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*Published Version:*
Vichi, S., Infantino, A., Zagni, F., Cicoria, G., Braccini, S., Mostacci, D., et al. (2020). Activation studies for the decommissioning of PET cyclotron bunkers by means of Monte Carlo simulations. RADIATION PHYSICS AND CHEMISTRY, 174, 1-12 [10.1016/j.radphyschem.2020.108966].

*Availability:*
This version is available at: https://hdl.handle.net/11585/760579 since: 2024-08-27

*Published:*
DOI: http://doi.org/10.1016/j.radphyschem.2020.108966

# Activation studies for the decommissioning of PET cyclotron bunkers by means of Monte Carlo simulations

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**During the operational life of a PET cyclotron, the concrete walls of the vault are activated by secondary neutrons. For the dismantling of such accelerator facilities, a considerable amount of low level solid radioactive waste has to be characterized and disposed. To decrease future dismantling costs and complexity, the amount of radioactive waste has to been prospectively evaluated in the design phase, then confirmed at the time of planning decommissioning. In this work, the Monte Carlo code Fluka was used for the assessment of the activation of the bunkers of two different facilities: the 16.5 MeV GE PETtrace at S. Orsola-Malpighi Hospital in Bologna and the 18 MeV IBA Cyclone 18/18 HC at the Bern University Hospital (Inselspital). The simulations were validated by means of experimental measurements performed in our previous works: non-destructive, in field measurements using a portable CZT detector were performed in Bologna; while core drilling samples were extracted from the bunker and measured in laboratory with an HPGe detector in Bern. The activity of the most important radionuclides in the concrete walls of the bunker, namely Eu-152, Mn-54, Co-60, Sc-46, Zn-65 and Cs-134 resulted within the range of 0.01 – 2 Bq/g. The consistency between Monte Carlo results and experimental measurements was within a factor 2 - 3 for most radionuclides, except for Eu-152, Sc-46, Zn-65. The activity concentrations estimated at each position considered exceeds the clearance levels of the new Directive 2013/59/Euratom.**
**The results of this work demonstrate that Monte Carlo simulations based on FLUKA are adequate to assess the residual activation levels, a fundamental information to foresee, plan and optimize the decommissioning of a cyclotron based PET centers.**

**Keywords: Neutron Activation; PET Cyclotron; Decommissioning; Monte Carlo; Radiation Protection.**

## 1. Introduction

Applications of particle accelerators to medicine are continuously growing, especially in cancer diagnosis and treatment. In particular, cyclotrons for the production of radioisotopes for Positron Emission Tomography (PET) are nowadays common in hospitals and more than 1200 PET cyclotrons are currently estimated to be in operation worldwide [1]. During irradiation with proton beams, the production of secondary neutrons leads to the activation of the components of the cyclotron itself and of the surrounding building materials. The number of installed cyclotrons increased essentially in the last 20 years. Nowadays, a significant number of the older systems, installed at the end of the 1990s or beginning of the 2000s is at, or close to, the end of life or already in the decommission phase. The number of cyclotrons in decommissioning will necessarily grow in the next years. The decommissioning of medical accelerators is therefore a very up-to-date theme of research since information is limited, if not lacking, insofar as the activation of the materials and subsequent implications on radioactive waste management procedures.
Considering PET cyclotrons, the (p,n) reaction exploited in the daily production of $^{18}F$ from $^{18}O$ represents the main source of secondary neutrons. The activation of surrounding material via neutron capture determines the production of short and long-lived radionuclides.
The induced activation poses a critical issue from the radiation protection point of view since it may potentially produce radioactive waste that needs specific handling and disposing procedures [2-7]. In particular, the activation induced has to be characterized following the European radiation protection directive (Directive 2013/59/Euratom) [8].
Furthermore, to decrease future dismantling costs, which might even be larger than the cost of installation, decommissioning has to be considered at an early stage, during the design and planning phase of an accelerator facility. This will make it possible to identify any critical issue as well as possible countermeasures to be taken during the construction phase. Evaluating in advance the amount of activation at the end of the cyclotron lifetime is strategic to plan an appropriate decommissioning approach.
Evaluation of induced radioactivity is a challenging task, due to all the complex physical phenomena involved. Type and level of activation strongly depend on several factors such as: the type of accelerator and targets, the beam energy and intensity, the workload of the accelerator, the geometry of the bunker and materials composition. For these reasons, a specific decommissioning plan is necessary for each individual facility.
Relatively few studies have been published on neutron activation of the concrete and their majority refers to high energy accelerator facilities used in fundamental research [9-13]; only few studies concern PET cyclotron facilities [14-17].
Accurate assessment of residual activation is challenging. As far as neutron fluxes are concerned, experimental measurements during the operation of the cyclotron are complex, time consuming and not always feasible for every irradiation condition. Furthermore, the monitoring or periodical measurement of the activation of building materials, such as concrete, is not straightforward; it can not always be implemented, or it may be feasible only in a limited amount of positions. On the other hand, an analytical approach is not possible without introducing strong approximations that affect significantly the accuracy of the results. For all of these reasons, an approach based on Monte Carlo simulations represents a valuable option.
This work describes an approach applied to two real cases: the GE PETtrace facility of the Sant'Orsola-Malpighi Hospital in Bologna and the IBA CYCLONE 18/18 HC facility at the Bern University Hospital (Inselspital). An accurate model of each facility was realized and used in the Monte Carlo simulation. On the other hand, these two facilities are currently in operation, making it possible to perform experimental measurements inside their bunkers to compare experimental and simulated results.

An analysis of these two cases can be used as guidance for many other facilities, since these two accelerators are the most commonly found worldwide for the production of PET radioisotopes [18].

## 2. Materials and methods

The main features of the two facilities considered in this work are reported in Table 1.

**Table 1 Main features of the two cases studies analysed in this work**

| | S.Orsola-Malpighi Hospital (Bologna) | Inselspital (Bern) |
|---|---|---|
| **First year of operation** | 2002 | 2012 |
| **Cyclotron** | GE PETtrace | IBA Cyclone 18 HC |
| **Energy** | 16.5 MeV | 18 MeV |
| **Acceleration plane** | Vertical | Horizontal |
| **N° of targets** | 6 | 8 |
| **N° of irradiations per day** | 1-2 | 1-3 |
| **Typical irradiation conditions** | $^{18}$F - 60 µA for 100 min<br>$^{11}$C - 70 µA for 25 min | $^{18}$F – 70 µA for 90 min<br>(140 µA dual beam) |
| **Activity typically produced at EOB (End Of Bombardment)** | 220-250 GBq of $^{18}$F<br>140-185 GBq of $^{11}$C | 300 – 500 GBq of $^{18}$F |
| **Workload in a year** | ≈ 25000 µAh for $^{18}$F<br>≈ 4000 µAh for $^{11}$C | ≈ 59000 µAh of $^{18}$F |

The Monte Carlo code FLUKA was used to quantify the radioactivity induced. FLUKA is a general purpose code for modelling particle transport and interactions with matter; it covers an extended range of applications spanning from proton and electron accelerator shielding to calorimetry, dosimetry, detector design, and radiotherapy [19,20]. FLUKA allows detailed calculations of induced radioactivity, as reported in several comprehensive benchmark studies [21-24]. Even if the original field of application of FLUKA was high energy physics, it has been gradually extended and nowadays it is widely resorted to also in medical physics applications at lower energy ranges [25-27].

A detailed model of the two cyclotron facilities was developed and implemented to assess the neutron flux around the accelerator and activation induced in the concrete bunker walls. To validate the models the results of Monte Carlo simulations were compared with experimental measurements conducted in the two facilities, as described in a previous work [6]. For this purpose, only gamma emitter radionuclides were considered, since experimental measurements of beta emitting radionuclides are cumbersome. Short half-lives radionuclides (below 75 days) are of no interest for this study, considering typical cooling times before decommissioning.

As a general consideration, activation of concrete is strongly dependent on the composition of the materials, including trace elements whose concentration is typically not accurately known. Even small differences in material composition may affect final results significantly. For this reason, the methodology presented in this work does not intend to assess activation with a high level of precision, applicable to all scenarios and facilities. Nevertheless, the results obtained give an order of magnitude of the typical levels of activation in cyclotron's bunkers, offering useful guidance also in different situations.

### 2.1 FLUKA Monte Carlo model of the GE PETtrace facility in Bologna

The GE PETtrace is a compact cyclotron with vertical acceleration plane capable of accelerating $H^-$ ions up to an energy of 16.5 MeV. The unit installed at the S.Orsola Malpighi Hospital in Bologna is in operation since 2002 in the routine production of PET radiopharmaceuticals. The workload of this accelerator is characterized by typical daily irradiations at 60 µA with an irradiation time of 100 minutes for the production of Fluorine-18 using a Niobium body target, and by daily productions of $^{11}$C, bombarding for 20 - 30 minutes at 70 µA. The cyclotron is used five days per week. The cyclotron is equipped with six targets, two of which are dedicated to the production of Fluorine-18. A detailed MC model of the GE PETtrace cyclotron was realized to allow accurate transport of neutrons produced inside the Fluorine target by (p,n) reactions. As shown in Figure 1, the model includes all the major components of the cyclotron that are expected to interact with the neutrons generated: the magnet and magnet poles (iron), the vacuum chamber (aluminum), a schematic model of the coils (copper), of the dees (copper) and of the body of the internal ion source (brass). Data on the dimensions and features of the cyclotron and its components were taken from the technical sheets and reference manuals of the vendor [28].

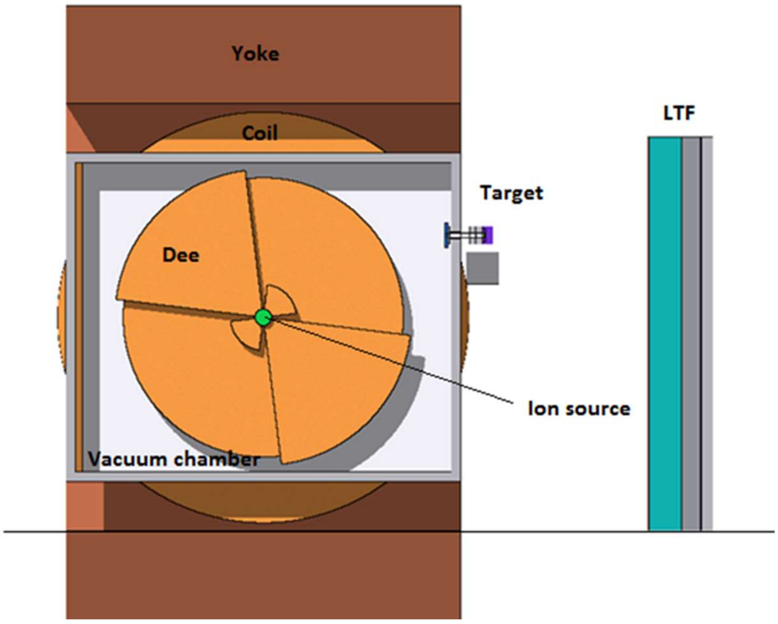


**Figure 1 - FLUKA MC model of the internal structure of the GE PETtrace cyclotron.**

The target system modelled was the PT800 18F Nb 25 target system (Figure 2), comprising a niobium chamber, the target body (Aluminium) and two Havar™ foils (25 µm and 50 µm thick). Havar is an alloy of cobalt (42%), chromium (20%), nickel (20%) and traces of manganese, molybdenum, iron and others [29].

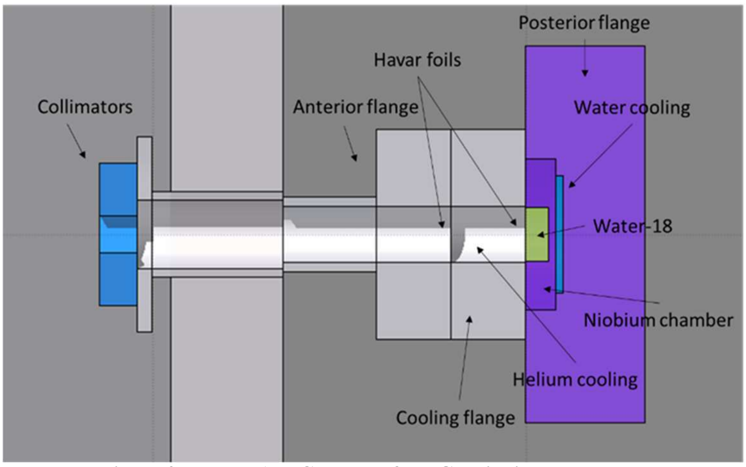


**Figure 2 – FLUKA MC model of the GE niobium target assembly**

The cyclotron bunker was modelled on the basis of the original construction drawings. The inner dimensions of the bunker are: 650 cm by 535 cm with a height of 350 cm and with 200 cm thick concrete walls (Figure 3). Reinforcement bars were also modelled since their contribution to the activation is significant.

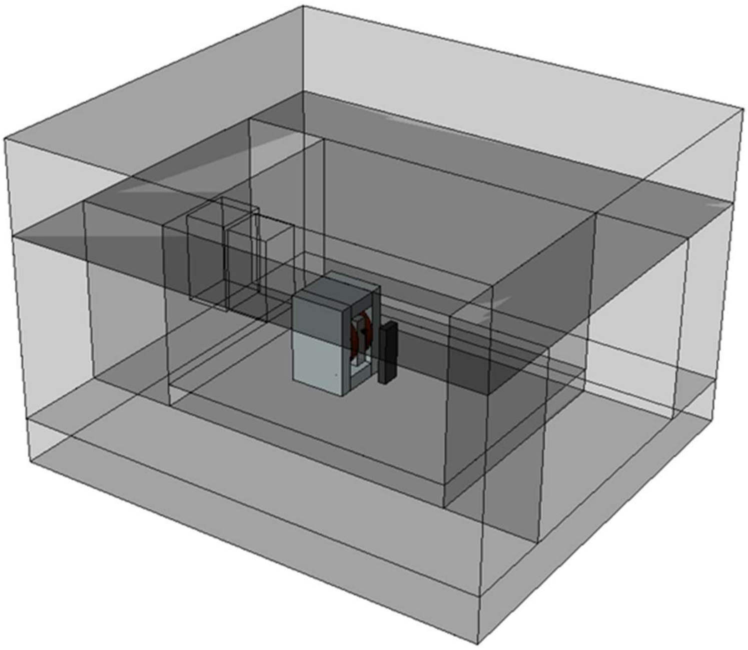

**Figure 3 FLUKA Monte Carlo model of the cyclotron bunker of S. Orsola Malpighi Hospital.**

The average density of concrete was 2.35 g/cm$^3$, as reported in technical sheets. The exact compositions of concrete and reinforcement rods were unknown. Since a chemical analysis was not possible, the standard Portland concrete present in FLUKA was used. This library does not include elements as cesium, cobalt and europium, the amount of those elements was taken from literature data [8]. The composition used in the simulation is reported in Table 2.

**Table 2 - Concrete composition used for the S. Orsola cyclotron bunker modelling (elements marked with * are taken from literature).**

| *ELEMENTS* | *Mass Fraction* | *ELEMENTS* | *Mass Fraction* |
|---|---|---|---|
| ***Si*** | 0.24 | ***Ba**** | 0.00025 |
| ***Ca*** | 0.037 | ***Ni**** | 3.1E-5 |
| ***Al*** | 0.018 | ***Co**** | 1.9E-6 |
| ***Fe*** | 0.016 | ***Nb**** | 2.3E-6 |
| ***K*** | 0.0038 | ***Eu**** | 6.6E-8 |
| ***Mg*** | 0.0035 | ***Cs**** | 1.2E-6 |
| ***Na*** | 0.0013 | ***O*** | 0.667 |
| ***Mn**** | 0.0011 | ***H*** | 0.01 |
| ***Ti**** | 0.00095 | ***C*** | 0.001 |

For the reinforcement rods, the composition used was a combination of steel SS316LN present in FLUKA adding impurities concentration values present in literature (Table 3) [9].

**Table 3 - Reinforcement rods composition (elements marked with * are taken from literature).**

| *ELEMENTS* | *Mass Fraction* | *ELEMENTS* | *Mass Fraction* |
|---|---|---|---|
| ***Cr*** | 1.85E-01 | ***Si*** | 1.00E-02 |
| ***Mn*** | 2.00E-02 | ***C*** | 3.00E-04 |
| ***Fe*** | 6.71E-01 | ***P*** | 4.50E-04 |
| ***Ni*** | 1.13E-01 | ***S*** | 3.00E-04 |
| ***Co**** | 1.00E-04 | | |

## 2.2 FLUKA Monte Carlo model of the IBA Cyclone 18 HC facility in Bern

The IBA Cyclone 18 HC is a fixed-energy cyclotron with a horizontal acceleration plane, capable of accelerating negative hydrogen $H^-$ ions at an energy of 18 MeV. The unit in Inselspital Bern was installed in 2012. This facility is conceived for commercial routine radioisotope production, as well as for multi-disciplinary research, the two activities running in parallel. The cyclotron has eight independent exit ports, one of which connected with a 6 m long external transport beam line terminated in a second bunker, dedicated to scientific activities. Four ports are connected with fluorine liquid targets and are used for the routine production of PET radionuclides. One port is equipped with a solid target station for the production of non-conventional radioisotopes[30]. The average

workload of the accelerator is equivalent to 3 daily irradiations at 70 µA for 90 minutes alternating the four liquid target for the production of Fluorine-18.
In this case again, a detailed MC model was developed (Figure 4). The model includes all the major components: the yoke (steel); the poles (steel), with their hills and valleys; a schematic model of the coils (Copper); the vacuum chamber; the holes in the lower yoke needed for vacuum pumping and to introduce the RF power into the RF cavities. Finally, the eight target exit ports, four fitted with fluorine-18 target stations, and one connected to the external beam line (Aluminium), with two quadrupoles (Copper).

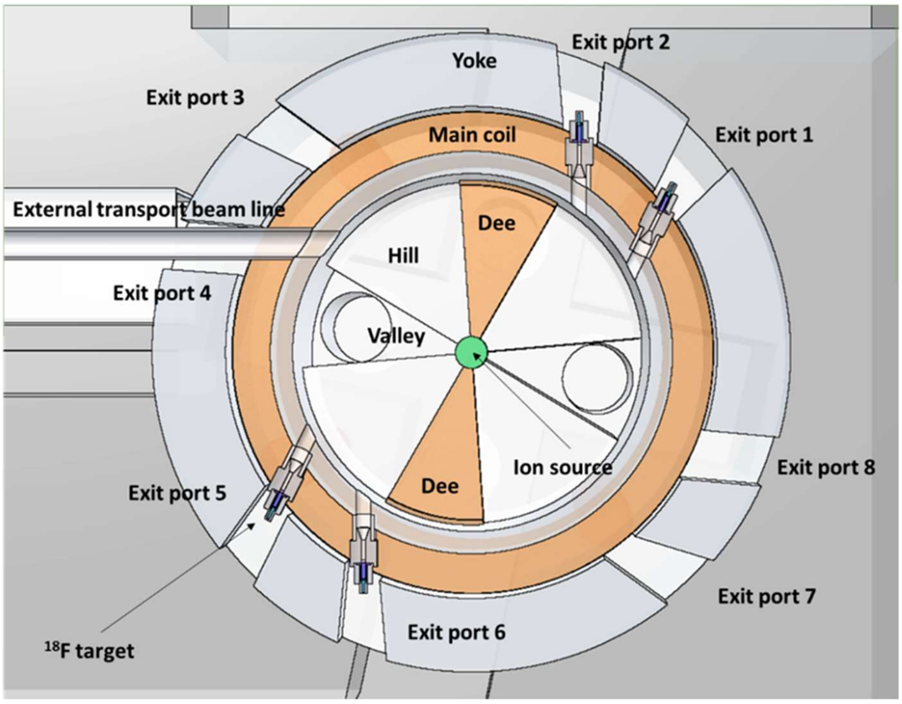

**Figure 4 - Horizontal view of the IBA CYCLOPE 18/93D FLUKA model implemented in this work.**

The target system modelled was the IBA Nirta® large volume liquid target (Figure 5). The model includes the aluminum structure, the niobium chamber, the water-cooling system, the helium-cooling system, the collimator, titanium foil (12.5 µm) and Havar foil (35 µm).

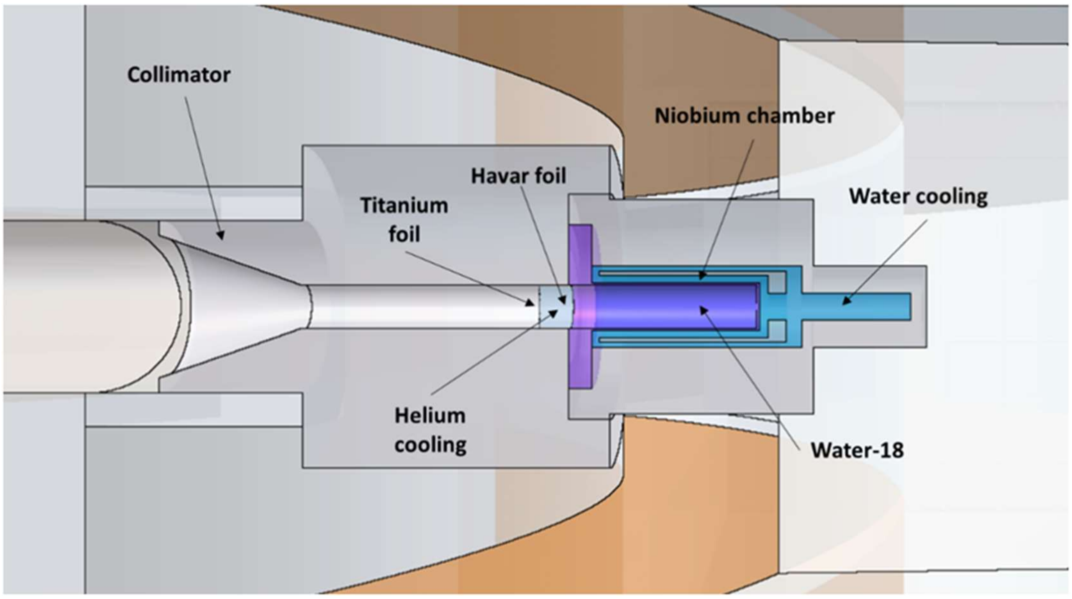

**Figure 5 - FLUKA model implemented in this work of IBA Nirta Liquid Target large volume.**

The MC model of the cyclotron vault was implemented on the basis of the original construction drawings: the building comprises two separated vaults, a vault for the cyclotron (4.00 m (width) x 4.75 m (length) x 3.00 m (height)), connected by a duct on a 1.8 m thick wall with a second vault (4.00 m (width) x 5.40 m (length) x 3.00 m (height)) for the external beam transfer line (BTL) (Figure 6). Also in this case the model includes the reinforcement rods.

**Figure 6 - Vertical view of 3D FLUKA MC model of cyclotron bunker.**

During the construction of the Bunker a sample of concrete was taken from the walls and sent to EMPA *Material Science & Technology* laboratories (Dübendorf, Switzerland) for an XRF Analysis to determine concrete composition. The qualification of 1st and 2nd period elements (from H to Ne) by XRF generally cannot be considered reliable in the case of inorganic solid matrices. Moreover, the amounts of some elements as cesium, cobalt and europium appeared to be below the limit of detection. Also in this case, the concrete composition was integrated with literature data, as reported in Table 4.

**Table 4 - XRF detected element in concrete sample (elements marked with * are taken from literature).**

| *ELEMENTS* | *Mass Fraction* | *ELEMENTS* | *Mass Fraction* |
|---|---|---|---|
| ***Na*** | 0.003 | ***Zn*** | 0.0001 |
| ***Mg*** | 0.008 | ***Sr*** | 0.0009 |
| ***Al*** | 0.036 | ***Zr*** | 0.0002 |
| ***Si*** | 0.16 | ***Ba**** | 0.00025 |
| ***P*** | 0.0008 | ***Ni**** | 3.1E-05 |
| ***S*** | 0.006 | ***Co**** | 1.9E-06 |
| ***Cl*** | 0.0004 | ***Nb**** | 2.3E-06 |
| ***K*** | 0.0104 | ***Eu**** | 6.6E-08 |
| ***Ca*** | 0.36 | ***Cs**** | 1.2E-06 |
| ***Ti*** | 0.003 | ***O**** | 0.3754 |
| ***Mn*** | 0.0005 | ***H**** | 0.05 |
| ***Fe*** | 0.024 | ***C**** | 0.001 |

The average density of concrete was assumed to be 2.28 g/cm$^3$, as reported in technical sheets. No accurate information regarding reinforcement rods composition was known, for this reason the composition of Table 3 was used here too.

**2.3 Physics settings of the Monte Carlo code and model of the neutron source**

The set of defaults called NEW-DEFA was used as basis for simulations. As in previous works [26, 27] , it proved to be the best compromise considering the activation of the main physical mechanisms, CPU-time usage and agreement with experimental results at this energy range. NEW-DEFA enables physical mechanisms such as transport of electrons, positrons, photons, low energy neutrons, delta ray production and heavy particle bremsstrahlung. For each simulation, two PHYSICS cards were also used to enable coalescence mechanisms and the new FLUKA evaporation model with heavy fragment evaporation.

Essentially, the most relevant neutron source is the production of $^{18}$F via the $^{18}$O(p,n)$^{18}$F reaction. Because of the relatively low neutron production rate (~$10^{-3}$ neutrons per incident proton), simulating protons as primary particles is quite inefficient to obtain low uncertainty results. The use of secondary neutrons as primary particles is a more efficient approach allowing to decrease by about three order of magnitude the number of primaries needed to obtain the same uncertainty results.

The spectrum of secondary neutrons resulting from the (p,n) reaction in daily production of $^{18}$F was first determined in a separate and simplified simulation. For this purpose, the proton beam and only the materials interacting with it were simulated and neutrons fluence as a function of energy was scored on a sphere surrounding the target. Considering the production cross section of the *$^{18}$O(p,n)$^{18}$F,* the proton transport threshold was set at 1 MeV.

The average neutron spectra per primary proton obtained is shown in Figure 8. The neutron spectrum related to the PETtrace shows a peak around 2.4 MeV, while the one related to the CYCLONE shows a peak around 2.8 MeV. The spectrum for the CYCLONE is higher due to the higher energy and larger cross section.

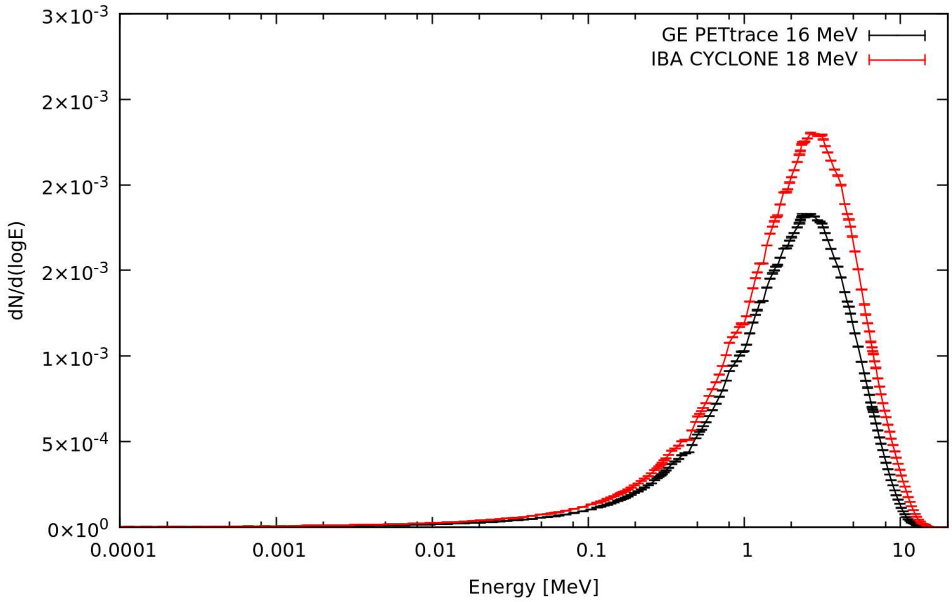

**Figure 8 - Neutron energy spectra per primary proton generated by the $^{18}O(p,n)^{18}F$ reaction in PETtrace target (black) and in CYCLONE target (red).**

The neutron multiplicity estimated, defined as the integrated number of neutrons produced by one incident proton, is equal to (3.99 ± 0.08) $10^{-3}$ [#neutrons/primary proton] for the PETtrace and (5.10 ± 0.01) $10^{-3}$ [#neutrons/primary proton] for the Cyclone. These results are in very good agreement with the data from the original works of Tesch [31], as reported by NCRP [7].
The neutron source intensities and spectra obtained were then used directly as the source term for activation assessments using an external subroutine. In FLUKA, a number of user routine templates are available and can be modified by the user to fulfil non-standard tasks. In this work, the *source.f* user routine was modified to model a point, isotropic source positioned inside the target. In both cases, the neutron spectrum was modelled as isotropic.

### 2.4 Scoring and residual activation assessment

The activation of the cyclotron vault walls and of the reinforcement rods was assessed using several RESNUCLEI cards; the activity was scored at different positions and depths for different life expectancies of the cyclotron.
Since RESNUCLEI scores nuclei on a region basis, 10 target positions were modelled inside the bunker walls as the sum of rectangular slabs (100x100x10 cm) to evaluate the in-depth activation profile and to score the distribution of activity concentration for each nuclide inside the walls (Figure 9 and 10).

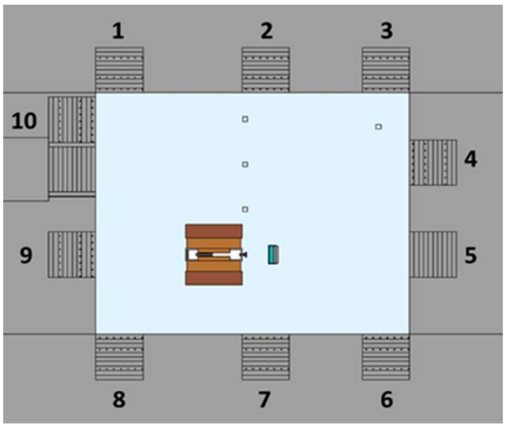

**Figure 9 - FLUKA MC model of PETtrace bunker where the positions in which activity concentration was estimated are indicated.**

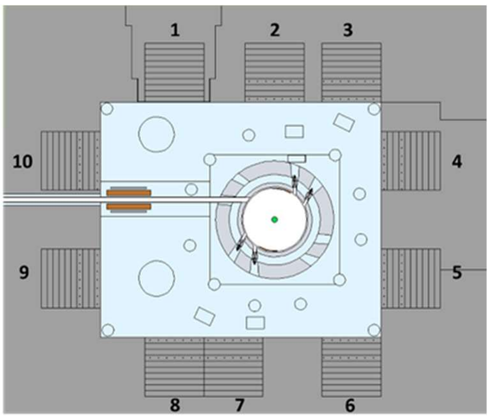

**Figure 10 – FLUKA MC model of CYCLONE 18/9 bunker where the positions in which activity concentration was estimated are indicated.**

RESNUCLEI results were expressed for each radionuclide in numbers of residual nuclei per cubic centimeter per primary neutron. The estimation of residual activity after different life expectancy of the cyclotron was done off-line by means of an external tool that was developed for this purpose.

The activity concentration per unit target current and unit irradiation time was estimated from the number of nuclei for each radionuclide $N_r$, as

$$A_r = N_r * \frac{1}{\rho} * n_n * \frac{ln(2)}{T_{1/2r}}$$ ***Equation 1***

Where $N_r$ is the number of nuclei per unit volume and per primary neutron, $\rho$ is the material density, $n_n$ is the number of neutrons generated from the target per unit current and per unit time and $T_{1/2r}$ is the radionuclide half-life expressed in second. $A_r$ can also be defined as the activity concentration after an irradiation of 1 second at 1 $\mu A$. On this basis, the saturation yield per unit current was calculated as

$$Y_{r,sat} = \frac{A_r}{1 - e^{\frac{-ln2}{T_{1/2r}}}}$$ ***Equation 2***

The activity induced in materials after irradiation was calculated as a function of the cooling time $t_c$using the following equation:

$$A_r(t) = Y_{sat} * I * \left(1 - e^{-\lambda t_{irr}}\right) * e^{-\lambda t_c}$$ ***Equation 3***

Where I is the irradiation current, $t_{irr}$ is the duration of the irradiation.
In order to compare the simulations with experimental data, the residual activation was estimated after 16 years of operation for the PETtrace facility and after 4 years for the CYCLONE facility. The average irradiation conditions described in paragraphs 2.1 and 2.2 were used.

## 3. Results

The main long lived radionuclides produced are reported in Table 5 together with the most probable production reactions with its cross-section, decay mode and most relevant gamma emissions. The clearance levels from Table A –Annex VII of Directive 2013/59/Euratom[8] are also indicated.

**Table 5 - Main radionuclides found with the most probable production reaction**

| Nuclide | $T_{1/2}$ | Reaction | Cross section | Abundance (%) | Decay mode | Main gamma emissions | Clearence level (Bq/g) |
|---|---|---|---|---|---|---|---|
| $^{154}Eu$ | 8.8 y | $^{153}Eu(n,\gamma)^{154}Eu$ | 312 b | 52 | β-, EC β+ | 123 keV (40%)<br>1274 keV (35%) | 0.1 |
| $^{152}Eu$ | 13.5 y | $^{151}Eu(n,\gamma)^{152}Eu$ | 9198 b | 48 | β-, EC | 121 keV (28%)<br>344 keV (26%) | 0.1 |
| $^{134}Cs$ | 2.1 y | $^{133}Cs(n,\gamma)^{134}Cs$<br>$^{134}Ba(n,p)^{134}Cs$ | 29 b<br>9 mb at En = 16 MeV | 100<br>2 | β-, EC | 604 keV (97%)<br>795 keV (85%) | 0.1 |
| $^{65}Zn$ | 244.3 d | $^{64}Zn(n,\gamma)^{65}Zn$ | 0.78 b | 49 | EC β+ | 1115 keV (50%) | 0.1 |
| $^{60}Co$ | 5.3 y | $^{59}Co(n,\gamma)^{60}Co$ | 37 b | 100 | β- | 1332 keV (99%)<br>318 keV (99%) | 0.1 |
| $^{46}Sc$ | 83.8 d | $^{45}Sc(n,\gamma)^{46}Sc$ | 27 b | 100 | β- | 889 keV (99%)<br>1120 keV (99%) | 0.1 |
| $^{54}Mn$ | 312.3 d | $^{55}Fe(n,p)^{54}Mn$ | 590 mb at En = 10 MeV | 6 | EC β+, β- | 834 keV (99%) | 0.1 |

As expected most of the production of long-lived radionuclides is due to the high capture cross section for thermal neutrons of some heavy elements like europium, typically present in trace amounts in concrete, or cobalt and cesium, present in trace amount both in concrete and in reinforcement structures. In the following the results obtained in the two study cases are reported.

### 3.1 Comparison of the Monte Carlo models with experimental measurements

The accuracy of Monte Carlo simulations was assessed through comparison with experimental measurements previously performed that can be found in Reference [6]. In Bologna, non-destructive measurements of the concrete walls were performed inside the bunker with a portable CZT detector. The detector used was the Kromek GR1 (Kromek™ , Sedgefield, UK) ), a portable gamma-ray spectrometer composed of a 1 cm$^3$ CdZnTe crystal with an energy resolution of 2.0 % FWHM at 662 keV. Measurements were performed during cyclotron maintenance in the three position shown in Figure 11. The efficiency calibration of the detector was obtained through Monte Carlo simulations in a previous work [32].

In Bern it was possible to collect three samples of concrete by making core drilling, and measure them in laboratory with a HPGe detector. The detector used had 30% relative efficiency (measured at 25 cm source-detector distance, relative to a 3''x 3'' NaI(Tl) detector) and a resolution of 1.8 keV at 1332 keV. The sampling positions are reported in Figure 12.

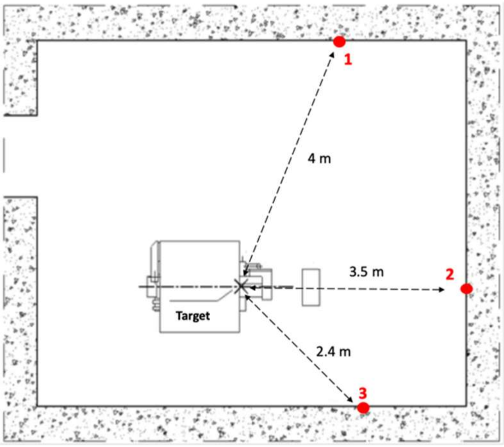


**Figure 11 - Kromek GR1 measurement positions inside the PETtrace bunker.**

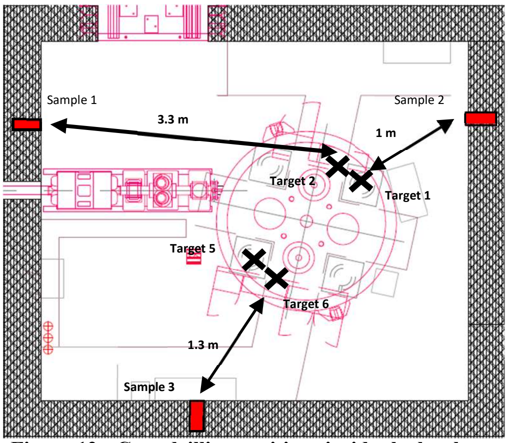


**Figure 12 - Core drilling positions inside the bunker of the Berrn medical cycloton.**

The results for the GE PETtrace facility are reported in Tables 5, 6 and 7. The consistency between Monte Carlo results and experimental measurements is generally within a factor 2 - 3 for most radionuclides, and within a factor of 6 for Eu-152. Discrepancies in activation assessment are probably due to the unknown real concrete composition, especially for what concern trace elements. The literature values used in the simulations could be quite different from the real trace element concentration. For example the absence of $^{46}$Sc in simulated results is due to the absence of $^{45}$Sc in the concrete composition modelled.

**Table 5 - Comparison between activity concentrations measured experimentally with the Kromek GR1 in Bologna and the corresponding activity concentrations estimated with Fluka for position 1.**

| | *Experimental* | | *Fluka* | | |
|---|---|---|---|---|---|
| *Nuclide* | *Activity concentration (Bq/g)* | *Uncertainty (%)* | *Activity concentration (Bq/g)* | *Uncertainty (%)* | *FLUKA/ Experimental* |
| $^{46}$Sc | 0.26 | 18 | - | - | - |
| $^{54}$Mn | 0.18 | 13 | 0.14 | 2 | 0.78 ± 0.10 |
| $^{60}$Co | 0.17 | 12 | 0.21 | 4 | 1.24 ± 0.16 |
| $^{152}$Eu | 0.12 | 23 | 0.75 | 6 | 6.25 ± 1.49 |
| $^{134}$Cs | 0.04 | 30 | 0.01 | 12 | 0.25 ± 0.08 |
| $^{154}$Eu | <0.02* | - | 0.08 | 9 | - |

**Table 6 - Comparison between activity concentrations measured experimentally with the Kromek GR1 in Bologna and the corresponding activity concentrations estimated with Fluka for position 2.**

| | *Experimental* | | *Fluka* | | |
|---|---|---|---|---|---|
| *Nuclide* | *Activity concentration (Bq/g)* | *Uncertainty (%)* | *Activity concentration (Bq/g)* | *Uncertainty (%)* | *FLUKA /Experimental* |
| $^{60}$Co | 0.04 | 17 | 0.13 | 16 | 3.25 ± 0.76 |
| $^{152}$Eu | <0.04* | - | 0.20 | 4 | - |
| $^{46}$Sc | <0.02* | - | - | - | - |
| $^{134}$Cs | <0.01* | - | 0.01 | 26 | - |
| $^{54}$Mn | <0.01* | - | 0.01 | 1 | - |
| $^{154}$Eu | <0.02* | - | 0.09 | 45 | - |

**Table 7 - Comparison between activity concentrations measured experimentally and the corresponding activity concentrations estimated with Fluka for position 3.**

| Nuclide | Experimental | | Fluka | | FLUKA/ Experimental |
|---|---|---|---|---|---|
| | Activity concentration [Bq/g] | Uncertainty (%) | Activity concentration [Bq/g] | Uncertainty (%) | |
| $^{152}$Eu | 0.36 | 13 | 1.72 | 4 | 4.78 ± 0.65 |
| $^{60}$Co | 0.19 | 12 | 0.48 | 3 | 2.53 ± 0.31 |
| $^{134}$Cs | 0.07 | 15 | 0.03 | 13 | 0.43 ± 0.13 |
| $^{54}$Mn | 0.08 | 15 | 0.20 | 1 | 2.50 ± 0.38 |
| $^{46}$Sc | 0.12 | 14 | - | - | - |
| $^{154}$Eu | <0.03* | - | 0.09 | 45 | - |

The activity concentrations of the three samples collected in the bunker of the IBA CYCLONE facility are reported in Tables 8, 9, and 10. In this case the consistency between FLUKA and experimental measurements is typically around a factor of 2, except for $^{46}$Sc and $^{65}$Zn. Also in this case the absence of $^{46}$Sc in simulated results is due to the absence of $^{45}$Sc in the concrete composition modelled.

**Table 8 - Comparison between activity concentrations measured experimentally and the corresponding activity concentrations estimated with Fluka for sample 1.**

| Nuclide | Experimental | | Fluka | | FLUKA/Experimental |
|---|---|---|---|---|---|
| | Activity concentration [Bq/g] | Uncertainty (%) | Activity concentration [Bq/g] | Uncertainty (%) | |
| $^{60}$Co | 0.17 | 3 | 0.21 | 2 | 1.20 ± 0.04 |
| $^{152}$Eu | 0.10 | 2 | 0.10 | 3 | 0.98 ± 0.03 |
| $^{46}$Sc | 0.10 | 3 | - | - | |
| $^{134}$Cs | 0.03 | 3 | 0.01 | 19 | 0.41 ± 0.08 |
| $^{65}$Zn | 0.02 | 5 | 0.10 | 3 | 6.19 ± 0.36 |
| $^{154}$Eu | 0.01 | 8 | 0.02 | 16 | 0.10 ± 0.02 |
| $^{54}$Mn | 0.01 | 11 | 0.01 | 41 | 0.51 ± 0.22 |

**Table 9 - Comparison between activity concentrations measured experimentally and the corresponding activity concentrations estimated with Fluka for sample 2.**

| Nuclide | Experimental | | Fluka | | FLUKA/Experimental |
|---|---|---|---|---|---|
| | Activity concentration [Bq/g] | Uncertainty (%) | Activity concentration [Bq/g] | Uncertainty (%) | |
| $^{46}$Sc | 0.59 | 3 | - | - | |
| $^{152}$Eu | 0.52 | 1 | 0.22 | 2 | 0.41 ± 0.01 |
| $^{60}$Co | 0.32 | 3 | 0.35 | 3 | 1.08 ± 0.05 |
| $^{65}$Zn | 0.11 | 4 | 0.37 | 2 | 6.19 ± 0.28 |
| $^{134}$Cs | 0.21 | 2 | 0.09 | 11 | 0.41± 0.05 |
| $^{54}$Mn | 0.09 | 4 | 0.04 | 13 | 0.40 ± 0.05 |
| $^{154}$Eu | 0.06 | 3 | 0.02 | 10 | 0.28 ± 0.03 |

**Table 10 - Comparison between activity concentrations measured experimentally and the corresponding activity concentrations estimated with Fluka for sample 3.**

| *Nuclide* | *Experimental* | | *Fluka* | | *FLUKA/Experimental* |
|---|---|---|---|---|---|
| | *Activity concentration [Bq/g]* | *Uncertainty (%)* | *Activity concentration [Bq/g]* | *Uncertainty (%)* | |
| $^{152}Eu$ | 0.41 | 2 | 0.19 | 3 | 0.47 ± 0.02 |
| $^{46}Sc$ | 0.38 | 3 | - | - | |
| $^{60}Co$ | 0.21 | 3 | 0.29 | 2 | 1.35 ± 0.05 |
| $^{134}Cs$ | 0.21 | 2 | 0.09 | 9 | 0.44 ± 0.04 |
| $^{65}Zn$ | 0.06 | 5 | 0.39 | 2 | 6.98 ± 0.38 |
| $^{54}Mn$ | 0.08 | 5 | 0.03 | 15 | 0.41 ± 0.06 |
| $^{154}Eu$ | 0.05 | 3 | 0.02 | 43 | 0.34 ± 0.15 |

The results obtained for the two facilities show the importance of having accurate information on the composition of the concrete actually used in the construction of the vault. Nevertheless, they demonstrate that FLUKA can be satisfactorily used to assess the order of magnitude of the residual induced radioactivity, with accuracy increasing with better knowledge of the composition of the materials.

### 3.3 Assessment of the residual activation

Once the accuracy of Monte Carlo results was assessed in some reference points, the activation at different positions and for different life expectancy of the cyclotron was estimated. As reported in Table 11, the activity concentration inside the bunker of the S.Orsola-Malpighi hospital was calculated for the 10 positions of Figure 9. The activity concentration refers to the first 10 cm of thickness inside the bunker wall and was calculated as the average value of activity concentration in concrete and in reinforcement rods after 14 years of cyclotron operation.

**Table 11 - Activity concentration of the main long lived radionuclides in the first 10 cm inside the bunker wall of the PETtrace facility in Bologna at different positions.**

| | Nuclides | Eu-154 | Eu-152 | Cs-134 | Co-60 | Mn-54 |
|---|---|---|---|---|---|---|
| Activity concentration [Bq/g] | Position 1 | 0,041 ± 0,002 | 0,421 ± 0,006 | 0,016 ± 0,001 | 0,360 ± 0,016 | 0,131 ± 0,003 |
| | Position 2 | 0,087 ± 0,003 | 0,830 ± 0,009 | 0,021 ± 0,001 | 0,443 ± 0,014 | 0,156 ± 0,003 |
| | Position 3 | 0,082 ± 0,003 | 0,712 ± 0,008 | 0,036 ± 0,002 | 0,442 ± 0,012 | 0,092 ± 0,002 |
| | Position 4 | 0,085 ± 0,003 | 0,876 ± 0,010 | 0,024 ± 0,001 | 0,490 ± 0,016 | 0,143 ± 0,003 |
| | Position 5 | 0,093 ± 0,004 | 0,562 ± 0,006 | 0,035 ± 0,002 | 0,191 ± 0,006 | 0,012 ± 0,001 |
| | Position 6 | 0,089 ± 0,003 | 0,878 ± 0,009 | 0,038 ± 0,002 | 0,473 ± 0,014 | 0,128 ± 0,002 |
| | Position 7 | 0,140 ± 0,004 | 1,961 ± 0,014 | 0,042 ± 0,002 | 0,842 ± 0,021 | 0,519 ± 0,005 |
| | Position 8 | 0,042 ± 0,003 | 0,403 ± 0,006 | 0,017 ± 0,001 | 0,218 ± 0,010 | 0,011 ± 0,001 |
| | Position 9 | 0,043 ± 0,002 | 0,514 ± 0,007 | 0,014 ± 0,001 | 0,260 ± 0,010 | 0,032 ± 0,001 |
| | Position 10 | 0,041 ± 0,002 | 0,460 ± 0,007 | 0,012 ± 0,001 | 0,237 ± 0,009 | 0,018 ± 0,001 |

As expected, the highest specific activation was found at the locations closest to the targets, in particular at position 7. The maximum activity concentration was estimated to be 1.96 Bq/g for Eu-152 and 0.84 Bq/g for Co-60. In Figure 13, the in-depth activation profile for position 7 is shown together with the clearance level of 0.1 Bq/g.

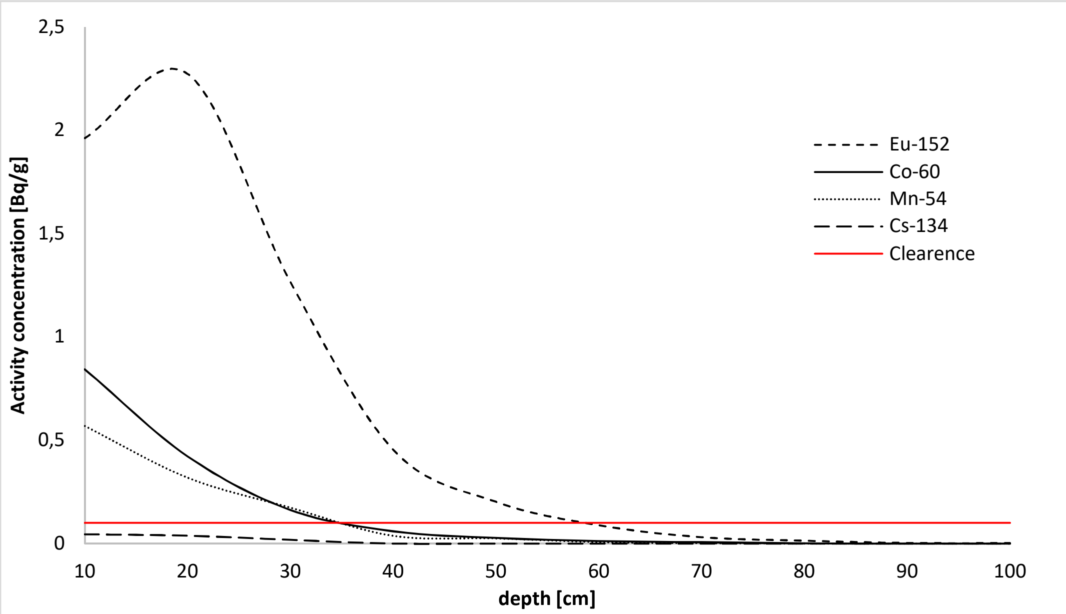


**Figure 13 - In-depth activation profile in position 7 of the PETtrace facility in Bologna.**

The activity concentration is higher in the first 40 cm and decreases with increasing depth becoming lower than 0.1 Bq/g after about 60 cm and almost negligible after the first 70 cm. The activation level is lower in positions 1, 8, 9 and 10 due to the combination of distance and relative position with respect to targets. Furthermore, the presence and position of reinforcement rods in the concrete is, as expected, very important. As it can be noted, the position with the lowest activation for $^{60}$Co and $^{54}$Mn is position 5. This is due to the fact that at this position the wall is made of concrete bricks only and reinforcement rods are not present.
The activity concentrations in the Inselspital bunker, after 4 years of cyclotron operation, are reported in Table 12. Also in this case, the activity concentration refers to the first 10 cm inside the wall and was calculated as the average value of activity concentration in concrete and reinforcement rods.

**Table 12 - Activity concentration of the main long-lived radionuclides in the first 10 cm inside the wall of the CYCLONE bunker in Bern at different positions.**

| | Nuclides | Eu-154 | Eu-152 | Cs-134 | Co-60 | Mn-54 |
|---|---|---|---|---|---|---|
| Activity concentration [Bq/g] | **Position 1** | 0,004 ± 0,001 | 0,051 ± 0,002 | 0,020 ± 0,004 | 0,354 ± 0,027 | 0,034 ± 0,006 |
| | **Position 2** | 0,008 ± 0,001 | 0,134 ± 0,003 | 0,082 ± 0,010 | 0,247 ± 0,037 | 0,007 ± 0,001 |
| | **Position 3** | 0,017 ± 0,002 | 0,179 ± 0,004 | 0,090 ± 0,011 | 0,297 ± 0,030 | 0,021 ± 0,004 |
| | **Position 4** | 0,018 ± 0,002 | 0,217 ± 0,004 | 0,087 ± 0,011 | 0,348 ± 0,026 | 0,038 ± 0,007 |
| | **Position 5** | 0,009 ± 0,001 | 0,076 ± 0,002 | 0,027 ± 0,006 | 0,056 ± 0,011 | 0,007 ± 0,001 |
| | **Position 6** | 0,008 ± 0,001 | 0,107 ± 0,002 | 0,044 ± 0,007 | 0,192 ± 0,034 | 0,013 ± 0,002 |
| | **Position 7** | 0,018 ± 0,002 | 0,189 ± 0,004 | 0,091 ± 0,011 | 0,308 ± 0,023 | 0,034 ± 0,006 |
| | **Position 8** | 0,009 ± 0,001 | 0,099 ± 0,002 | 0,036 ± 0,008 | 0,241 ± 0,024 | 0,017 ± 0,003 |
| | **Position 9** | 0,004 ± 0,001 | 0,088 ± 0,003 | 0,024 ± 0,005 | 0,127 ± 0,019 | 0,013 ± 0,002 |
| | **Position 10** | 0,001 ± 0,001 | 0,100 ± 0,003 | 0,013 ± 0,003 | 0,205 ± 0,016 | 0,004 ± 0,001 |

The walls of the bunker are mostly activated in positions 3, 4 and 7, as those are the positions nearest to the targets. In those positions, an activity concentration of about 0.2 Bq/g and 0.3 Bq/g is expected for $^{152}$Eu and $^{60}$Co, respectively. The lowest activation is present at positions 9, 10 and 5. Position 1 presents the highest activity concentrations for $^{60}$Co and $^{54}$Mn, due to the fact that it corresponds to the bunker door, where a high concentration of steel is present. The in depth activation profile at position 4 is reported in Figure 14.

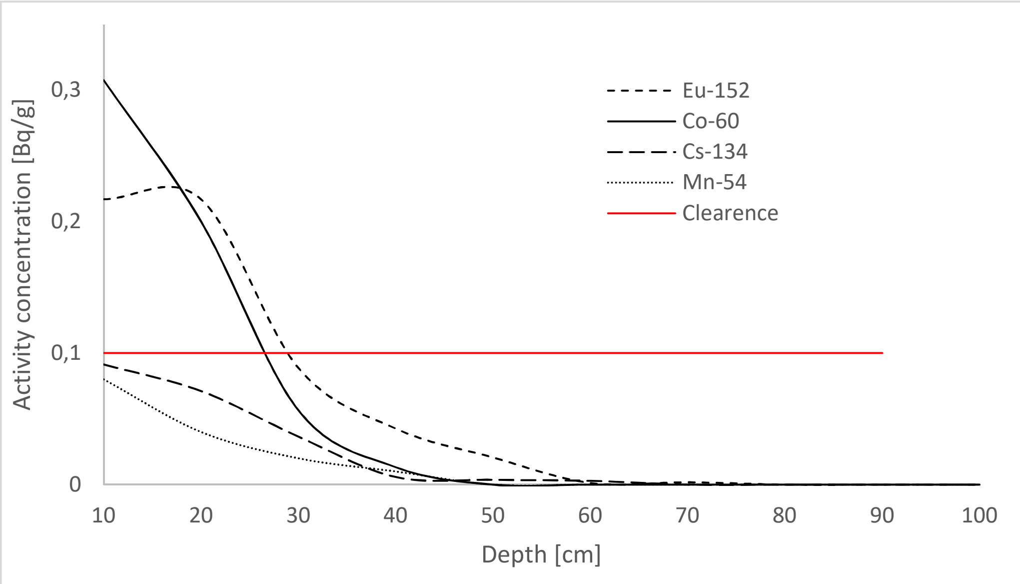


**Figure 14 - In-depth activation profile in position 4 of the CYCLONE facility in Bern.**

The activity concentration estimated at each of the positions of Table 11 and 12 exceeds the clearance levels of the new Directive 2013/59/Euratom (Table A – Annex VII) [8]. A selective removal of activated concrete can be performed considering conservatively the activation profiles of most activated positions (Figure 13, Figure 14).
The activation level was also assessed considering different life expectancies of the two facilities. As an example, activity concentrations in the most activated positions for the two facilities are reported in Figure 15 and 16 as a function of the years of operation of the facility.

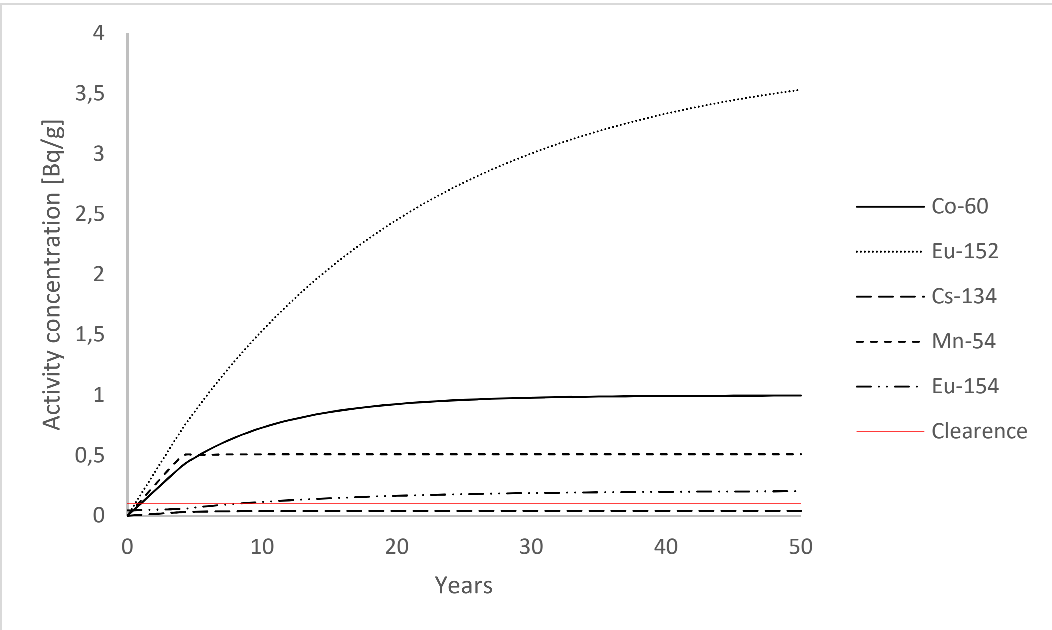


**Figure 15 - Prediction of residual activity at decommissioning, in the first 10 cm inside the wall at position 7 considering different life expectancy of the PETtrace facility in Bologna.**

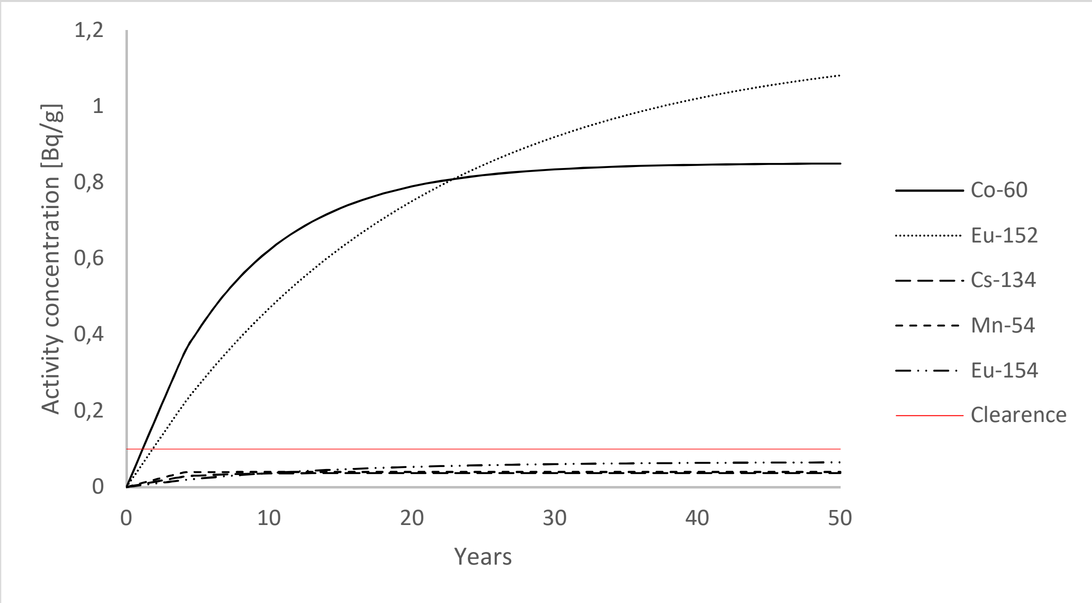


**Figure 16 - Prediction of residual activity at decommissioning in the first 10 cm inside the wall at position 4 considering different life expectancy of the CYCLONE facility in Bern.**

Prediction of future activation levels shows that the activity concentration of Cs-134 and Mn-54 remains basically constant after 5 years of operation, while for Co-60 it does not vary considerably after about 20 years. For Eu-152 and Eu-154, the case is different: due their much longer half-lives, saturation is not reached. From Figures 15 and 16, it also appears evident that the clearance level is exceeded already in the first years of operation.

**4 Conclusions**

The aim of this work was to develop methodologies for the assessment of activation induced in a bunker of a medical cyclotron, using the Monte Carlo code FLUKA.

The results of the simulations were compared with gamma spectrometry measurements. The results obtained demonstrate that FLUKA can be used satisfactorily to assess the order of magnitude of the residual radioactivity mostly induced by the neutron fields generated during proton irradiations for the production of radioisotopes for medical applications. The discrepancies between data and simulations are mainly due to differences between real and modelled material compositions, the accuracy of the results increases with a better knowledge of the composition of the materials. This methodology can be therefore valuable to assess in advance the level of activation of a cyclotron bunker in view of defining the optimum decommissioning plan. In particular, assessment of residual activation via Monte Carlo simulation allows planning the selective removal of the parts of the bunker walls that exceed clearance limits and have therefore to be considered as radioactive waste.

The results presented in this paper are instrumental for an accurate planning of new facilities, for which the decommissioning phase has to be considered as an important part of their life-cycle. Several factors are relevant in order to minimize the amount of nuclear waste produced during the operational phase of the facility:

- an appropriate choice and quality control of the materials used to build the bunker;
- an accurate analysis of the components, including trace elements;
- proper design and position of the reinforcement bars, possibly avoiding to place bars in the first 30 - 60 cm of the walls.

These aspects are also of paramount importance for a reliable simulation of the ongoing levels of activation.

Furthermore, in order to allow a periodical experimental assessment of the level of induced activation, it is recommended to provide for the installation of about 50 cm deep removable drilled bores located in chosen positions (in front of the target in particular); as an alternative, at the time of pouring the concrete of the vault, it is suggested to obtain an adequate number of test samples of concrete, to be left in specific position inside the vault, to help monitoring the activation in time.

**Acknowledgements**

We acknowledge the support by the Radiopharmacy teams at the S. Orsola Malpighi Hospital in Bologna and of SWAN Isotopen AG in Bern.

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