A method for radiological characterization based on fluence conversion coefficients

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A method for radiological characterization based on fluence conversion coefficients

Robert Froeschl
CERN, 1211 Geneva 23, Switzerland
E-mail: robert.froeschl@cern.ch

Abstract. Radiological characterization of components in accelerator environments is often required to ensure adequate radiation protection during maintenance, transport and handling as well as for the selection of the proper disposal pathway. The relevant quantities are typical the weighted sums of specific activities with radionuclide-specific weighting coefficients. Traditional methods based on Monte Carlo simulations are radionuclide creation-event based or the particle fluences in the regions of interest are scored and then off-line weighted with radionuclide production cross sections. The presented method bases the radiological characterization on a set of fluence conversion coefficients. For a given irradiation profile and cool-down time, radionuclide production cross sections, material composition and radionuclide-specific weighting coefficients, a set of particle type and energy dependent fluence conversion coefficients is computed. These fluence conversion coefficients can then be used in a Monte Carlo transport code to perform on-line weighting to directly obtain the desired radiological characterization, either by using built-in multiplier features such as in the PHITS code or by writing a dedicated user routine such as for the FLUKA code. The presented method has been validated against the standard event-based methods directly available in Monte Carlo transport codes.

1. Introduction
The radiological characterization of components after their use in an accelerator facility is an important task for operational radiation protection, due to the need to define adequate radiation protection measures during maintenance, transport or handling of a component or due to the need to select a proper disposal pathway.

The relevant radiological hazard factor for radiological characterization can often be written as a weighted sum, with radionuclide specific weighting coefficients, of specific activities.

The commonly used methods for obtaining the required specific activities are briefly presented and analyzed. This analysis highlights the need of an alternative method providing fast convergence, automatic normalization and good visualization capabilities.

A new method based on fluence conversion coefficients is developed that satisfies these requirements. The formulation of the method and its implementation is presented and its capabilities are demonstrated on selected examples.

Finally, promising future applications of this method are discussed.

2. Radiological characterization
The radiological hazard factor, denoted $H$, relevant for radiological characterization can often be written as a weighted sum, with radionuclide specific weighting coefficients $w_b$, of specific
activities $A_b$: 

$$H = \sum_b \frac{1}{w_b} A_b = \sum_b \frac{1}{w_b} \sum_r \sum_e T_{br} P_{re} m_e$$

(1)

The mass specific activity $A_b$ of radionuclide $b$ can often be written in terms of the production rate $P_{re}$ of radionuclide $r$ from the chemical element $e$

$$P_{re} = \frac{N_A}{M_e} \sum_{i=p,n,\pi^+,\pi^-} \int \phi_i(E) \sigma_{i,e,r}(E) dE,$$

(2)

where $N_A$ is Avogadro’s constant and $M_e$ is the atomic weight of the chemical element $e$. The sum is typically extended over protons ($p$), neutrons ($n$) and charged pions ($\pi^+, \pi^-$) for high energy hadron accelerators. In this formulation, the natural isotope abundances for each element $e$ are taken into account, i.e. the cross section $\sigma_{i,e,r}(E)$ is an abundance weighted average of the cross sections of each isotope of element $e$. Furthermore, $\Phi_i(E)$ denotes the radiation fluence for the various secondary particles ($i = p, n, \pi^+, \pi^-$). The expression $\sum_e P_{re} m_e$ corresponds to the production rate of isotope $r$ in the whole component.

The time evolution of the specific activity of radionuclide $b$, i.e. the build–up of all radionuclides $r$ and the full decay chain leading to isotope $b$, can be described by the matrix $T_{br}$, involving the time-dependent beam intensity profile and typically expressed using the bateman coefficient $[1, 2]$.

2.1. Standard methods for radiological characterization

Monte Carlo radiation transport simulation codes can be used to compute the radiological hazard factor $H$ needed for radiological characterization. There are two standard methods for these calculations that are often employed when using Monte Carlo simulation codes. These two methods are presented in the next paragraphs and their specific use in the FLUKA $[3, 4]$ and PHITS $[5, 6, 7]$ Monte Carlo radiation transport simulation codes are discussed.

Event based: The first standard method to obtain the required specific activities is based on the events of creation of radionuclides during interactions in the radiation transport. In FLUKA, the RESNUCLE card provides this capability, either scoring production yields or directly activities for a given irradiation profile. This scoring is based on regions defined in the geometry of the simulation.

In PHITS, the T-Yield tally can score production yields, also based on regions defined in the geometry of the simulation. The time evolution is also accounted for in the post-processing step.

Fluence spectra based: The second standard method to obtain the required specific activities is based on the calculation of fluence spectra in the Monte Carlo simulation and then weighting these spectra with radionuclide production cross-sections in a post-processing step. The time evolution is also accounted for in the post-processing step.

In FLUKA, the USRTRACK card provides this capability whereas in PHITS the T-Track tally can be used for this purpose.

The post-processing can be done with various codes, e.g. JEREMY $[8, 9]$ or ActiWiz $[10, 11]$.

A detailed comparison of the these two types of standard methods is presented in table 1. This comparison clearly demonstrates the need for an alternative method with fast convergence, automatic normalization and good visualization capabilities for a large number of cells.
Table 1. Comparison of the two standard methods for radiological characterization, described in section 2.1 and the fluence conversion coefficients method, described in section 3.

<table>
<thead>
<tr>
<th>Method</th>
<th>Pros</th>
<th>Cons</th>
</tr>
</thead>
<tbody>
<tr>
<td>Event based</td>
<td>Built-in capability with reduced human error probability</td>
<td>Convergence can be slow for regions away from main interaction zone</td>
</tr>
<tr>
<td></td>
<td>Very good for small number of regions close to main interaction zone</td>
<td>Region-based: Mass normalization has to be done manually</td>
</tr>
<tr>
<td></td>
<td>Uses cross-sections from MC code</td>
<td>Difficult visualization</td>
</tr>
<tr>
<td></td>
<td>Material compositions from MC input</td>
<td>Cumbersome for large number of regions</td>
</tr>
<tr>
<td>Fluence spectra based</td>
<td>Better convergence properties</td>
<td>Cross-sections decoupled from MC code</td>
</tr>
<tr>
<td></td>
<td>Possibility to evaluate alternative material compositions</td>
<td>Region-based: Volume normalization has to be done manually</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Difficult visualization</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cumbersome for large number of regions</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Higher post-processing needs</td>
</tr>
<tr>
<td>Fluence conv. coeff. based</td>
<td>Fully integrated into existing tool-chain</td>
<td>Cross-sections decoupled from MC code</td>
</tr>
<tr>
<td></td>
<td>Very good visualization of larger number of regions</td>
<td>Higher pre-processing needs</td>
</tr>
<tr>
<td></td>
<td>Automatic mass normalization</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Better convergence properties</td>
<td></td>
</tr>
</tbody>
</table>

3. Fluence conversion coefficients method

The radiological hazard factor $H$ in equation 1 can be expressed in terms of fluence conversion coefficients, denoted FCCs. This can be achieved by inserting equation 2 into equation 1, approximating the integrals over the particle energies by discrete sums and then re-ordering the summation in a way to move the particle-type summation and the energy summation as well as the particle fluences to the front.

$$H = \sum_b \frac{1}{w_b} \sum_{r,e} T_{br} P_{re} m_e$$

$$\approx \sum_b \frac{1}{w_b} \sum_{r,e} T_{br} \left( \frac{N_A}{M_e} \sum_{i=p,n,\pi^+,\pi^-} \sum_E \Phi_{i,E} \sigma_{i,e,r}(E) \right) m_e$$

$$= \sum_{i=p,n,\pi^+,\pi^-} \sum_E \Phi_{i,E} \left( \sum_{b,r,e} \frac{1}{w_b} T_{br} \frac{N_A}{M_e} \sigma_{i,e,r}(E) m_e \right)_{K_{i,E}}$$

Equation 3 now expresses the radiological hazard factor $H$ in terms of fluence conversion coefficients $K_{i,E}$, yielding also the definition for the $K_{i,E}$ coefficients. The basic idea of the presented method is to precompute the fluence conversion coefficients $K_{i,E}$ for a given radiological characterization task and then to apply them on-line during the Monte Carlo radiation transport simulation.
3.1. Implementation
A dedicated code for the pre-computation of the desired fluence conversion coefficients has been developed. This code takes the material composition, the irradiation and cool-down profile as well as the radionuclide specific weight coefficients for the hazard factor as input from the user. Based on these data and the cross-section data set of the JEREMY code [8], it computes the fluence conversion coefficients as defined in equation 3 and stores them either as F77 source code snippet for a dedicated FLUKA user routine or as PHITS multipliers in the PHITS input file format.

The code is written in the python programming language [12] and therefore automatically exposes a scripting interface. It has a very compact code base, consisting of 300 lines of code plus 1000 lines of code for the time evolution that is based on the JEREMY code, with decay data from JEFF 3.1.1 [13]. However, it is designed so that it can be easily interfaced with other cross-section data sets or time evolution engines.

The application of the fluence conversion coefficients during the Monte Carlo radiation transport simulation is achieved in FLUKA by a dedicated fluscw user routine that is then applied to a USRBIN scoring. In PHITS, the multipliers can be applied to a T-Yield tally. In both codes, the scoring of the radiological hazard factor is performed on a mesh geometry and is therefore not directly coupled to the region definitions of the simulation geometry.

4. Validation
A simulation of a proton beam hitting a copper target has been performed to verify the correctness of the fluence conversion coefficients method with respect to the FLUKA code. The copper target is a cylinder with a radius of 5 cm and a length of 80 cm. The target has been segmented in the simulation geometry into 4 parts of 20 cm length for scoring purposes. The proton beam with a momentum of 1 GeV/c is impinging onto the target at the center of the circular front face. The target is surrounded by a cylindrical-symmetric structure of air, concrete, cast iron and concrete. The layout of the simulation geometry is shown in figure 1.

Several radiological quantities have been scored with FLUKA as well as with the fluence conversion coefficients method in the copper target and the shielding structure. These quantities comprise saturation activities of selected radionuclides, their activities after certain irradiation and cool-down periods and multiples of the Swiss Exemption Limit, that is the sum of the specific activities weighted by radionuclide-specific exemption limits defined in the Swiss Radiation Protection Ordinance [14]. A comparison for these quantities is presented in table 2. The agreement is very good and differences larger than the statistical uncertainties can arise from the difference in the underlying radionuclide cross-section set in the used version of FLUKA (FLUKA2011 2c.5) and the ones used to compute the fluence conversion coefficients.

5. Examples and future applications
The capabilities of the fluence conversion coefficients method are demonstrated with several examples. This is followed by the discussion of promising future applications of the fluence conversion coefficients method.

5.1. Examples
The first example is the calculation of the multiples of the Swiss Exemption Limit for concrete after 10 years of irradiation followed by 2 years of cool-down for the validation geometry (see section 4) with the PHITS code. The results are shown in figure 2 for a beam intensity of 1 proton per second on the target.

The following examples show multiples of the Swiss Exemption Limit after 10 years of irradiation followed by 2 years for different materials for the CHARM facility [15] that is located in the CERN PS East Experimental Area. It receives a primary proton beam from the CERN
Figure 1. Cut through the cylindrical-symmetric simulation geometry used for the validation of the fluence conversion coefficients method with respect to the FLUKA code. The cylindrical-symmetric scoring volumes are indicated by the orange arrows.

Table 2. Comparison of several radiological quantities scored with FLUKA as well as with the fluence conversion coefficients method for validation purposes.

<table>
<thead>
<tr>
<th>Radiological Quantity</th>
<th>Material</th>
<th>Region</th>
<th>FLUKA</th>
<th>FFCs</th>
<th>FLUKA/FCCs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co-60 Production Yield</td>
<td>Copper</td>
<td>T1</td>
<td>1.98e-06±0.8%</td>
<td>1.536e-06±0.1%</td>
<td>1.29±0.81%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>T4</td>
<td>5.82e-09±14%</td>
<td>4.897e-09±2.1%</td>
<td>1.19±14%</td>
</tr>
<tr>
<td>Na-24 Production Yield</td>
<td>Concrete</td>
<td>Sh1</td>
<td>2.91e-09±3.3%</td>
<td>2.668e-09±0.4%</td>
<td>1.09±3.3%</td>
</tr>
<tr>
<td>Specific Activity (10y, 2d)</td>
<td></td>
<td>Sh3</td>
<td>8.62e-11±16%</td>
<td>8.688e-11±2.2%</td>
<td>0.924±16%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sh4</td>
<td>3.15E-10±3.3%</td>
<td>2.885e-10±0.4%</td>
<td>1.09±3.3%</td>
</tr>
<tr>
<td>Na-22 Specific Activity (10y, 2y)</td>
<td>Concrete</td>
<td>Sh1</td>
<td>1.20E-10±2.8%</td>
<td>1.443e-10±0.3%</td>
<td>0.832±2.8%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sh3</td>
<td>2.25E-12±16%</td>
<td>3.291e-12±1.8%</td>
<td>0.952±16%</td>
</tr>
<tr>
<td>Mn-54 Specific Activity (10y, 2y)</td>
<td>Cast Iron</td>
<td>Sh2</td>
<td>1.78e-10±1.5%</td>
<td>1.641e-10±1.7%</td>
<td>1.08±2.3%</td>
</tr>
<tr>
<td>Multiple of Swiss Exemption Limit (10y, 2y)</td>
<td>Concrete</td>
<td>Sh1</td>
<td>4.16e-12±5.7%</td>
<td>4.575e-12±0.9%</td>
<td>0.969±5.8%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sh3</td>
<td>4.16e-12±5.7%</td>
<td>4.575e-12±0.9%</td>
<td>0.969±5.8%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sh2</td>
<td>2.37e-10±2.9%</td>
<td>2.339e-10±0.5%</td>
<td>1.01±2.9%</td>
</tr>
</tbody>
</table>

* Production Yields in [nucl/(primary)]
Specific Activity in [(Bq/g)/(primary/s)]
Multiple of Swiss Exemption Limit in [1/(primary/s)]

PS with a beam momentum of 24 GeV/c that impinges on a copper or aluminum target. The beam is slowly extracted with a maximum pulse length of 350 ms and a maximum average design beam intensity of $6.7 \cdot 10^{10}$ protons per second. The layout of the CHARM facility is shown in figure 3 and a discussion of the radiation protection aspects of the CHARM facility can be found in [16].

The multiples of the Swiss Exemption Limit after 10 years of irradiation with $6.7 \cdot 10^{10}$ protons per second followed by 2 years of cool-down is shown for concrete in figure 4 and for cast iron in figure 5. These results can be used in the future during the decommissioning phase to establish
a zoning that identifies shielding structures that are expect to be activated below the Swiss Exemption Limits and that are therefore candidates for clearance from regulatory control.

Figure 2. Multiples of the Swiss Exemption Limit computed with the PHITS code for concrete after 10 years of irradiation followed by 2 years of cool-down for the validation setup described in section 4.

Figure 3. Integration drawing of the CHARM facility in the CERN PS East Experimental Area
**Figure 4.** Multiples of the Swiss Exemption Limit at beam line height computed with the FLUKA code for concrete after 10 years of irradiation followed by 2 years of cool-down for the CHARM facility described in section 5.1.

**Figure 5.** Multiples of the Swiss Exemption Limit at beam line height computed with the FLUKA code for cast iron after 10 years of irradiation followed by 2 years of cool-down for the CHARM facility described in section 5.1.
5.2. Future applications

The fluence conversion coefficients method is very general in its nature. These are some of the promising future fields of application of this method:

**Radioactive waste pre-characterization:** The shielding structures for new accelerator facilities are often designed using Monte Carlo radiation transport simulations. The radiological characterization of these structures, based on the fluence conversion coefficients method, can be already performed under conservative assumptions for the exploitation parameters. Estimates for the radioactive waste quantities as well as costs for its disposal can then be computed based on the obtained radiological characterization results.

**Clearance zoning:** It is often practical to establish a zoning for maintenance or decommissioning works to delimit the areas where waste might be cleared from regulatory control from areas where the waste will have to be treated as radioactive waste. This type of zoning might be established using the fluence conversion coefficients method for a quite extensive family of waste components.

**Earth activation:** The prediction of the activation of earth around accelerator tunnels might be required for radiation protection aspects of civil engineering works close to the tunnels or to prove the compliance with activation limits derived from water regulation. The fluence conversion coefficients method might be employed in both assessments.

**Planning of activation experiments:** One recurring task for the planning of activation experiments is the optimal selection of the masses for activation detectors for different materials, reaction channels and detector placement locations. A map of the spatial distribution of the saturation activities for a given material and reaction channel might be used to derive the optimal activation detector mass at various locations. This has to be done by taking also the irradiation and cool-down profile as well as the response of the gamma-spectrometry setup used for the activity measurements of the activation detectors after irradiation into account.

An example of a saturation activity distribution map is shown for the production of Na-24 in aluminum in the CHARM facility (described in section 5.1) in figure 6.

6. Summary

The fluence conversion coefficients method for radiological characterization has been presented based on the analysis of the capabilities of the standard methods for radiological characterization.

It is based on the reformulation of the often required weighted sum, with radionuclide specific weighting coefficients, of specific activities, also denoted as radiological hazard factor, as summation of the particle type dependent and energy dependent fluences with a set of fluence conversion coefficients. These fluence conversion coefficients can be precomputed for a given radiological characterization task and then applied on-line during the Monte Carlo radiation transport simulation. This method has been developed for both the FLUKA and PHITS Monte Carlo radiation transport codes.

The fluence conversion coefficients method is complementary to the standard methods for radiological characterization. Its main advantages are fast convergence, automatic mass normalization and good visualization capabilities for a large number of cells. The scoring of the radiological hazard factor can be performed on a mesh geometry and is therefore not directly coupled to the region definitions of the simulation geometry.

The correctness of the fluence conversion coefficients method and its implementation has been verified with respect to the FLUKA code. The capabilities of the fluence conversion coefficients method have been demonstrated by the example and promising future applications have been pointed out.
Figure 6. Saturation activity distribution of the production of Na-24 in aluminum at beam line height in the CHARM facility described in section 5.1.

The fluence conversion coefficients method provides very flexible, powerful and extensible tools for radiological characterization at accelerator facilities using Monte Carlo radiation transport simulations.

Acknowledgments
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