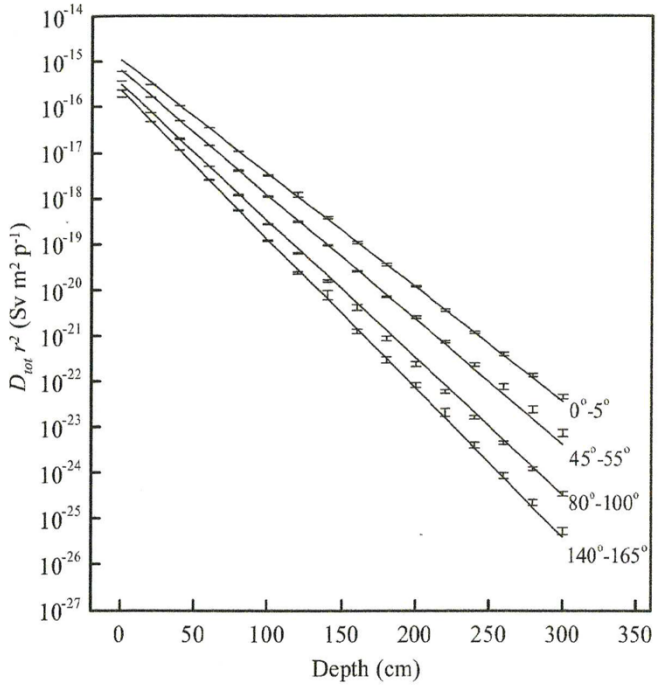


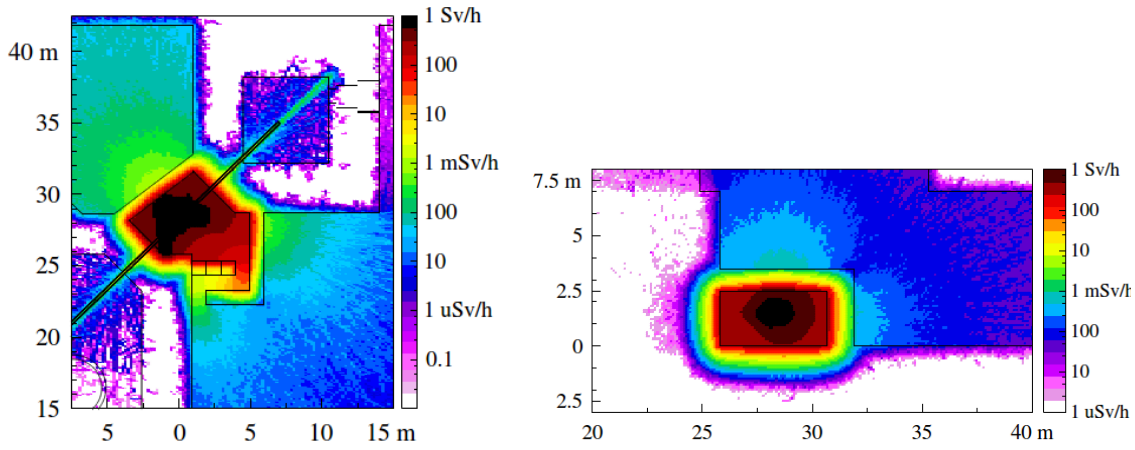
Figure 24 Transmission through concrete at different angles, [35].

Table 5 Beam losses from the cyclotron to the bunker RI3, as evaluated in [33]

Energy (MeV)	Evaluated beam loss %	Current lost per μA on target (nA)
35	0.58%	5.8
50	0.31%	3.1
70	0.17%	1.7

Table 6 Shielding thickness calculated on the evaluated beam current lost along the beam line.

Energy (MeV)	Current lost per 500 μA on target	Source term at 0° (Sv h^{-1} m^2) per 10^{12} proton	Source term at 0° (Sv h^{-1} m^2) per calculated beam current	At 15 m (mSv/h)	Factor *x*	Shielding thickness (cm)
35	2.9 μA	0.3	5.4	24	2.4 10^5	220
50	1.5 μA	0.7	6.6	4.4	4.4 10^4	180
70	0.85 μA	2.0	10.6	47	4.7 10^5	235



(a) (b)

Figure 25 Neutron ambient equivalent dose rate due to the beam lost during transport in the L3b vault. All along the installation perimeter the walls have 1 m thickness. On the left a plan view is shown and on the right there is a front view. Values have been normalized for 200 nA current.

5.3 ENVIRONMENTAL IMPACT OF THE INDUCED RADIOACTIVITY

5.3.1 Earth and groundwater

The shielding beneath the irradiation bunker is 3 meters thick and it is completely made in concrete, so the eventuality that radioactive nuclides are generated in the earth after a direct irradiation by the proton beam is excluded.
The radionuclides generated in the most internal layer of the shielding are kept into the structure. They shall not migrate to the environmental matrices thanks to the isolation of the building with respect to the ground of the dig.
The only possibility to find activation in the environmental samples is the transmission of neutrons through the shielding and the absorption of those neutrons by the earth and groundwater and consequent generation of radioactive nuclei. It has been evaluated, then:

a) the chance for the radionuclides produced in the groundwater, to be found in the water supply system in the immediate neighborhood of the laboratory
b) the radioactivity possibly filter through the groundwater causing a potential contamination of the drinkable water.

A numerical simulation has been run using as source parameters: proton beam energy of 70 MeV and 0.2 mA current on a rubidium chloride target deposited on copper. It has been supposed to last the irradiation 10000 days in order to take into account the build up of long half-life radionuclides.
The target has been placed in the center of a room with concrete floor, 2.35 g/cm^3 density and 3 m thickness. Beneath the floor, at 90° with respect to the target two samples have been placed, a water and an earth sample of 0.4 m^3 each.
The earth used for the calculations has density of 1,6 g/cm^3 and the elemental composition is specified in Table 7. This weight molecular composition is referred to a sample collected between 7.5 and 8.5 meters underground by the dig of the SPES installation.

Table 7 Weight composition of the earth at the LNL, 8 meters underground.

Molecule	Units	Value
Na_2O	% p/p	0.52
MgO	% p/p	4.22
Al_2O_3	% p/p	5.08
SiO_2	% p/p	19.4
K_2O	% p/p	0.69
CaO	% p/p	24.8
MnO	% p/p	0.03
TiO_2	% p/p	0.24
Fe_2O_3	% p/p	1.32

P_2O_5	% p/p	0.10
SO_3	% p/p	0.09
Barium	mg/kg	35
Cesium	mg/kg	< 100
Cobalt	mg/kg	1.7
Chrome	mg/kg	4.8
Europium	mg/kg	< 0.5
Phosphorus	mg/kg	197
Nickel	mg/kg	5.4
Copper	mg/kg	0.9
Rubidium	mg/kg	< 0.5
Strontium	mg/kg	152
Titanium	mg/kg	54
Vanadium	mg/kg	27
Zinc	mg/kg	12

The sample water has been considered salts and impurities-free, because it was supposed that the radioactive elements possibly dissolved in water were coming from the ground around it, so they have been included in the ground calculations.
Results show that the specific activity in both samples is well below 0.01 Bq/g, following the conservative hypothesis of a continuous irradiation lasting 10000 days (less than 30 years).

Table 8 Radioactivity induced in the earth sample beneath the irradiation bunker.

Nuclide	$T_{1/2}$	Bq per kg of the earth sample	Effective dose coefficient per introduction by ingestion (adults > 17 years old) Sv/Bq	Total effective committed dose per introduced weight unit[3] uSv per kg
Cs-134	2,06 y	$2,4\ 10^{-3}$	$1,9\ 10^{-8}$	$\sim 10^{-5}$
Mn-56	2,6 h	$1,7\ 10^{-3}$	$2,5\ 10^{-10}$	$\sim 10^{-7}$
Fe-55	2,7 y	$1,8\ 10^{-3}$	$3,3\ 10^{-10}$	$\sim 10^{-7}$
Cr-51	27,7 d	$1,6\ 10^{-2}$	$3,8\ 10^{-11}$	$\sim 10^{-7}$
Ca-45	162,6 d	$3,0\ 10^{-2}$	$7,1\ 10^{-10}$	$\sim 10^{-5}$
K-43	22,3 h	$1,7\ 10^{-2}$		~
K-42	12,4 h	$2,5\ 10^{-2}$	$4,3\ 10^{-10}$	$\sim 10^{-6}$
Ca-41	10^5 y	$8,9\ 10^{-7}$	$1,9\ 10^{-10}$	$\sim 10^{-11}$
Ar-39	269 y	$5,5\ 10^{-5}$		~
K-38	7,6 m	$6,8\ 10^{-2}$		~
Ar-37	35,0 d	$3,4\ 10^{-1}$		~
P-32	14,2 d	$5,6\ 10^{-3}$	$2,4\ 10^{-9}$	$\sim 10^{-5}$
Si-31	157,3 m	$9,7\ 10^{-3}$	$1,6\ 10^{-10}$	$\sim 10^{-6}$
Al-28	2,2 m	0,3		~
Mg-27	9,5 m	$3,0\ 10^{-2}$		~
Na-24	14,96 h	0,2	$4,3\ 10^{-10}$	$\sim 10^{-4}$
Na-22	2,6 y	$7,2\ 10^{-3}$	$3,2\ 10^{-9}$	$\sim 10^{-5}$
F-18	109,77 m	$1,7\ 10^{-2}$		~
N-16	7,13 s	$6,6\ 10^{-2}$		~
O-15	122 s	0,17		~
C-14	5730 y	$1,7\ 10^{-6}$	$5,8\ 10^{-10}$	$\sim 10^{-9}$
N-13	10 m	$1,7\ 10^{-2}$		~
C-11	20,4 m	$3,4\ 10^{-2}$	$2,4\ 10^{-11}$	$\sim 10^{-7}$
Be-7	53,3 d	$1,2\ 10^{-2}$	$2,8\ 10^{-11}$	$\sim 10^{-7}$
H-3	12,33 y	$1,2\ 10^{-3}$	$1,8\ 10^{-11}$	$\sim 10^{-8}$

[3] It is only reported the order of magnitude.

On the left of the graph in Figure 26 it is possible to see that the radioactivity of short halflife nuclides reach the saturation activity on the first day of irradiation, so rapidly their radioactivity will decay (the curves of ^{16}N and ^{38}K coincide, so do the curves of ^{13}N and ^{18}F). For long halflife radionuclides the one contributing most to the water radioactivity is tritium ($T_{1/2}$ =12.33 years). Even considering a very long irradiation period the saturation activity is lower than 1 mBq/g.
In Table 8 a list of the radioactive elements possibly found in the ground close by the facility with the respective quantities.
The paths for the radionuclides to reach the public are:

- Dilution in the groundwater and transfer to the public water supply system
- Through absorption by the plants
- Food chain

Tipically radionuclides with short halflife do not constitute a hazard because they decay faster than they transfer to the groundwater time. On the other side radionuclides with long halflife do not reach the saturation activity. For these reasons radionuclides with halflife in the range of a few hours to 100 years are taken into consideration (for completeness in the table all the radionuclides theoretically produced have been reported).
For each of them it has been indicated: the halflife, the specific radioactivity in the sample of earth/water, the dose to radioactivity coefficient considered for adults of age 17 and older, as from the Decree 230/95 and modifications (units of Sv/Bq). In the last column, it has been reported the effective committed dose, obtained multiplying the values of the previous two columns. It must be bear in mind that not all of the calculated radionuclides are soluble, so it is reasonable to take into account a percentage within 0,1 and 1% of the total calculated activity

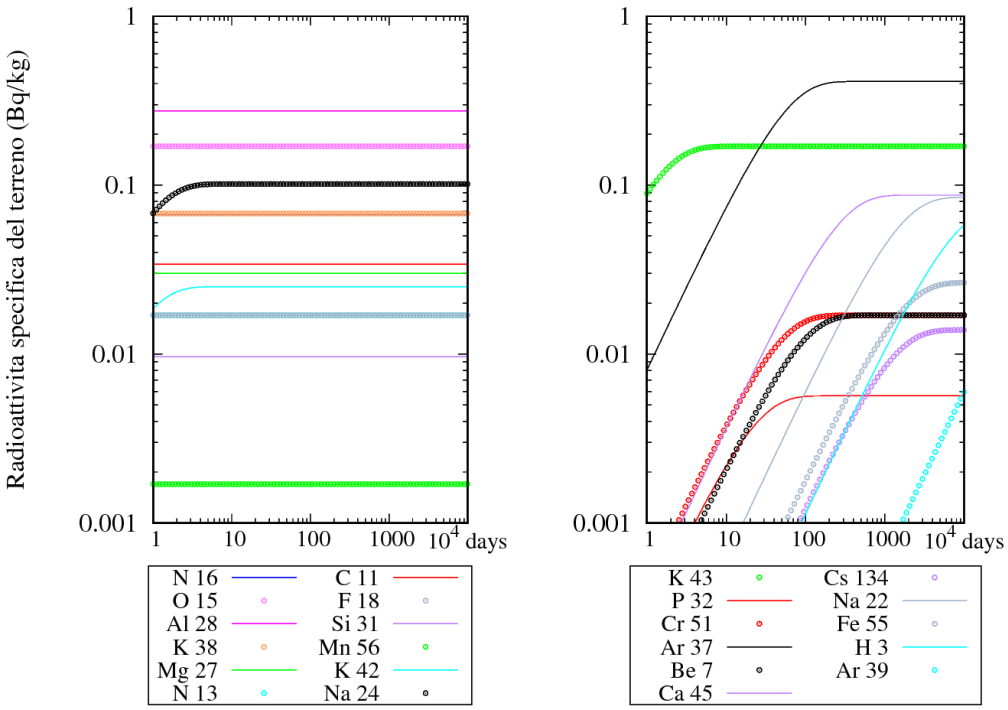


Figure 26 Build up of the specific radioactivity in the ground, due to the continuous irradiation of a ^{85}Rb target. On the left radionuclides of $T_{1/2}$ shorter than one day, on the right $T_{1/2}$ shorter than 300 years.

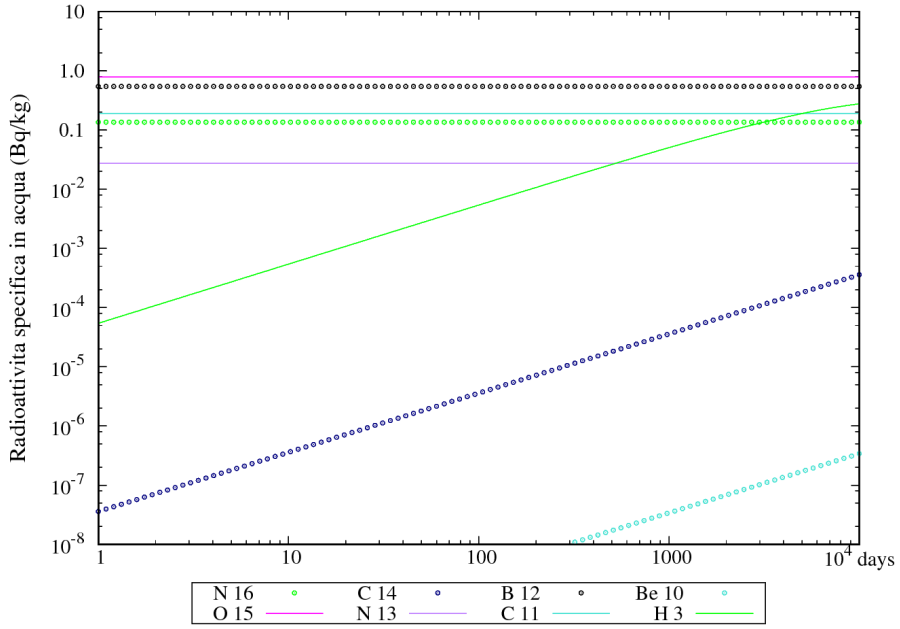


Figure 27 Radioactivity concentration in groundwater.

For what concerns the earth activation, the specific radioactivity is well below 10 mBq/g, with a great contribution from ^{37}Ar, ^{28}Al. Metallic elements do not show a significant level of radioactivity.

5.4 AIRBORNE ACTIVATION

In this paragraph, the results of the calculations of radioactivity induced in air are shown. As it will be shown, the goal - and the reference value for the radioactivity concentration - is 1 Bq per gram of air. This value is the concentration limit for unauthorized release of short halflife radionuclides (less than 75 days) to the environment, as stated in section 30 of the Decree 230/95 as modified, explained in paragraph 6.3.

5.4.1 Irradiation vault RI#1-2-3

The air inside the irradiation vaults is activated by the secondary neutron absorption emitted during the irradiation of the target. Standard air density is 1.205 10^{-3} g cm^{-3} and composition, as implemented in the calculations modelling, is N_2 (75,5 %), O_2 (23,2 %), Ar (1,3 %) and C (0,12 %).
The considered ventilation plant works to withdraw the air from the outside and release it to the irradiation bunker after filtering and conditioning in order to have standard humidity and temperature.
The absorption cross section for neutrons by the air components have been shown in Table 9, in particular ^{41}Ar has cross section higher than 1 barn for thermal neutrons, as seen in Figure 28.
The air change inside the vault occurs at a rate suitable to guarantee a negative pressure of 60 Pa with respect to the rooms close by. From the measurements done in the first commissioning operations, it has been seen that this value is reached changing the air at a rate of 500 m^3/h.
The amount of extracted radioactivity has been calculated through the application of the equations explained in paragraph 2.2.1 and the evaluation of the production rates with the Monte Carlo code FLUKA. The two-step approach for airborne activity evaluation, as explained in paragraph 3.1.1, has been followed.
In Table 9 the production rates of the radionuclides generated in air and the radioactivity concentration of the air extracted from the bunker have been reported. The irradiation parameters for these calculations were proton energy 70 MeV and current 100 μA. The value in the fourth column (radioactivity concentration in units Bq per gram of air) refers to a continuous irradiation lasting one day, so that the approximation of equilibrium situation of production and removal of radioactive species can be applied without further corrections. It has to be noted that, when the irradiation time is shorter than one day (few hours in many cases) the calculated quantities might overestimate the effective production

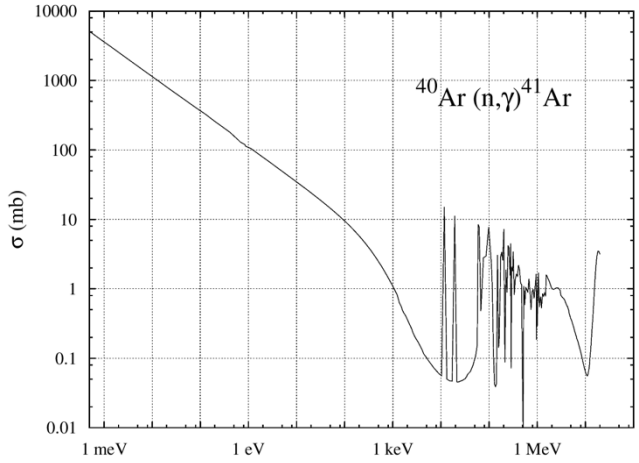


Figure 28 Cross section of the reaction ^{40}Ar (n,γ) ^{41}Ar. For thermal neutrons, the cross section is about 1 barn [20].

Table 9 Production rates in air, evaluated for the irradiation of a RbCl target with 70 MeV energy proton beam at 100 μA current.

Radionuclide	$T_{1/2}$	Atomi/prot. on target	Bq/g released
^{3}H	12,33 y	$4{,}4\ 10^{-6}$	$4{,}5\ 10^{-5}$
^{7}Be	53,3 d	$1{,}5\ 10^{-5}$	0,01
^{11}C	20,3 min	$6{,}1\ 10^{-5}$	136
^{13}N	9,96 min	$3{,}4\ 10^{-5}$	116
^{15}O	122,2 sec	$8{,}6\ 10^{-6}$	50
^{41}Ar	109,34 min	$2{,}0\ 10^{-7}$	0,12
Total			302

The radioactivity concentration extracted is 302 Bq/g. Scaling for a proton current of 500 μA the concentration at the extraction becomes 1510 Bq/g.
The ventilation system conveys the air of the whole installation to a unique stack, so that the radioactivity concentration has to be averaged over the total amount of air extracted, that is about 75000 m^3 ($9\ 10^7$ grams). The final concentration becomes, then, about 10^{-5} Bq/g.
The release to the environment is controlled by a spectrometric measurement system in order to interlock the cyclotron in case of concentration release exceeding 1 Bq/g.

5.4.2 Beamline vault

In vault hosting the beamline that drives the proton beam from the cyclotron to the irradiation bunker, the radioactivity concentration induced in air because of the proton beam losses is about 600 Bq/g, considering the facility working at full power (500 μA, 70 MeV protons on target). For this area there is no ventilation system foreseen, so the policy will be to limit the access to that area until a concentration less than 1 Bq/g is reached.
For the specific case, after a continuous working period of one day, the waiting time will be on the order of 20 hours. During full power irradiation, most likely, an immediate maintenance intervention on the beam line equipment won't be requested, so that 20 hours is a reasonable time to wait before entering.
The calculations shown so far take into account the high intensity parameters and dedicated vaults. The low intensity beamline, namely the one dedicated to cross section measurements, where the maximum proton current is 100 nA, will not present hazards form this point of view since the produced radioactivity is less than 1 Bq/g, so there are no restrictions to the access related to air activation risks.

5.5 Cooling Water Activation

5.5.1 Cyclotron cooling system

Taking into account the volume of the cooling pipes close to the proton beam (that is the water directly irradiated by the secondary radiation due to the beam current lost during the acceleration stage), the ratio of the total flow rate in water cooling circuit to the volume of the pipes mentioned provides the water change rate for activation calculations. The small irradiated volume considered flows in the whole circuit to, ideally, come back in its initial position close to the irradiation point, after a time calculated as the ratio of the tank volume to the flow rate.
The induced radioactivity, calculated for an irradiation period of one hour, must take into account the continuous flow rate of the cooling water.

5.5.2 Target cooling system

An evaluation of the activation of the targt cooling water has been made, considering a proton beam current of 100 μA and energy 70 MeV. The results refer to the production of radionuclides due to the beam (proton or secondary) interaction with the water components, while other occurrences are not taken into account. It may be, for example, the case of ^{7}Be, produced in other materials and then released to the cooling water through leaching phenomena. This is not accounted for, but can still contribute to the coolant radioactivity when measured.
The radionuclides mostly contributing to the total radioactivity are: ^{15}O ($T_{1/2}$ 2.1 min), ^{13}N ($T_{1/2}$ 10 min), ^{11}C ($T_{1/2}$ 20 min) and ^{7}Be ($T_{1/2}$ 53 days). Tritium (^{3}H, $T_{1/2}$ 12.33 years)

contribution is very poor, due to its long halflife and, consequently, the very slow build up in the water cooling circuit.
With an irradiation period of a few hours, every radionuclide – exception made for ^{7}Be – reach saturation activity. After about 6 hours the specific radioactivity decays to less than 1 Bq/g.

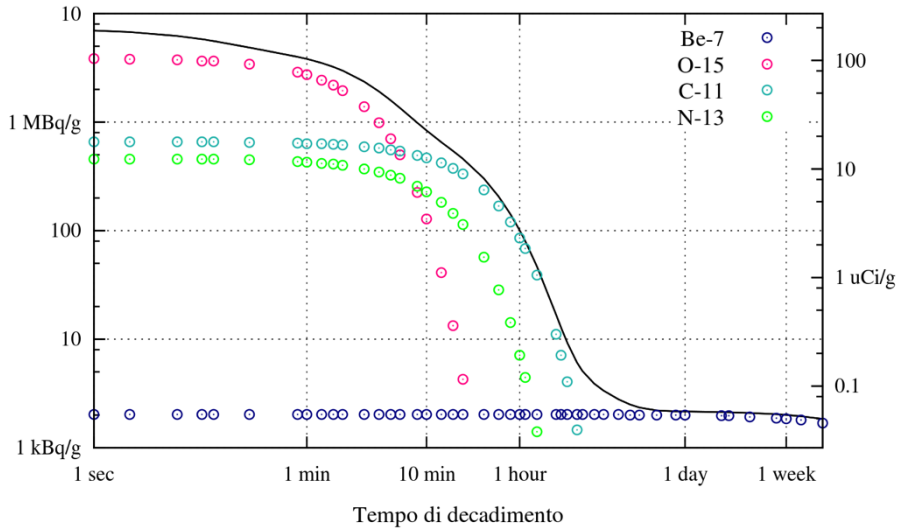


Figure 29 Radioactivity induced in the target cooling water, as a function of the cooling time, after an irradiation of 1 day with proton beam energy of 70 MeV and 100 μA current. The sull line represents the total radioactivity. Values have been reported as multiples of Bq units on the left axis and submultiples of the Ci units on the right axis.

The radioactivity concentration of ^{7}Be after an irradiation lasting 1 day is about 2 kBq/g, as shown in **Errore. L'origine riferimento non è stata trovata.**29. In Figure 30 it has been represented the buildup evolution of ^{7}Be and ^{3}H as a function of the irradiation period. In view of preparing treatment and/or retrieval procedures for the target cooling water, it will be referred to the specific activity of ^{7}Be because, after a few hours of cooling time, the residual radioactivity will be only due to this isotope.

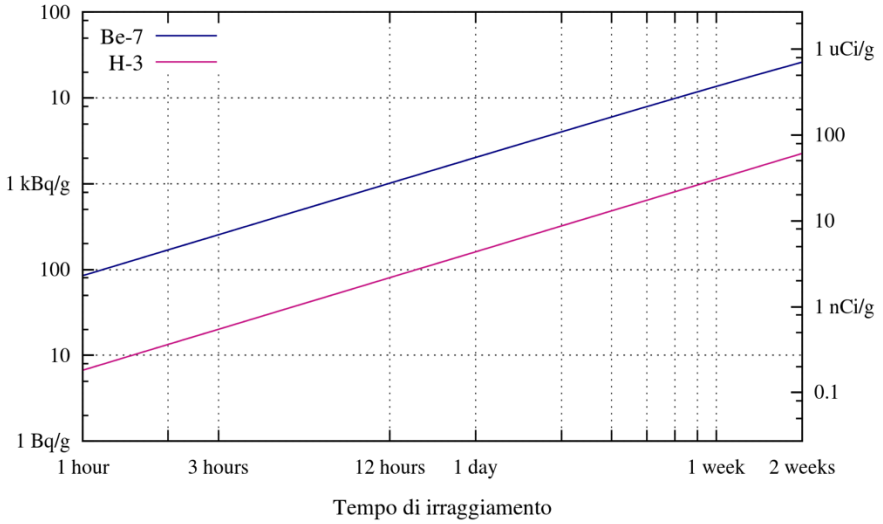


Figure 30 Build up of the radioactivity due to ^{7}Be and ^{3}H as a function of the irradiation time, with proton beam energy of 70 MeV and 100 μA current. Values have been reported as multiples of Bq units on the left axis and submultiples of the Ci units on the right axis.

5.6 TARGET ACTIVATION

Considering the specific case of the irradiation of rubidium (either natural or highly enriched in the isotope ^{85}Rb), the target is chemically processed not before one week after the irradiation, in order to let the contaminant isotope ^{83}Rb decay, so that the ratio ^{82}Rb to ^{83}Rb is optimized. Thus, after an irradiation, the radioactive target will be left inside the irradiation bunker for a period longer than 6 days.

Entering in the irradiation cave during this period is forbidden, the gamma dose rate can still be as high as hundreds of mSv/h as a function of the distance from the target station, or in presence of a local shielding [36], as a function of both distance and orientation in space of the shielding and the beam line opening (the dose rate is much lower in presence of a local shielding, but the target will be transferred unshielded to the radiochemistry, so its radioactivity should still be accounted for).

Once transferred the target to the radiochemistry laboratory for treatment, the support structure – schematically assumed to be an aluminum box in the simulations – remains inside the irradiation cave. The residual dose rate, due to this structure activation, at one meter distance is about 100 mSv/h after the end of the irradiation, and 100 μSv/h after one week by the end of irradiation, that is the instant when the target is transferred to the radiochemistry. A detailed time evolution of the residual dose rate is reported in Table 10.

When a local shield is used, the ambient equivalent dose rate strongly depends on the direction of approach to the irradiated target. From the simulation results it has, infact, been seen that the gamma radiation emission at an angle of 180° can still be important, despite the presence of a local shielding.

Table 10 Gamma dose rate at the distance of 1 meter from the target when the target is in or out of the bunker. The local shield is considered here.

Decay period	Dose rate at 1 meter distance from the support		Dose rate at 1 meter distance from the target	
	0°	180°	0°	180°
1 sec	24,8 mSv/h	85 mSv/h	30,4 mSv/h	875 mSv/h
1 hour	14,1 mSv/h	21,2 mSv/h	17 mSv/h	245 mSv/h
1 day	100 uSv/h	745 uSv/h	160 uSv/h	27,6 mSv/h
1 week	2 uSv/h	52 uSv/h	5,8 uSv/h	2,9 mSv/h
10 days	1,4 uSv/h	27 uSv/h	3,6 uSv/h	2,1 mSv/h
2 weeks	0,8 uSv/h	20 uSv/h	2,7 uSv/h	1,8 mSv/h

Table 11 Gamma dose rate at the distance of 1 meter from the target when the target is in or out of the bunker. The local shield is not considered here.

Decay period	Dose rate at 1 meter distance from the Al support	Dose rate at 1 meter distance from the irradiate target
1 sec	91,4 mSv/h	575 mSv/h
1 hour	59,3 mSv/h	401 mSv/h
6 hours	46,7 mSv/h	283 mSv/h
1 day	20,3 mSv/h	172 mSv/h
3 days	2,2 mSv/h	143 mSv/h
1 week	37 uSv/h	123 mSv/h
2 weeks	11 uSv/h	105 mSv/h

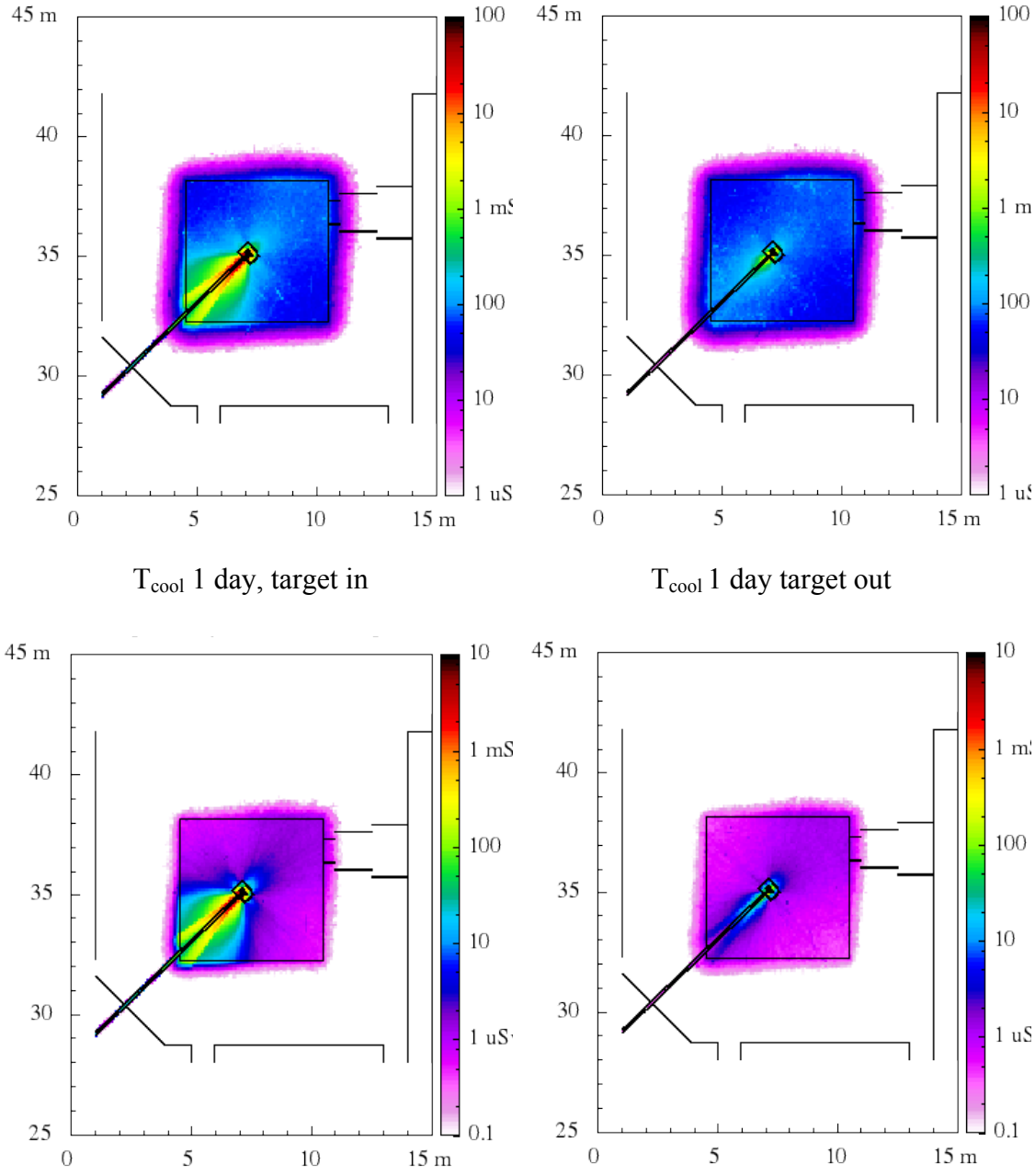


Figure 31 Residual gamma dose rate after an irradiation of 5 hours with proton beam current of 100 μA. Top left: after one day cooling, with target in the bunker; bottom left: after one week cooling with the target in bunker; top right: after one day cooling, target removed from the bunker; bottom right: after one week cooling target removed from the bunker.(on x and y axis relative positions in meters can be read). Local shielding of the target on place.

After the target is transferred to the radiochemical laboratories, the bunker is normally classified as controlled area and the access is ruled, scheduling with due time in advance, duration, procedures and type of the intervention.

In Figure 32 and Figure 33 the contribution to the dose rate from the target and the support as a function of the decay time is shown. It is also represented the different contribution from the two main radioisotopes generated in aluminum, ^{22}Na and ^{24}Na, responsible respectively of the aluminum radioactivity on long and short timescales.

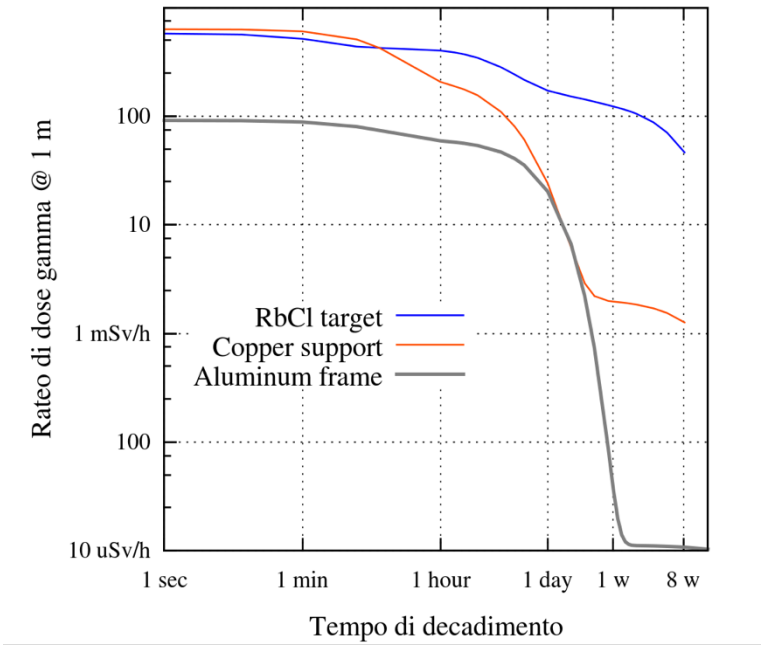


Figure 32 Residual gamma dose rate after the irradiation of a RbCl target with protons of energy 70 MeV and current 100 μA, lasting 5 days. Contribute to the dose rate from RbCl, from the copper support and from aluminum frame have been here represented.

5.7 RADIATION PROTECTION OF THE RADIOCHEMICAL LABORATORY

5.7.1 Laboratory facilities for radioisotope production

The design of a good radiochemistry laboratory takes into account the flow of work as the radionuclides are prepared, irradiated, recovered, purified, quality checked and packaged for transport [40, 41]. As a result of these activities, the laboratory facility should contain areas for each. In the facility under study a laboratory dedicated to spectrometry should be foreseen, as well, in order to conduct the required analysis on the samples used for cross sections evaluation. There will be a target processing area, which is usually contained in a shielded enclosure known as a ‘hot cell’. These hot cells shield the operator from the high levels of radioactivity present. The purification of the radionuclide or the conversion of a radionuclide into a radiopharmaceutical is usually carried out in these hot cells. There will be a quality control (QC) area, where the purity of the radionuclides or radiopharmaceuticals is checked [38, 39].

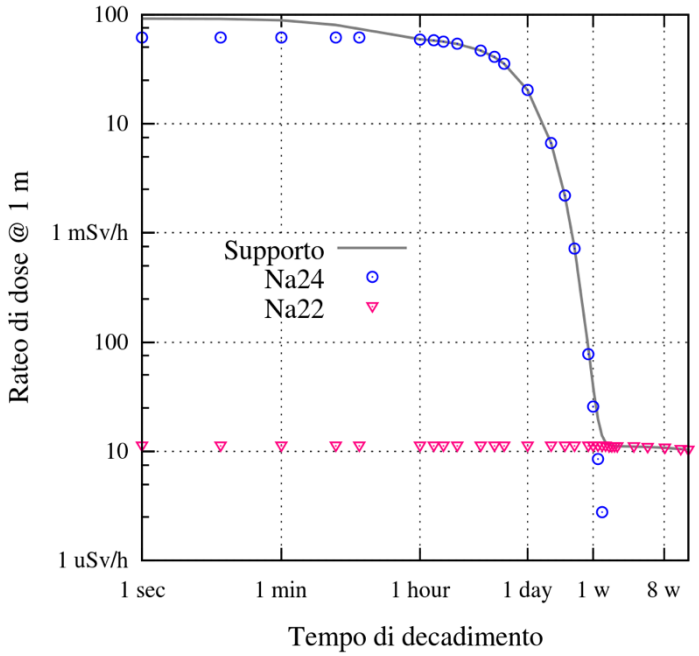


Figure 33 Residual gamma dose rate due to activation of the aluminum frame. The contribute of ^{22}Na and ^{24}Na has been highlighted, since they provide the highest dose rate of long and time-scales, respectively.

5.7.2 Classification

According to the radionuclides used, the radioactivity handled at any one time and the nature of the work involved, the radionuclide laboratory is classified as type I, II or III. [42], as from Table 12. The radioactivity has to be evaluated taking into account the multiplication coefficients of Table 13.
Recommendations concerning safety systems, containment and ventilation are set for the different laboratory classes, in order to avoid any spread of contamination.
The radiochemical laboratory of the LARAMED project will handle radioisotopes of *High radiotoxicity*: the radioactivity of the target will, for sure, exceed 100 mCi thus the laboratory must be classified as class I.
From the point of view of radiation protection, each area interested by the radioactivity flow should be classified as radiation area, in particular will be controlled areas those dedicated to: target processing, radiopharmaceutical preparation, target material recovery, temporary storage for potentially contaminated waste, quality checking, decontamination. The radiological classification is usually reviewed after the first

auditing and once defined, with due detail, the operative procedures for the radioactivity handling.
Every area with transit and/or manipulation of radioactivity is monitored with gamma radiation detectors and portable contamination monitors will be available where contamination hazard exists for the workers and for the working surfaces.
Exiting from the laboratory is subject to measurement with hand, cuff and foot surface contamination monitor: if no contamination is detected the worker can leave the area, while in presence of a contamination alarm the source of contamination must be investigated.

Table 12 Classification criteria for laboratories where unsealed sources are handled.

Radioisotopes group	Radiotoxicity	Minimum radioactivity[4]	Unit or laboratory type		
			III	II	I
			Radioactivity		
I	Very High	3700 Bq (100 nCi)	≤ 370 kBq (≤ 10 uCi)	370 kBq ÷ 370 MBq (10 uCi ÷ 10 mCi)	> 370 MBq (> 10 mCi)
II	High	37 kBq (1 uCi)	≤ 3700 kBq (≤ 100 uCi)	3700 kBq ÷ 3700 MBq (100 uCi ÷ 100 mCi)	> 3700 MBq (> 100 mCi)
III	Modest	370 kBq (10 uCi)	≤ 37 MBq (≤ 1 mCi)	37 MBq ÷ 37 GBq (1 mCi ÷ 1 Ci)	> 37 GBq (> 1 Ci)
IV	Low	3700 kBq (100 uCi)	≤ 370 MBq (≤ 10 mCi)	370 MBq ÷ 370 GBq (10 mCi ÷ 10 Ci)	> 370 GBq (> 10 Ci)

Table 13 Multiplication coefficients of the radioactivity handled for the classification asset.

[4] Below the minimum radioactivity the classification criteria are not applied.

Type of operation	Solid sources with scarce surface contamination	Liquids, solutions, suspensions	Solid sources with surface contamination	Gas, powders, liquids or solids with high vapour pressure
	Coefficient			
Storage and solid waste	1000	100	10	1
Very Simple Operation (elution, dilution)	100	10	1	10^{-1}
Simple operation	10	1	10^{-1}	10^{-2}
Complex operation	1	10^{-1}	10^{-2}	10^{-3}

Workers handling radioactivity will be classified as radiation workers, more precisely taking into account the duties and the working time. The access will be only granted to personnel that is adequately trained and classified.
Those workers who have to stay in the areas previously classified as controlled areas, will be classified as exposed radiation workers. If they have to be in those areas occasionally and not continuously, the classification will be done on a case-by-case basis.

5.7.3 Structures and Equipment Requirements

Some aspects are highlighted here without great detail (it can found in the recommendations, [42]).

VENTILATION

In general class I, II and III laboratories must be provided of adequate ventilation system in order to avoid any spread of contamination during both normal operation and in case of accident. In particular air should not be recycled internally, but introduced from outside after appropriate conditioning.
The air flow should be from less contamination areas to higher potential contamination areas, that is to say that areas with the higher potential contamination hazard must be kept in depression regime with respect to the others (opportune differential pressure class-dependent are defined in [42].
There must filters at the exhaust fit for the type and quantity of the effluent.

SURFACE MATERIALS AND FURNITURE

All the surface materials and furniture of the radionuclide laboratory shall be selected in a manner that makes them easy to clean. Attention shall also be paid to the following matters:

- The floor and the surfaces of working benches shall be made of materials impermeable to moisture and resistant to ordinary chemicals, such as dilute acids, alkalis and organic solvents.
- Joints and gaps shall be filled.
- The walls and the ceiling shall be made of materials that have a smooth surface.
- The working areas must be equipped with only the minimum furniture needed, the coatings of which do not accumulate dust and are easy to clean.

WASTE TREATMENT

For the waste produced in the LARAMED laboratories, it will be foreseen to store them temporarily if they contain radionuclides of half-life shorter than 100 days and if they reach, in 5 years, the exemption limits defined in the Decree 230/95.
If those constraints are not met, there will be an agreement with an authorized company to periodically collect and dispose them.
The systems and procedures to store the waste inside the lab will be suitable for the type and activity of the radioactive sources handled. In particular, they will be labeled to identify in detail the type of waste, the radionuclide contained, the activity, the type of operation that has produced it and as many other information as possible to trace the radioactivity inside the laboratory.
The effluent coming from basins or decontamination showers will be also collected in dedicated tanks, equipped with monitoring and sampling systems.
When the exemption limits will be reached, the waste will be disposed of in agreement with the environmental management requirements.

5.8 POTENTIAL EXPOSURES

The licensing procedure for installations with radiological risk, explicitly requires to estimate potential exposures (section 115-ter of the Decree 230/95 as modified). The scenario described in the following paragraphs show the possible consequences of technical plants failure or the default in the response of dedicated safety systems.

5.8.1 Pneumatic transfer block

During the pneumatic transfer of the irradiated target to the radiochemistry, there might be a failure causing the target to be blocked:

a) In the duct inside the roof of the irradiation bunker

b) In the transfer line above the roof on the way to the hotcell[5]

In case (a) the roof shielding is thick enough to guarantee in the working areas (bunker and laboratories around) a dose rate value as low as affordable for a maintenance aimed at the target recovery (about 10 μSv/h one week after the EOB, with the target caught at approximately half of the roof).
In case (b) it is needed to shield the target locally, probably with pre-assembled lead mobile shields

5.8.2 Radioactive air accidental release

It has been seen that the air coming from the irradiation bunker is highly activated, paragraph 5.5.1. Nevertheless, the irradiation bunker is not the only vault in the whole installation interested by radiation sources and ventilation system. This implies that all the air coming from the different areas are conveyed to a unique stack.
Being the irradiation bunker, the vault with highest radioactivity concentration, the common air removal process helps to reduce the average radioactivity concentration at the stack, resulting in a value consistent with the legal limits.
Suppose that for a failure of the system the air coming from the bunker RI#3 is directly removed, without being mixed with air from the other vaults. If the time required for the air to reach the stack is not taken into account, the radioactivity concentration immediately released to the environment is about 1500 Bq/g that is 1,8 MBq/m^3 and 0,2 MBq.s^{-1} if we consider a ventilation rate of 400 m^3/h. The detector used in the SPES installation to monitor the air concentration at the stack, is a spectrometric system that continuously samples the air and gives an interlock signal when the threshold of 1 Bq/g is reached. The measuring time can be tuned but obviously the longer the measure lasts, the more accurate the indication on the radioactivity released is. A reasonable time has been found to be 5 minutes.
In the examined case, the worst scenario is that the release of 0,2 MBq.s^{-1} lasts 5 minutes, time after which the system breeds an interlock and stop the beam delivery by the cyclotron.
The potential radioactivity concentration at a certain distance by the stack is obtained in the following.
At the downwind distance of X meters from the stack, the transverse area of the radioactive plume is S

$$S = \pi C_y C_z X^{2-n} \quad m^2 \qquad (1)$$

[5] Since the system is not yet defined in detail, the one presented here is just a hypothesis of how the transfer line could be built.

With C_y and C_z diffusion constants on the y and z plans, and n is an index used to take the turbulence into account.
It has been shown that, under widely different atmospheric conditions, the combination of these parameters gives a transverse section of the plume - at about 30 m downwind - practically independent from the atmospheric condition, with an average radius of the plume of about 5.7 m. Hence, as a first approximation, the average plume radius R, at a distance X meters downwind can be represented by

$$R = 5.7 / 30\, X = 0.19\, X \quad \text{meters} \tag{2}$$

For a release of air activity at a rate Q Bq/s when the wind speed is u m/s, the average activity concentration in the plume will be q Bq/m^3 obtained from

$$q = Q / \pi\, R^2\, u \quad \text{Bq/m}^3 \tag{3}$$

However, near the groundlevel the plume is assumed to have a semicircular cross section where activity that touches the ground is reflected back into the plume, making the concentration twice that given above. It is also assumed that the activity is uniformly distributed over the plume cross section. Substituting for R from equation (2), the activity concentration at X meters downwind from a release of Q Bq/s with the wind speed u m/s becomes

$$q = 18\, Q / u\, X^2 \qquad \text{Bq/m}^3 \tag{4}$$

At large distances and low wind speeds the activity will significantly decay in transit. As an example, it is reported the concentration of radioactivity in the plume downwind from the point of release as a function of the distance from the source for different wind speeds,
Figure 35.

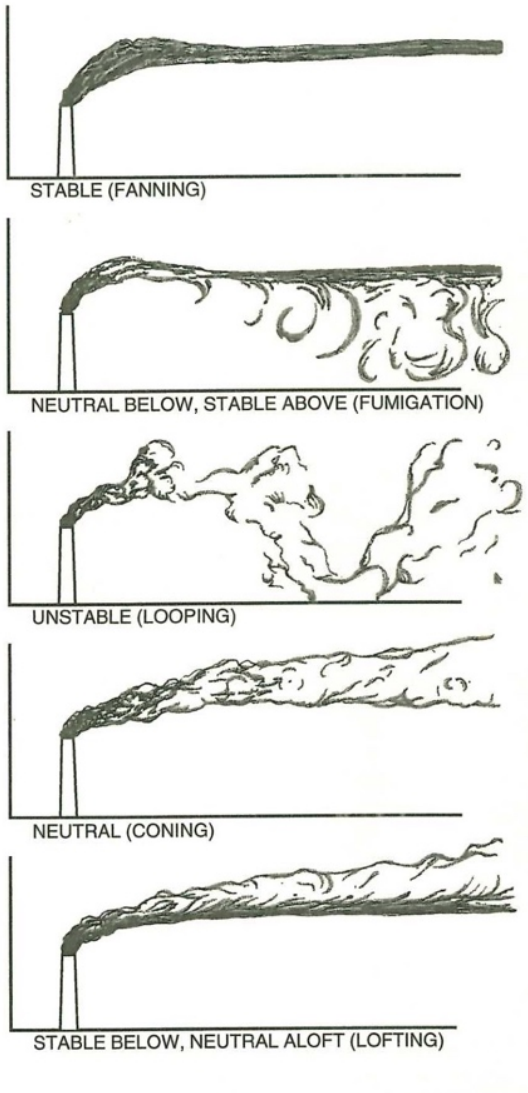


Figure 34 The effect of atmospheric stability effect on plumes, from [43].

Table 14 Dose rates from large volumes of activated air and water

Position	uSv/h per MBq m^{-3}	
	Beta	Gamma
Semi-infinite cloud	100	270
Water surface	0.13	0.35

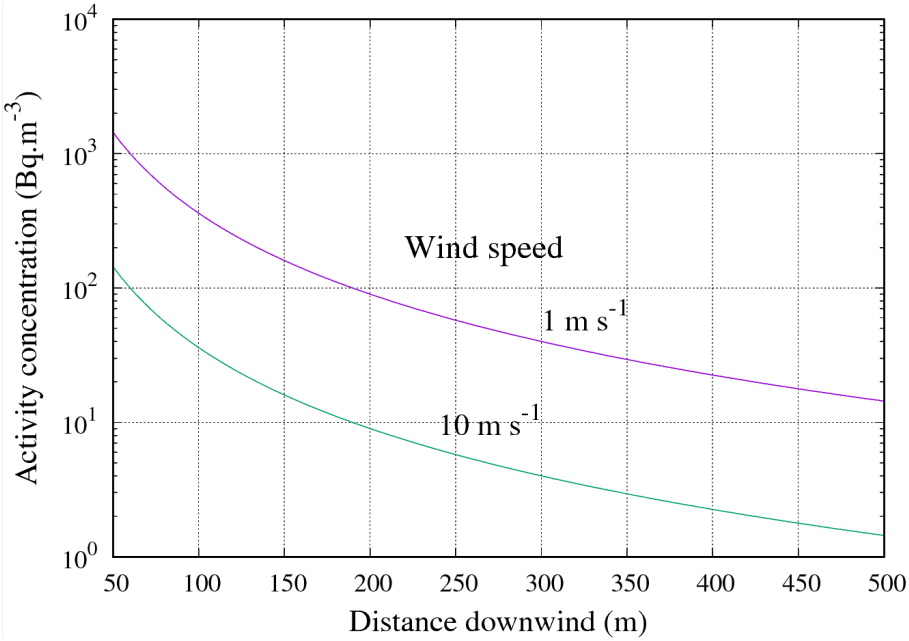


Figure 35 Radioactivity concentration in a radioactive plume as a function of distance downwind from a release at a rate of 0.2 MBq.s^{-1}.

Using the dose rate conversion factors given in Table 14, the beta dose rate in the plume at a distance X meters downwind from a release of Q Bq/s when the wind speed is u m/s becomes

$$D_\beta = 100\ q = 1.8\ Q\ /\ u\ X^2 \qquad \text{nSv/h} \qquad (5)$$

And the gamma dose rate, where the plume radius is given by equation (2) approximates to

$$D_\gamma = 1.2\ R\ q = 2.0\ Q\ /\ u\ X \qquad \text{pSv/h} \qquad (6)$$

In the present case, with an emission rate of Q = 0.2 MBq.s^{-1}, with a wind speed of u = 1 m/s and at the distance X = 200 m (distance where the first houses can be found around the laboratory) the obtained dose values are

D_β = 9 nSv/h
D_γ = 2 nSv/h

The dose rates calculated above are those in an average plume of radioactive air and would represent the possible instantaneous values that occur during the release. In the case where wind frequency and speed is known as a function of direction, equation (6), adopting suitable activity decay correction, could be used to estimate the integrated gamma dose rate in different direction and at different distances downwind from the point of release. However, as the concentration of the radioactivity in the plume is inversely proportional to the wind speed, it will be in calm conditions that the dose rates will be highest and at low wind speeds the wind direction tends towards being random.
Assuming all wind directions are equally likely, then at a distance X meters from the source, the released activity could be considered to pass through a circular plane of length $2\pi X$ and height $\sqrt{\pi R}$ with velocity u. The long-term average radioactivity concentration at X meters from a release of Q Bq/s then becomes

$$q = Q / 11\, X\, R\, u \qquad \text{Bq/m}^3 \qquad (7)$$

where R is the plume radius in meters and u the wind speed in m/s. The resulting gamma dose rate corresponding to this concentration will be given by equation

$$D = 3.2\, R \qquad \text{uSv/h per MBq m}^{-3} \qquad (8)$$

Then the long-term integrated gamma dose D(tot), per GBq of activity released is

$$D\,(\text{tot}) = 0.11/\, u\, X \qquad \text{uSv/GBq} \qquad (9)$$

With the obtained results, considering a total released radioactivity of about 0.06 GBq (release of 0.2 MBq/s lasting 5 minutes) the total dose to the population at 200 m distance is 0.03 nSv. If this type of failure occurs 10 times in one year, the annual dose rate would be 0.3 nSv/y, that is neglibile from a radiologic point of view.
If the same model is applied to the chronic release of 1 Bq/g during the whole working year, (suppose 10^4 hours/year of operation), the total annual release would be about 5 GBq and the long-term integrated gamma dose about 2.5 nSv/h.
It is underlined that the wind speed has been assumed, prudentially, equal to 1 m/s so that the integrated dose is overestimated. Furthermore, a correction factor must be introduced to take into account the radioactive decay in transit.

5.8.3 Burning of the irradiated target

It has been calculated the effective dose in case of fire of a rubidium chloride target, one week after the end of the irradiation.
This scenario is considered to be realistic only in case of a failure of the fire detection and fire extinguishing system in the laboratory and in the hot cell.

The total effective dose has been obtained using the HotSpot code, taking into account the external exposure in presence of the plume, the inhalation of radioactive species due to the plume submersion and the introduction after radioactivity resuspension (resuspension factor calculated according to the Maxwell-Anspaugh model).
The selected weather condition was low wind (1m/s speed at 10 meters height and 0.59 m/s at the emission point, 4 meters height) from north, corresponding to the stability Pasquill classes.
The dose distribution as a function of the distance from the emission point has been represented in
Figure 36.
The red curve represents the worst case and shows that the maximum dose release, corresponding to about 20 mSv, falls 100 meters distant from the emission point. In the particular case of the SPES building, this distance is outside from the laboratory perimeter, but still far from the first houses.

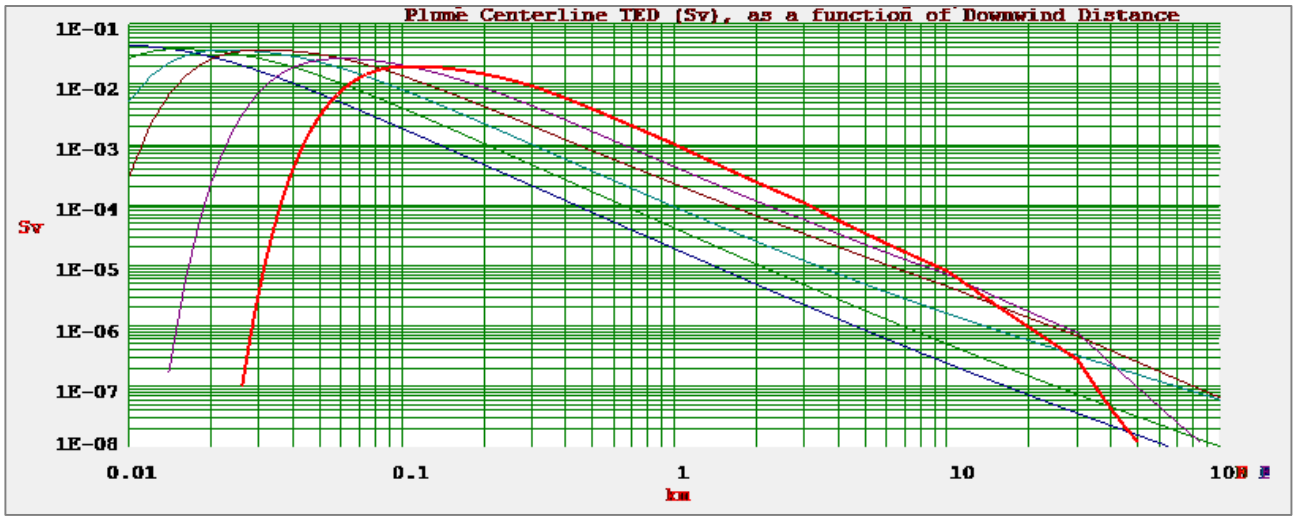


Figure 36 Effective dose distribution as a function of the distance from the point of emission in case of fire of the RbCl target, according to different atmospheric stability classes.

6. Regulatory Basis for Radiation Protection

6.1 GENERAL REGULATORY REGIME

In Italy, the regulatory regime for nuclear activities is based largely on the following legislative instruments:

- Framework Act on the Peaceful Uses of Nuclear Energy (No. 1860 of 31 December 1962) introduces a general regime based on a series of procedural requirements such as notifications and licenses.
- Legislative Decree No. 230 of 17 March 1995 related to the safety of nuclear installations and the protection of workers and the general public against the hazards of ionizing radiation arising from the peaceful uses of nuclear energy. This decree provided, inter alia, for the implementation of existing Euratom Directives on radiation protection.
- Legislative Decree No. 241 of 26 May 2000 which amends and completes the previous decree, taking into account the provisions of Council Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation, in particular with regard to natural sources of ionizing radiation, interventions and possible exposure.
- Legislative Decree No. 187 of 26 May 2000, which implements Council Directive 97/43/Euratom of 30 June 1997 on health protection of individuals against the dangers of ionizing radiation in relation to medical exposure, and repealing Directive 84/466/Euratom.
- Articles 25, 26 and 29 of Law No. 99 of 23 July 2009 include enabling provisions empowering the Government to issue one or more implementing decrees providing rules for the siting of new nuclear power plants, the licensing process for the construction, operation and dismantling of those plants, as well as rules for interim storage and the final disposal of nuclear waste.

These instruments, in addition to other acts, decree and regulations which do not concern nuclear activities exclusively, constitute the framework for nuclear legislation in Italy [44].

6.2 RADIATION PROTECTION

Legislative Decree on the safety of nuclear installations and the protection of workers and the general public against the hazard of ionizing radiation (No. 230/95), as completed by the Decree No. 241/00, is the basic text in Italy governing radiation protection. In furtherance of the general framework established by Act on on the Peaceful Uses of Nuclear Energy, the decree also introduces a regime for supervising the safe conduct on nuclear activities and, in general, all activities involving the use of ionizing radiation. This regime will be supplemented by the adoption of a number of implementing decrees, in the same manner as those made in implementation of Presidential Decree No. 185/64. Some examples shall be mentioned. A Decree of the Prime Minister is to prescribe the numerical values and other conditions which determine the scope of application of Decree No.230/95. This implementing decree will be made on the proposal of the Ministries for the Environment and Health, in conjunction with the other ministries and following consultations with the ISPRA and the other organizations concerned. Provisionally, these values and conditions are established in Annex1 to Decree No.230/95, as amended by Annex1 to Decree No. 241/00. Another Prime Ministerial Decree (made under a similar procedure) is to establish the maximum dose limits for workers and the public. These dose limits are established, in the meantime, by Annexes III and IV to Decree No. 230/95, as amended by Annex 1 to Decree No. 241/00.

6.2.1 Protection of workers

As provided by Decree No. 230/95, the responsibility for the radiation protection of workers lies with the Ministry for Labour and Social Security (acting through the Labour Inspectorate) according to Section 59, the local authorities of the National Health Service and the ISPRA.

Those entities, including the state, the regions, the provinces, the communes, public bodies, educational establishments and research laboratories, which expose workers to the hazards arising from ionizing radiation in the course of their work, must comply with the provisions of Decree No. 230/95. The general rules for the radiation protection of workers, like Presidential Decree No.185/64, are based upon the Euratom basic standards. This source is expressly cited in particular in the provisions for the adoption of dose limits (Section 96) mentioned above. Decree No.230/95 also regulates work in mines where radioactive substances are present and, in particular, defines the obligations of employers in relation to the radiation protection of workers in the mining industry. The conditions governing the application of these provisions are set out in a Decree of the Minister for Industry, who is also responsible for inspection activities to ensure protection against ionizing radiation risks. In particular, specific provisions have been introduced as regards the protection of outside workers covered by Council Directive90/641/Euratom of 4 December 1990 on the operational protection of outside workers exposed to the risk of ionising radiation during their activities in controlled areas. This involves, inter alia, the

establishment for each worker of a personal radiation logbook with which he must be provided in compliance with the above Directive.
Finally, another very important innovation is the inclusion of provisions allowing the ALARA principle to be applied to the employer's activities. Besides incorporating the latest Euratom Basic Standards, Decree No.241/00 also regulates the protection of workers exposed to natural sources of ionizing radiation (Sections 10 bis et seq) as well as crew members of high altitude flights.

6.2.2 Protection of the public

Decree No. 230/95 also contains provisions on the protection of the public against ionizing radiation. The Ministry for Health is responsible for such protection and must, in particular, through its National Health Service, control all sources of ionizing radiation to avoid any contamination of the public and of the general environment (Section 97). Regional and provincial commissions have been set up to give advice on radiation protection and related problems.
Dose limits and maximum permissible concentrations for the public are established in accordance with Section 96 of Decree No. 230/95, in compliance with applicable Community Directives. Provisions of Decree No. 187/00 concern the protection of persons during medical exposure in compliance with Council Directive 43/97/Euratom of 3 June 1997 on health protection of individuals against the dangers of ionizing radiation in relation to medical exposure and repealing Directive 84/466/Euratom. These provisions repeal those provisions in Decree No.230/95 (Sections 109 to 114) which implemented Directive 84/466/Euratom. The Ministry for Health must adopt the necessary measures to give full effect to Decree No. 187/00. Such measures will cover the conditions governing training of staff, the criteria for approval of radiological equipment, justification of certain exposures etc.

6.2.3 Protection of the environment

The most important provisions of Decree No. 230/95 relating expressly to the environment are Sections 100 and 104. Decree No. 241/00 also requires that an optimum level of environmental protection be maintained thereby ensuring that the exposure limits set out in the Euratom Basic Standards are observed.
In the event of an accident during an operation which involves radioactive substances, if the environment is effected, the operator must intervene to prevent the risk of subsequent Nuclear Legislation in OECD Countries contamination or injury to persons (Section 100 of Decree No. 230/95). The Prefect of the Province and the local authorities of the National Health Service must be informed immediately.
Whereas Section 104 provides that the Ministry for the Environment is responsible for monitoring ambient radioactivity, the monitoring of foodstuffs and beverages is entrusted to the Ministry for Health with overall technical co-ordination being ensured by the ISPRA (see infra, Part II "Institutional Framework"). All monitoring is carried out by

national and regional networks, the latter acting under directives issued by the above-mentioned ministries.
The activities of the ISPRA are also governed by relevant directives from these ministries, and by Section 35 of the Euratom Treaty. Its functions include the co-ordination and standardization of measurements, the introduction of new measuring stations etc.
The situation described above is one result of the referendum held on 18 April 1993 which abolished the powers of Local Health Units as regards the environment, entrusting these powers to the ISPRA and to other relevant regional and provincial departments and organizations concerned.

6.3 RADIOACTIVE WASTE MANAGEMENT

Decree No. 230/95 implemented Council Directive 92/3/Euratom concerning transfers of radioactive waste. Circular No. 236 of the Ministry for Economic Development of 28 October 1994, adopted in order to implement this directive into Italian legislation pending Decree No. 230/95, was basically transcribed into this decree. Pursuant to that directive, a prior license is required for transfer, import, export and transit of radioactive waste (Section 32). The procedure governing such licenses is laid down in a decree of the Minister for Economic Development. At the international level, Italy ratified the 1972 London Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter on 30 April 1984.
When the radioactive waste meets specific requirements, the Decree No. 230/95 is not applied. In particular, according to section 30 and 154, those waste or materials to be reuse/recycle/dispose of, containing radionuclides with half-life shorter than 75 days and in concentration lower than 1 Bq per gram.

6.4 THE AUTHORIZATION FRAMEWORK

According to section 29 of the Decree 230/95 and subsequent modifications, each activity entailing a risk of exposure to ionizing radiation, must be prior authorized according to the amount of radioactivity or to the characteristics of the radiation source involved. The authorization can be A or B category. For both of them the authorization is granted taking into account the following aspects:

- the location of the installation
- the appropriateness of: spaces, radiation protection structures, uses of the source, the equipment and personnel competence
- consequences of possible accidents
- procedures for the removal or disposal of radioactive wastes to the environment.

Category A or B are specifically requested according to the amount of radioactivity handled (different limits for sealed and unsealed sources), to the type of equipment producing ionizing radiation and to the type and intensity of the generated radiation field (the limits are explicitly described in Annex IX).

Category A authorization is granted by the Minister for Economic Development in agreement with the Minister for the Interior and Ministers for Labour, Health and the Environment taking the remarks of ISPRA and of the involved regions into account.
Category B license is released by the prefect of the province in agreement with other competent technical authorities.
For the applications described in this thesis a license of Category A is requested. In order to obtain a license of this type, the description of the activity to be licensed must be presented together with a technical report edited by the Qualified Expert (QE). This report will include:

a) the description of both the spaces interested by the activity object of the license, and the neighboring areas, together with the classification of the areas according to section 82;
b) criteria followed by the QE to classify both the areas and the workers operating in there
c) operations to be done, equipment and radiation sources to be used in one place or to be moved elsewhere in the area
d) identification of potential exposure and actions to prevent exposure and confine the consequences on workers and general public
e) radioactive waste production and management (it also includes possible reuses or recycling).
f) Civil works required for the activity and commissioning tests
g) Actions in view of the decommissioning
h) Evaluation of the dose to the workers and to the reference group during normal operations
i) Potential exposure: evaluation of the spread of contamination in time and space
l) Physical monitoring of radiation protection (dose evaluation, environmental monitoring, whatever in the definitions of duties of the QE)
m) worker's safety and protection; training program and professional qualification of the workers
n) in case of medical application, the contribution to the environmental burden by the radioactivity administered to the patient for diagnosis or therapy.

6.5 Classification of radiation areas

A classified radiation area is a delimited space where the access is ruled according to radiation protection criteria. Two types of classification exist: classified and supervised area, they will be defined in the following paragraphs.
Normally there are procedures defined by the QE to rule the exit of personnel and materials from classified radiation areas, due to the risk of contamination spread.
There are also procedures to be followed to access radiation areas. In Table 17 and Table 18 a description is given of the situations that can be found in a radiation environment

(during normal operation or in case of emergency) and the safeguards to take to safely access those areas.

6.5.1 Controlled Radiation Area

This is an area where the doses received by the workers may exceed $3/10^{ths}$ of the annual dose limit established for exposed workers (ref. Table 15).
Entrances to a controlled area bear a yellow warning sign with the words "Controlled Radiation Area: No Entry to Unauthorised Personnel", and the standard black and yellow trefoil sign denoting a radiation hazard.
Moreover, the access in a controlled radiation area is ruled by procedures specified by the QE and transmitted to the employer.

6.5.2 Supervised Radiation Area

Those places where there is the risk for a worker to receive a dose higher than the limits established for general public (ref. Table 15) but are not appointed as classified areas, are supervised areas. They are demarcated by a yellow sign with the words "Supervised Radiation Area".

6.6 CLASSIFICATION OF RADIATION WORKER

In general, workers dealing with a risk of exposure to ionizing radiation in their working life can be either considered as non-exposed workers or classified as exposed workers.
It must be bear in mind that a non-exposed worker can still perform activities entailing a risk of exposure to ionizing radiation, but the dose received must be below the established limits for the general public, expressed in Table 15.
Those classified as exposed workers should receive an annual dose not exceeding the limits expressed in Table 15.
Radiation exposed workers are further defined as category A or B. Category A workers are those individuals whose effective dose might exceed 6 mSv in one year or the equivalent dose for the lens of eye, skin, hands, forearms, feet and ankles might overcome $3/10^{ths}$ of established limits. Exposed workers who are not classified as category A are category B.
Two details are noteworthy about the radiation worker classification: first, the classification is done by the QE following the job description provided by the employer. Second, both categories share the same annual limits and the introduction of the "$3/10^{ths}$" criterion is used to identify those exposures needing further investigation.
Table 16 is an overview of the significance of the exposure levels in terms of medical inspection needs. It is useful not only to have a reference of the exposure limits, but mainly to understand what is the average professional exposure during one's working life.

Table 15 Dose limits established for general public and for exposed workers according to the Decree 230/95 and subsequent modifications.

	General public	Exposed Workers
	(mSv/year)	
Effective dose	1	20
Lens of the eye (equivalent dose)	15	150
Skin, average on 1 cm^2 whichever exposed surface (equivalent dose)	50	500
Hands, forearms, feet, ankles (equivalent dose)	–	500

Table 16 Possible guidelines to the significance of exposure to radiation. (Sullivan)

Exposure	Significance
3.5 Sv	50% chance of survival
> 1 Sv	Serious to lethal
> 50 mSv	Requiring medical checks
50 mSv. y^{-1}	Occupational Dose Limit
15-50 mSv. y^{-1}	Strict dose control necessary
5-15 mSv. y^{-1}	Professional Exposure
< 5 mSv. y^{-1}	Minimum control necessary
1 mSv. y^{-1}	Natural background
10 uSv. y^{-1}	Insignificant

6.6.1 Radiation Protection constraints

The physical surveillance of the workers and the population is done taking into account the limits explained in Table 15. Anyway, it is advised to set radiation protection constraints, lower than the dose limits, to be read as project objectives in order to comply with the safety standards and maintain a safety margin.

The evaluations of the present work take into account work/public dose constraints and radioactivity of the equipment/instruments of the installation and of the environmental matrices.

Namely, the following constraints have been set:

- Public and non-exposed workers: 0.5 mSv per year.
- Exposed workers (both category A and B): 5 mSv per year.
- Radioactivity concentration in environmental matrices: 1 Bq per gram, or total radioactivity of the considered radionuclide less than the values of table I-1 of the cited decree.
- Radioactivity concentration in equipment/instrumentation: 1 Bq per gram, or total radioactivity of the considered radionuclide less than the values of table I-1 of the cited decree.

The materials to be disposed of will be subject to the procedures actually adopted at the LNL, at the licensed accelerators: once identified, measured and labelled the materials will be characterized by the radiation protection service with gamma spectrometry. In particular, the small samples and the small-size pieces will be analysed in laboratory while the equipment, activated parts of the beam line or, in general, big-size instrumentation will be analyzed directly on the field with a portable spectrometric system.

The access criteria of Table 17 and Table 18 have been set to comply with the dose limits. In particular, a gamma dose rate threshold corresponding to 3 μSv/h has been set. Below this threshold, a radiation-exposed worker could, hypothetically, work for the entire working year (approximately 1690 hours per year) without exceeding the yearly dose constraints. For this reason, above the threshold any intervention must be previously authorized and the duration should be limited (procedures are set case-by-case). The nature of these table is, clearly operative, in order to give an indication to workers and activity responsible of the attention to be paid to the dose rates involved during operations and intervention.

The public reference group for the dose evaluations are identified as the people working in the university fields by the laboratory perimeter (10 meters, south direction, as shown in Figure 37). For accidental release of radioactivity, it has been considered that the closest inhabitants are at 200 meters distance from the SPES building.

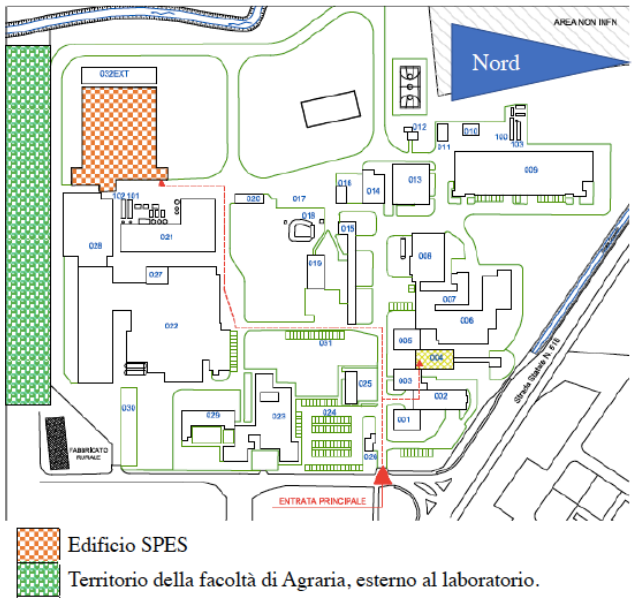


Figure 37 Laboratory layout. The SPES building highlighted in red and the reference group site (external to the LNL) in green, direction south.

Table 17 Access criteria in controlled area as a function of the neutron dose rate.

Neutron dose rate, R_n [μSv/h]	Authorized personnel	Nature and frequency of the operation	Notes
< 0,01	Personnel previously authorized to access the areas at the level -1	Unrestricted	Measure gamma dose rate and beta dose rate in case of *hands on* intervention
0,01 < R_n < 3	Radiation exposed workers	Urgent/important	EQ authorization required. Personal dosimeters to be worn.

Table 18 Access criteria in controlled area as a function of the gamma dose rate.

Gamma dose rate, R_γ [μSv/h]	Authorized personnel	Nature and frequency of the operation	Notes

< 0,10	Personnel previously authorized to access the areas at the level -1	unrestricted	Measure beta dose rate in case of *hands on* intervention on the beamline and the cyclotron
0,1 < Rγ < 3	Radiation workers	Necessary operation (first operation phases), routine operations at regime	EQ authorization required during the first operation phase (beamline commissioning, maintenance scheduling). Personal dosimeters to be worn. Measure beta dose rate in case of *hands on* intervention and wear finger dosimeters to evaluate equivalent dose to the hands. During normal operations the authorization of the EQ is no more required.
3 < Rγ < 10	Radiation worker	Limited duration (maximum 2 hours)	EQ authorization required during the first operation phase (beamline commissioning, maintenance scheduling). Personal dosimeters to be worn. Measure beta dose rate in case of *hands on* intervention and wear finger dosimeters to evaluate equivalent dose to the hands.
10 < Rγ < 100	Radiation exposed worker	Urgent/important intervention, limited duration (max 1 hour). Frequency less than once per month.	EQ authorization required.. Personal dosimeters to be worn (same as the above line)
100 < Rγ < 500	Radiation exposed workers	Urgent/needed intervention to contain the radioactivity spread and protect equipment/instrumentation and structures. Frequency less than 3 times per year	Opportunity evaluation of the intervention by the installation responsible[6] and authorization of the EQ.

[6] The responsibility chain expressed in this table may vary according to the installation policy, but it is essential to establish it clearly before any operation starts.

$R\gamma > 500$	Radiation exposed workers, category A	Essential intervention to preserve the istallation. Not more than once per year..	Opportunity evaluation of the intervention by the installation responsible[7] and authorization of the EQ

[7] The responsibility chain expressed in this table may vary according to the installation policy, but it is essential to establish it clearly before any operation starts.

7. Measurements and validation during the cyclotron commissioning

The 70 MeV proton cyclotron of the LNL-INFN is already installed and the commissioning tests started in May 2016.
After the first delivered/extracted beam at low current, a measurement campaign has been conducted, both on field and with samples collected from cooling effluents, dust and miscellaneous beam line equipment.
In this chapter, the results of the measures will be shown, together with the related instrumentation involved, the methods followed and the comparison with the calculated values.
The first tests have been led to prove the ability of the machine to accelerate and extract a proton beam with maximum 70 MeV energy and 700 μA current on two separate extraction ports.
The tests done so far include:

- Injection of the H^- beam and acceleration without extraction at maximum energy and current up to 3 μA
- Extraction of the proton beam with maximum current of 3 μA on silver and copper faraday cups inside the cyclotron vault
- Extraction of the proton beam with maximum current 500 μA on the beam dump (copper thick target shielded with polyethylene and lead) in the irradiation bunker.

The radiological survey has been organized in such a way to exclude any radiation dose outside of the building (through passive dosimeters placed on the external perimeter of the building), to avoid unforeseen exposure of the personnel accessing the controlled areas or staying in the control room (through ionization chambers and rem-counters), to keep the activation of the environmental matrices and accelerator structures under control, thus avoiding any spread of contamination.
In the following paragraphs, it will be given an overview of the instrumentation actually on site, the measurements done during the first tests and, where possible, the comparison of the measurements with the expected values calculated with the FLUKA code.

7.1 INSTRUMENTATION

The external perimeter of the building has been monitored through track-etch detectors (CR-39) for neutron dosimetry [45] and TLD for gamma dosimetry. Passive dosimeters have also been placed inside the building, in critical points corresponding to shielding penetrations and ventilation ducts coming from controlled areas.
Inside the building, mobile monitoring stations have been placed: they include ionization chambers for gamma dose rate (measuring range: 100 nSv/h– 1 Sv/h, energy range: 30 keV – 7 MeV) and extended range rem counters for neutron dose rate measurements (measuring range 1 nSv/h – 100 mSv/h, energy range 25 meV – 5 GeV). Additional, extended range, Geiger-Mueller counters have been placed inside the cyclotron vault and in the beam dump bunker. The distribution of the described equipment is shown in Figure 38.
The radioactivity concentration in air is continuously measured via a spectrometric system, specifically designed to have a sensitivity of 0.1 Bq/g. Two separate systems are used to monitor the radioactivity concentration inside the irradiation cave and at the stack.

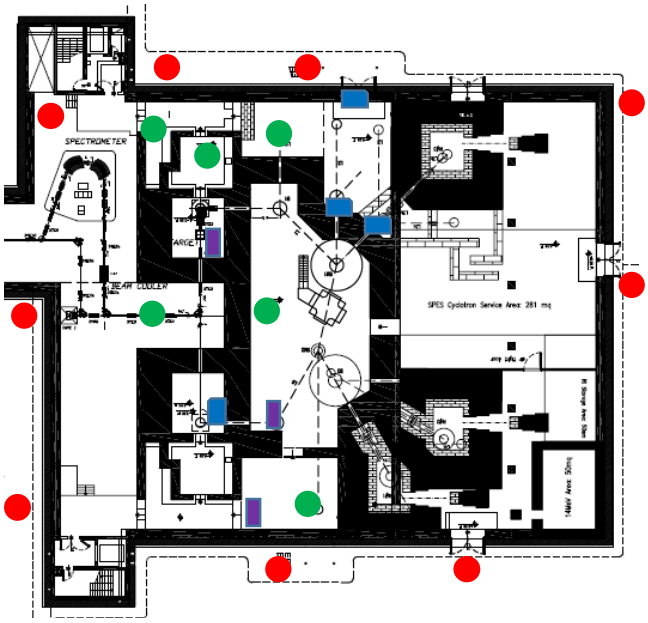


Figure 38 Environmental monitoring plan: neutron dosimeters around the external perimeter (red circles), neutron-gamma dosimeters (blue rectangles), n-gamma mobile stations (green circles) and extended range GM (purple rectangles).

7.2 MEASUREMENTS

7.2.1 Environmental survey

During the low current tests, the beam has been sent on two Faraday cups in silver and copper, with maximum beam power of 210 Watt (70 MeV energy and 3 μA current). The indication of the rem counter for the neutron ambient equivalent dose rate (AEDR) at the reference point, with beam on the Ag faraday cup, was 465 mSv/h/μA, the calculated value was approximately 500 mSv/h/μA (see **Errore. L'origine riferimento non è stata trovata.**39, at coordinate -15 m on x-axes and 7.5 m on y-axes, each axes minor tic is 2.5 meters, corresponds to the yellow-orange value on the scale).
During the irradiation of the copper faraday cup, the neutron dose rate at the measurement point, as indicated in Figure 40, was between 200 and 250 mSv/h/μA (approximate coordinates -15 m on x-axes and 17.5 m on y-axes, on each axis minor tic is 2.5 meters). This indication can be used as a reference during the irradiation of the beam dump, or in future operations with the SPES target. Since the shielding between the bunker of the beam dump and the cyclotron vault is thick enough to shield the neutron dose coming from the beam dump irradiation, it comes out that the dose rate in A1 is exclusively due to the beam losses along the beam line. The neutron dose rate in A1 proves useful to indicate whether the beam losses are close to nominal value or out of range.

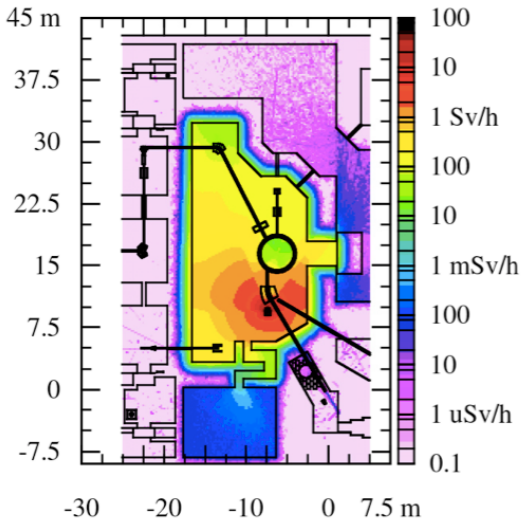


Figure 39 Neutron ambient equivalent dose rate during the irradiation of the silver faraday cup with 3 μA, 70 MeV protons.

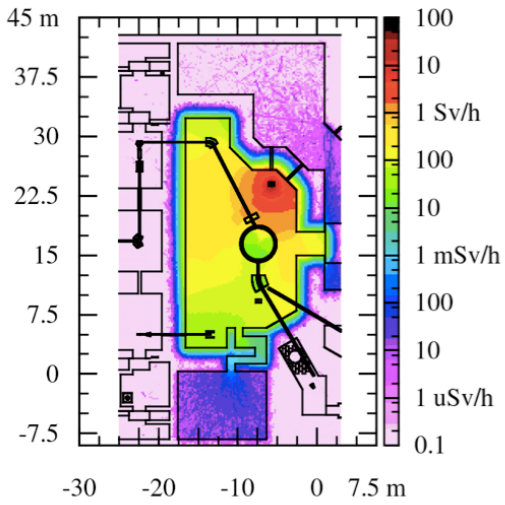


Figure 40 Neutron ambient equivalent dose rate during the irradiation of the copper faraday cup with 1 μA, 70 MeV protons.

Inside the bunker housing the beam dump the expected neutron AEDR is close to 10 Gy/h, a prohibited environment for neutron active measurements through, for instance, rem counters. The only instrument placed in the vicinity of the beam dump is an extended GM counter. The measured gamma AEDR during the irradiation of the beam dump is 160 mSv/h per 100 μA proton current at 70 MeV energy. This is confirmed by calculations (Figure 41, yellow-green spot at coordinates -20 x-axes, 24 y-axes).

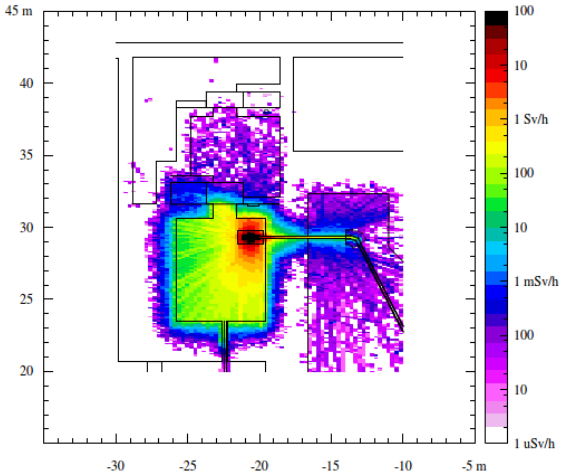


Figure 41 Gamma ambient equivalent dose rate in the bunker, during the irradiation of the copper beam dump with protons of 70 MeV energy and 100 μA current.

Unfortunately, in high neutron dose rate environments even the high range GM had
During the irradiation, all the critical points outside of the shielding were constantly monitored and – except for the accelerator cave, the irradiation bunker and respective pre-bunkers – nowhere the n-gamma AEDR exceeded the background.

7.2.2 Activation monitoring

The total irradiation period of the tests done so far is about 30 hours, so the build-up of long half life species is not enough to detect those elements through a gamma spectrometry. Nevertheless, some short-medium half-life elements have been found in the targets directly irradiated (Ag and Cu faraday cups) and in matrices activated by secondary radiations.
The direct irradiation of the Ag faraday cup with 3 μA proton current and 70 MeV proton beam energy, lasted approximately 2 hours. It was possible to identify some nuclides through gamma spectrometry, using a portable detector (Table 19).

Table 19 Nuclide identification from the gamma spectrometry of the Ag faraday cup.

Nuclide	Activity (Bq/unit)
Na-22	$2.03\ 10^{5}$
K-40	$1.37\ 10^{7}$
Mn-54	$2.02\ 10^{5}$
Co-58	$6.96\ 10^{5}$
Zn-65	$1.94\ 10^{6}$
Y-92	$1.94\ 10^{6}$
Zr-97	$1.05\ 10^{6}$
Rh-105	$1.07\ 10^{7}$
Cd-109	$1.09\ 10^{10}$
Sn-113	$2.88\ 10^{6}$
Sb-122	$1.66\ 10^{6}$
Te-132	$6.05\ 10^{5}$

The irradiation of the beam dump (thick copper target shielded by 35 cm polyethylene and 5 cm lead) with protons of 70 MeV energy, current in the range 100 to 500 μA, lasted non-more than 30 hours in total. The residual ambient equivalent gamma dose rate due to copper activation was about 150 μSv/h on the external shielding surface still 2 weeks after the end of irradiation.
The high neutron fluxes during the beam dump irradiation, caused the air inside the bunker to be activated, as well. The measures where done sampling the air directly inside the bunker and at the stack, where the air coming from the ventilation system of the facility as a whole, is sent: with a ventilation rate of approximately 400 m^3/h to keep the bunker under pressure, at the maximum power tested (35 kW) the radioactivity

concentration in air was 200 Bq/g, while with FLUKA calculations the expected value was approximately 500 Bq/g. In the same condition the radioactivity at the stack was below 1 Bq/g, with a total air flow of 40000 m^3/h and a path length from the bunker to the stack not less than 30 m, the readings have been shown in Figure 42. On the top figure, the detector reading of the air at the extraction is shown, with an indication of the reference concentration of 1 Bq/g (concentration limit for unauthorized release to the environment of short[8] half-life nuclides). The limit value has never been reached during the tests.
On the bottom figure, the blue indicators represent the radioactivity concentration of Ar-41, while the green are representative of the beta+ emitters, O-15, N-13 and C-11.

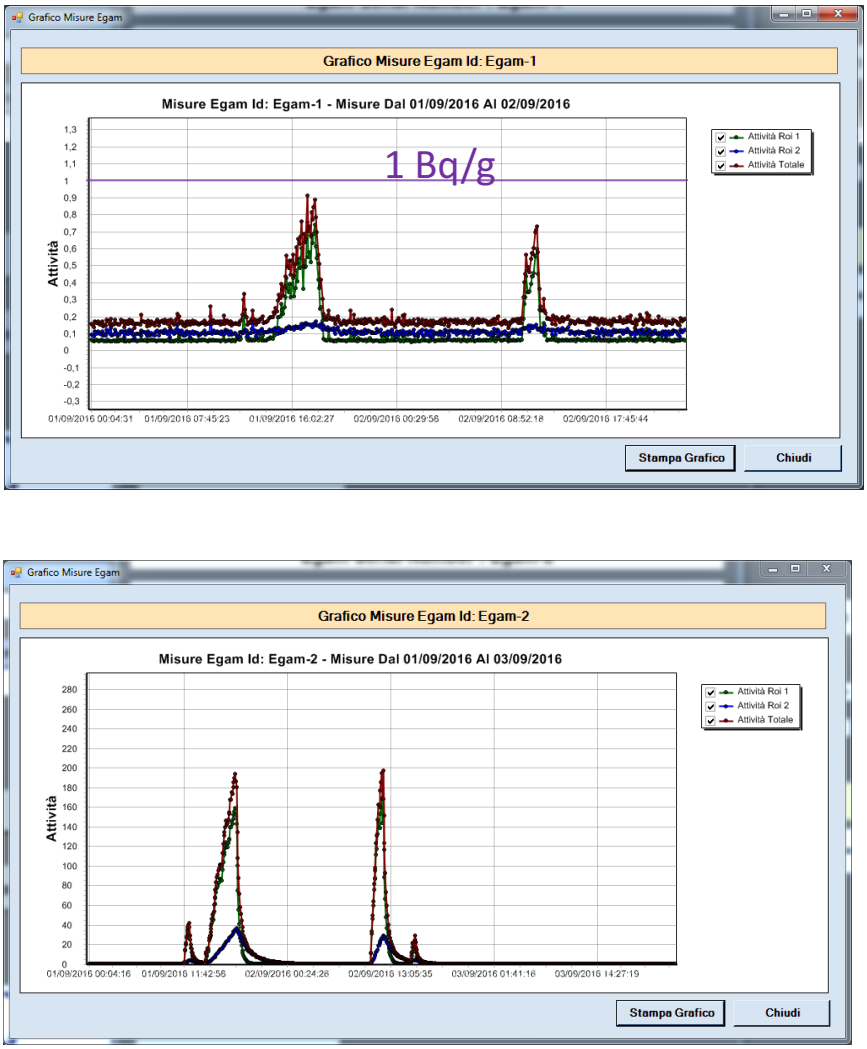


Figure 42 Radioactivity concentration in air: reading at the stack on top and inside the irradiation bunker (on bottom).

[8] Half-life shorter than 75 days.

A sample of the water in the cooling system of the cyclotron has also been analysed. The reference irradiation condition where: proton beam current 3 μA, energy 70 MeV, irradiation period 2 hours. In the calculation, it has been taken into account the total tank capacity, the water flow and the size of the pipes, to correctly evaluate water activation. While the radioactivity concentration of the beta+ emitters is consistent within a 20% error between measurements and calculation (approximately 2.5 MBq/liter calculated at EOB and 2 MBq/liter measured some minutes after the sample collection, Figure 43), the radioactivity of Be-7 measured (830 Bq/l) differs quite a lot form the calculation (50 Bq/l). Being this discrepancy still under investigation, the possible explication might be some leaching effect from copper.

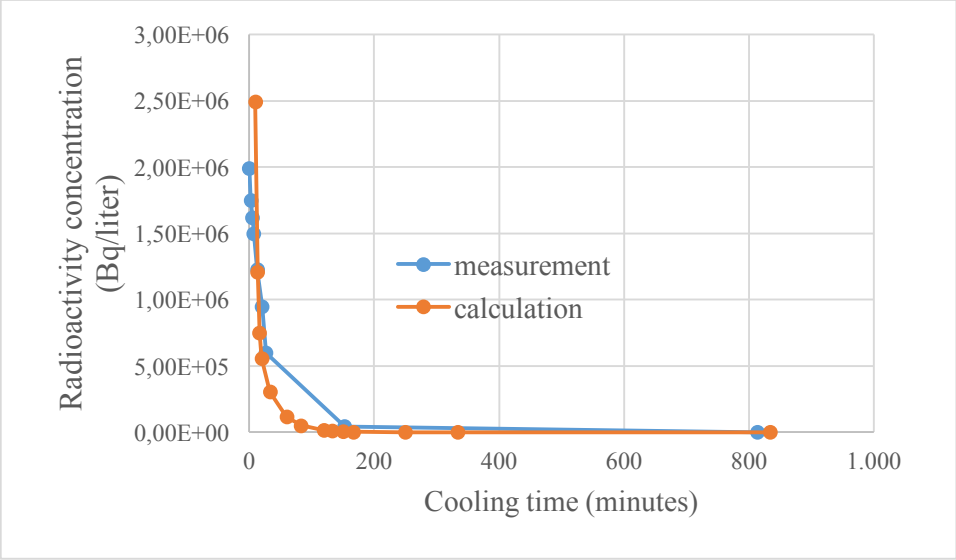


Figure 43 Comparison of the radioactivity concentration from O-15, N-13 and C-11 as calculated and as measured at different cooling times.

The water of the beam dump has not been sampled yet, but measures in contact with the tank showed a gamma AEDR of about 180 μSv/h during the irradiation at maximum power. The beam dump water tank has been temporarily confined with barriers at 2 meters distance, in future it will be shielded in order to avoid accidental overexposure of the personnel working close by. Being the irradiation period still too short, tritium has not been found but it is not excluded that, after a long run, it might be detected in the cooling water. In that case, a careful management of the water as a waste must be foreseen.
In order to evaluate possible contamination sources in future operations, a sample of the dust collected in the bunker was placed in the vicinity of the beam line and irradiated for two hours (irradiation by secondary neutrons). Despite the limited irradiation duration, 2 hours, with successive gamma spectrometry it was possible to identify the following species:

- Na-24 ($T_{1/2}$ 15 h), activity 144 Bq ± 5,
- Mn-56 ($T_{1/2}$ 2.6 h), activity 160 Bq ± 4,

- K-42 ($T_{1/2}$ 12.3 h), activity 16 Bq ± 1.4.

Confirmation of the presence of those species in the activated dust can be found in [46]. This measurement suggests that future maintenance or, more generally, any work done in the irradiation bunker after an irradiation period, must be done using masks, in order to avoid any possible introduction of radioactive particles. The irradiation bunkers should, anyway, be kept as clean as possible and the surfaces be adequately painted to be smooth.

7.2.3 Personal Monitoring

At this stage, all the people authorized to access the controlled areas of the facility have been classified as category B workers. Everyone is provided with passive and active dosimeters for n-gamma radiation. Whenever « hands-on » intervention was requested, finger dosimeters were supplied. After 4 months of operations, the collective dose has been 28 μSv, considered as negligible.

8. Conclusion

The LARAMED project of the LNL is a new research infrastructure for the study of innovative radionuclides in the field of nuclear medicine. High-energy accelerated proton beams colliding onto solid targets will provide the required radioactivity to study the production reactions and to set the appropriate chemical treatment for the radiopharmaceutical preparation.

Every stage of the whole procedure, from the target irradiation to the radiopharmaceutical packaging, entails a risk of exposure to ionizing radiation. An overview of the potential sources of exposure has been given, revealing that potential harm can be received not only during high-power irradiations but also during cross section measurements (requiring low-intensity proton beams), if adequate shielding structures or radiological safeguards are not foreseen.

All the aspects concerning the dose evaluation to the workers and to the reference public around the facility have been evaluated, considering the different routes of exposure during normal operation and in case of accident.

The studies have been done with the help of Monte Carlo simulations, to evaluate the source terms and to foresee possible activation of the materials.

It has been shown that the building and the infrastructures, at the state of play of the civil works, is adequate to mitigate the high dose levels achieved during the target irradiation. The mitigation effect has been considered from two main points of view:

- the potential external exposure of the worker and the population
- the potential activation/contamination of the environmental matrices, causing radioactivity to be transferred to the food chain and thus introduced into the human body, causing internal exposure.

The results of the evaluations have shown that the dose limits are never exceeded, exception made for the accident scenario of a fire of the irradiated target. The analyzed case, anyway, was a typical example of risk analysis when no safeguard is in place: the fire extinguishing system had been excluded and it was considered to be an emergency situation for the installation as a whole.

It has been seen that in each stage of the project there is room for safety improvements: in practical terms, the shielding for the irradiation bunkers can be increased in view of possible upgrades of the cyclotron, local shielding can be provided where needed, and the containment of radioactive effluents – as, for instance, that coming from cooling water tanks – can be more effectively monitored and contained. On the other hand, the

safety can be enhanced if adequate procedures are defined and put in place: it is of extreme importance the training of the personnel accessing radiation areas, in order to perform dose rate measurements and to evaluate the opportunity of interventions.
The systems that are still at the design level, for instance the pneumatic transfer system of the target from the irradiation bunker to the radiochemistry, have been considered and possible criticalities have been highlighted.
From the radiation protection service point of view, the monitoring system has been implemented trying to provide an overview as complete as possible of the radiation levels in every area during the machine operation and at the end of irradiation, for the detection of residual dose rate spots.
The monitoring system includes instruments for the collection of data both in laboratories and on the field. In particular, a system for the remote acquisition of the hot spots coupled to a portable HPGe spectrometer can prove an exceptional investigation system to detect the sources of activation on the cyclotron and the beam lines.
The measurements done during the commissioning of the cyclotron have been compared to the simulations, where the geometry and the irradiation conditions have been implemented as close as possible to the reality.
The operational quantities observed, as indicated by the monitors installed in the vicinity of the interaction points, were consistent with the expected values calculated with Monte Carlo simulations. The radioactivity induced in the environmental matrices and in the accelerator structures, even though in limited amounts, rise the attention on some important issues concerning the management of the facility, the maintenance program and the waste treatment.
The study presented in this work, together with the benchmark done during the commissioning of the cyclotron, includes the requested data for the licensing procedure of category A, in agreement with the Decree 230/95 and modifications.
In particular, it provides all the necessary informations to guarantee that the described activity responds to the basic radiation protection principles: justification, optimization and limitation.

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