Spatial distribution sampling and Monte Carlo simulation of radioactive isotopes

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Abstract

This work focuses on the implementation of a program for random sampling of uniformly spatially distributed isotopes for Monte Carlo particle simulations and in specific FLUKA.

With FLUKA it is possible to calculate the radio nuclide production in high energy fields. The decay of these nuclide, and therefore the resulting radiation field, however can only be simulated in the same geometry. This works gives the tool to simulate the decay of the produced nuclide in other geometries. With that the radiation field from an irradiated object can be simulated in arbitrary environments.

The sampling of isotope mixtures was tested by simulating a 50/50 mixture of $^{137}$Cs and $^{60}$Co. These isotopes are both well known and provide therefore a first reliable benchmark in that respect. The sampling of uniformly distributed coordinates was tested using the histogram test for various spatial distributions. The advantages and disadvantages of the program compared to standard methods are demonstrated in the real life case of the CERF-Facility at CERN.
Radioactive Isotope Simulation

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1 Introduction

For radiation physics and radiation protection, simulation of particle transport and energy deposition, as well as radioactivity production and decay is an important tool to get an estimation on the risk and consequences of the operation of a high-energy beam facility.

In high-energy particle accelerators we have two basic types of radiation, prompt radiation from the particle beam (particle cascades created by the beam hitting material) and residual radiation from the decay of produced radioisotopes. The main occurrences of activation at particle accelerators can be found at fixed target experiments and other points of beam interception (beam collimators, beam dumps etc.). Due to the particle cascade development not only the intercepting elements get irradiated but also the material and environment around them gets activated.

Since the activation level and therefore the residual radiation in such parts can get very high, simulations are needed to forecast the radiological situation. On the one hand for safe access to these specific areas, on the other hand for handling the activated material properly. The first part also includes preventing inhalation of activated air.

Specifically in the case of activated air, simulations are also used to evaluate potential risks for the environment in the case of uncontrolled release.

For the high energy particle accelerators at CERN, the LHC and its pre-accelerators (LINAC, PSB, PS, SPS), the most used simulation tool for radiation physics is FLUKA. [1, 2] (At other institutions other programs are also used.)

"FLUKA is a general purpose tool for calculations of particle transport and interactions with matter, covering an extended range of applications spanning from proton and electron accelerator shielding to target design, calorimetry, activation, dosimetry, detector design, Accelerator Driven Systems, cosmic rays, neutrino physics, radiotherapy etc." [3]

For calculating the decay of radioactive isotopes FLUKA, although in general a very powerful tool, only provides the possibility to use single radioisotopes as radiation source, whereas in reality mixtures of different isotopes as well as various spatial distributions are required to accurately simulate real-life situations.

Another important factor is the simulation of radioactive materials taken out of the experimental area and put in the laboratory. Here FLUKA is limited due to the fact that geometry cannot (easily) be changed during the simulation.

A typical example is the irradiation of a target over a period of time, during which it gets damaged by the beam. In order to continue experiments the target needs to be repaired. That is only possible in a laboratory. For safety reasons it is necessary to forecast the dose impact in the lab and its environment. This is not easily possible with current tools.

This work is aimed on addressing these issues.
2 Random Sampling Theory

Most of the general (pseudo) random number generators in computer simulations follow a uniform distribution on the interval \([0, 1)\). But when sampling coordinates in 3 dimensions, in non-Cartesian coordinate systems, mathematical methods are needed to ensure the correct transformation of the probability density functions (pdf). In the case of uniform spatial distributions this means that, uniformly independent distributed coordinates do not necessarily give a uniform spatial distribution in different coordinate systems. (For example see Section: 4.1)

To obtain uniformly distributed points in a given geometry and coordinate system, marginalization, the Bayes’ theorem of conditional probability, and the Inverse Transformation Method was used as explained below.

2.1 Sampling in arbitrary geometries and coordinate systems

Mathematically it is very convenient to use coordinate systems that best fit the actual geometries (spherical coordinates for spheres/ellipsoids, cylindrical coordinates for cylinders, etc.)

To illustrate the method used to sample the positions of the isotopes, an example of uniformly distributed point in a sphere or radius R is given below.

We start with the pdf of a uniform distribution in 3 dimensional Cartesian coordinates:

\[ p(x, y, z) = c \]

Now we want to use Spherical coordinates:

\[
\begin{align*}
    x &= r \sin \theta \cos \phi & r & \in [0, R] \\
    y &= r \sin \theta \sin \phi & \theta & \in [0, \pi] \\
    z &= r \cos \theta & \phi & \in [0, 2\pi]
\end{align*}
\]

For the coordinate transformation of the volume element with the Jacobi determinant we get,

\[
dV = dx \, dy \, dz = \left| \frac{\partial x}{\partial r} \frac{\partial x}{\partial \theta} \frac{\partial x}{\partial \phi} \right| \cdot \cdot \, dr \, d\theta \, d\phi = r^2 \sin(\theta) \, dr \, d\theta \, d\phi
\]

and therefore for the transformed pdf:

\[
\begin{align*}
    p(x, y, z) &= c \\
    p(r, \theta, \phi) &= r^2 \sin(\theta) \, p(x, y, z) \\
    &= r^2 \sin(\theta) \cdot c
\end{align*}
\]

Now the pdf needs to be normalized to determine the constant c:

\[
\begin{align*}
    \int_V p(r, \theta, \phi) \, dV &= 1 \\
    c \cdot \int_0^R \int_0^\pi \int_0^{2\pi} r^2 \sin(\theta) \, dr \, d\theta \, d\phi &= 1 \Rightarrow c = \frac{3}{4\pi R^3}
\end{align*}
\]
The normalized pdf in spherical coordinates is then:

\[ p(r, \theta, \phi) = \frac{3r^2 \sin(\theta)}{4\pi R^3} \]

To sample the coordinates we need to isolate them in the pdf. First the number of variables is reduced by one using marginalization:

\[ p_\phi(r, \theta) = \int_0^{2\pi} \frac{3r^2 \sin(\theta)}{4\pi R^3} \, d\phi = \frac{3r^2 \sin(\theta)}{2R^3} \]

With the conditional probability a single variable can now be isolated [4, p. 32f]:

\[ p(\phi | r, \theta) = \frac{p(r, \theta, \phi)}{p_\phi(r, \theta)} = \frac{1}{2\pi} \]

Note: In this case marginalization of all three variables would be possible since they are independent of each other. However if they are not independent the conditional probability must be used to get the correct result.

We do the last two steps again to get the other variables:

\[ p_\theta(r) = \int_0^\pi p_\phi(r, \theta) \, d\theta = \frac{3r^2}{R^3} \]

\[ p(\theta | r) = \frac{p_\phi(r, \theta)}{p_\theta(r)} = \frac{\sin(\theta)}{2} \]

With these pdfs, which only depend on one variable, the Inverse Transformation Method can now be used to get the desired random number distribution of the coordinates.

### 2.2 The Inverse Transformation Method

The inverse transformation method [5, p. 189f] is used to sample random numbers from an arbitrary pdf. Since the three pdf obtained in the last section are not uniformly distributed any more, this method is used to sample the corresponding random numbers from uniformly distributed random numbers. Due to the form of this method the corresponding cumulative distribution function (cdf) of the pdf needs to be invertible.

The pdfs are integrated to the corresponding cdfs and inverted:

\[
P_r(r) = \int_0^r \frac{3r^2}{R^3} \, dr = \frac{r^3}{R^3} \quad P_\theta(\theta | r) = \int_0^\theta \frac{\sin(\theta)}{2} \, d\theta = \frac{1-\cos(\theta)}{2} \quad P_\phi(\phi | r, \theta) = \int_0^\phi \frac{1}{2\pi} \, d\phi = \frac{\phi}{2\pi} \]

\[ r = P_r^{-1}(\xi_1) \text{ with } \xi_1 \in [0, 1] \quad \theta = P_\theta^{-1}(\xi_2) \text{ with } \xi_2 \in [0, 1] \quad \phi = P_\phi^{-1}(\xi_3) \text{ with } \xi_3 \in [0, 1] \]

\[ r = \sqrt[3]{\xi_1 \cdot R} \quad \theta = \cos^{-1}(1 - 2\xi_2) \quad \phi = \xi_3 \cdot 2\pi \]

With these random number dependencies, uniformly distributed points in a sphere can be sampled.
3 Isotope Decay Simulation

3.1 Methods of decay simulation in FLUKA

Radioactive isotopes decay with certain half-lives and have one or more so called decay channels. Most of the time a radioactive isotope decays into another radioactive isotope, leading to the creation of decay chains which end in stable isotopes. (See Figure 8 and 9) There are two methods implemented in FLUKA to simulate these decays and chains; the method based on the direct solution of the Bateman Equations and a so called (semi)-analog method that treats decays as Monte Carlo particles.

The Bateman Equations give an exact analytical solution for the decay and activity of a given isotope and its daughter products at the desired time.

\[
N_i(t) = \sum_{i=1}^{n} \left[ N_i(0) \times \left( \prod_{j=i}^{n-1} \lambda_j \right) \times \left( \sum_{j=i}^{n} \left( \frac{e^{-\lambda_j t}}{\prod_{p=i,p \neq j}^{n} (\lambda_p - \lambda_j)} \right) \right) \right]
\]

With \( N_i(t) \) being the number of isotopes of type \( i \) at time \( t \), and \( \lambda_i \) the corresponding decay rate of these isotopes to decay to isotopes of type \( i + 1 \).

In the (semi)-analog way, isotopes are treated the same as primary particles. They are sampled randomly according to their specific probability to decay in one chain or the other. Also the decay time of each particle is sampled randomly. The decay chains are followed until only stable isotopes are left. Therefore no time resolution is available with this method.

Currently in FLUKA the Bateman implementation only works for isotopes that are produced as secondary particles (i.e. through activation from a beam impact). If isotopes are started as a source only the (semi)-analog mode is available.

3.2 Simulation of a mixture of isotopes in FLUKA

Another limitation in FLUKA is that normally only one type of isotope can be used as a source per simulation. In reality in most cases a mixture of different isotopes is present. To use more than one isotope a so called user-routine needs to be programmed to provide FLUKA with the start terms for the isotope mix. To achieve the correct mix of isotope decays the sampling of the single isotopes was set in relation to their respective activity. The more active the isotope is, the higher the probability that it is set as a start particle in FLUKA. (See section: 4.2)


### 3.3 Units and Normalization in FLUKA

When simulating in FLUKA the results are usually given in units “per primary weight” unless special scoring options are used (See section: [3.4]). In the case of radioactive isotope simulation this means “per decay” since every started primary particle is essentially a decay. To normalize these results into a more useful unit the way FLUKA usually normalizes results needs to be changed. In FLUKA normalization happens with two variables: WTFLK and WEIPRI. WTFLK is the weight of one single particle and is usually set to 1. WEIPRI is the cumulative weight of all primaries:

$$WEIPRI = \sum_i WTFLK(i)$$

With these two variables FLUKA builds the weighted mean of the obtained results $X_i$:

$$\frac{\sum_i X_i WTFLK(i)}{WEIPRI}$$

A change in normalization can only be achieved with these two variables. To normalize the result to units “per second” the total activity ($A$) of the source has to be multiplied with the result:

$$\frac{Unit_{result \, decay}}{decay} \cdot A = \frac{Unit_{result \, decay}}{decay} \cdot \frac{decays}{s} = \frac{Unit_{result \, decay}}{s}$$

To achieve this WEIPRI was set to $\sum_i \frac{WTFLK(i)}{A}$ so weighting of the results can still be applied while normalization to “units per second” is enforced:

$$\frac{\sum_i X_i WTFLK(i)}{\sum_i \frac{WTFLK(i)}{A}}$$

### 3.4 Scoring in FLUKA

In FLUKA results are obtained by adding so called detectors to the input file. A detector is the Monte Carlo equivalent of a measurement instrument. Each detector type is designed to estimate one or more quantities. The final result is then the average value of the corresponding population. As in experimental measurements, it is possible to calculate a standard deviation by running several independent calculations. [6, Section 7.17]

No default detector is available since each scoring is dependent on the simulation setup. Therefore also the units of scored quantities depend on the setup. As explained above these can either be “per primary weight” or for example “per second”.

When using the “online” simulation method for radioactive decays in FLUKA, when radioisotopes occur as secondary particles through activation from a beam, usually an irradiation profile and cooling times are given and the result is in “units per second”. (e.g. pSv/s or Bq)

With the “offline” method, i.e. when starting radioisotopes as primary particles, only the (semi)-analog mode is available. This influences the scoring by altering the unit obtained as mentioned above. Also it is not possible to use cooling times in this mode, therefore the simulation has to be altered to get desired time dependent results.
4 Validation of sampling methods

4.1 Validation of Spatial Distribution sampling

As explained in chapter 2 it is necessary to transform the random numbers for spatial sampling according to the geometry and coordinate system.

Before validating the correctness of the coordinate dependencies obtained in section 2.2 an example is given why it is important to use special methods to sample the points. In this example the coordinates are sampled as follows:

\[
\begin{align*}
x &= R \cdot \xi_1 \sin(\pi \cdot \xi_2) \cos(2\pi \cdot \xi_3) \\
y &= R \cdot \xi_1 \sin(\pi \cdot \xi_2) \sin(2\pi \cdot \xi_3) \\
z &= R \cdot \xi_1 \cos(\pi \cdot \xi_2)
\end{align*}
\]

So the random numbers \(\xi_{1,2,3}\) are just plugged in with the boundaries for the coordinates. (See example in section 2.1) The graphical representation is given is Figure 1.

![Randomly sampled points in a sphere without using a special method to obtain the coordinates.](image)

Figure 1: Randomly sampled points in a sphere without using a special method to obtain the coordinates.

The resulting distribution is not uniform, this can be seen with the free eye. Now the results from section 2.2 are used to sample the same example as before:

\[
\begin{align*}
x &= R \sqrt[3]{\xi_1} \cdot 2\sqrt[3]{\xi_2} (1 - \xi_2) \cos(2\pi \cdot \xi_3) \\
y &= R \sqrt[3]{\xi_1} \cdot 2\sqrt[3]{\xi_2} (1 - \xi_2) \sin(2\pi \cdot \xi_3) \\
z &= R \sqrt[3]{\xi_1} \cdot (1 - 2\xi_2)
\end{align*}
\]

The visualization can be seen in Figure 2.
The resulting distribution (Figure 2) looks a lot more uniform than before (Figure 1). To quantify the quality and correctness of the distribution the histogram-test is used.

4.1.1 The Histogram-Test

The histogram-test is one of the easiest ways to check whether a distribution is correct or not. For the used example of a uniform distribution a horizontal line is expected, meaning that all bins are equally filled. One important thing concerning the histogram test is the correct choice of bins. In the used example it would be wrong to use equidistant radial bins (Figure 3). The result with this wrong bin spacing can be seen in Figure 4 and it is clear that this does not resemble a uniform distribution. The reason for that is that in the example uniformly distributed points in the volume of a sphere were sampled, meaning that in the same amount of volume should be the same amount of points. The right choice are equisized volume bins (Figure 5) which give the result in Figure 6.

Figure 2: Randomly sampled points in a sphere with special sampling methods.

Figure 3: Schematic of the sphere shells for the binning of the histogram in Figure 4.
Figure 4: Histogram with bins that represent sphere shells with the same thickness.

Figure 5: Schematic of sphere shells for the binning of the histogram in Figure 6. The volume in every shell is the same.
Figure 6: Histogram with bins that represent sphere shells with the same volume.

In Figure 6, the quality of the method used can be determined. The bins are filled equally except for some small statistical error which is expected.

In Figure 7, a sampling of all available geometries is shown.

Figure 7: Uniformly distributed random points for all available simple geometries.
4.2 Validation of Isotope Mixture sampling

To test the quality of the isotope sampling, a sphere containing 50% $^{137}\text{Cs}$ and 50% $^{60}\text{Co}$ was simulated. The electron- and $\gamma$-Spectrum are scored in binned histograms. In Figure 10, the probability per decay for electrons is shown. One can see the background from $^{60}\text{Co}$ and $^{137}\text{Cs}$ as well as specific Auger electron from the $^{137}\text{Ba}_{m}$ to $^{137}\text{Ba}$ decay (at 25.5 keV and 4.5 keV mean energy of the bin). The other peaks correspond to electron-emissions from different shells (M,L and K). In Figure 11 the gamma spectrum is plotted. The $^{137}\text{Ba}_{m}$ to $^{137}\text{Ba}$ $\gamma$-line at 662 keV and the two characteristic $^{60}\text{Co}$ $\gamma$-lines at 1174 keV and 1334 keV are the dominant peaks. The $^{60}\text{Co}$-lines have exactly a 50% probability as expected. The probability for the $^{137}\text{Ba}_{m}$-line is smaller because when looking at the decay chain of $^{137}\text{Cs}$ one can see that the $\gamma$-line has only a probability of 85% per decay. (Figure 8 and 9)

Figure 8: Decay diagram for $^{137}\text{Cs}$, Source: [9]

Figure 9: Decay diagram for $^{60}\text{Co}$, Source: [9]
Figure 10: Electron spectrum of a simulated 50% $^{137}$Cs and 50% $^{60}$Co source.
Figure 11: Gamma spectrum of a simulated 50% $^{137}\text{Cs}$ and 50% $^{60}\text{Co}$ source.
5 Case Study: CERF

5.1 The CERF Facility

"The CERN-EU high-energy Reference Field facility (CERF) provides a reference neutron field of characteristics similar to the field encountered at commercial flight altitudes (10 - 20 km), produced by cosmic rays interacting with the atmosphere. CERF provides a reference base for testing, intercomparing and calibrating passive and active instruments before their use on-board aircraft and in space." [10]

To simulate these radiation fields a beam hits a copper target in a shielded area (See Figure 12). The experiments are usually carried out over an extended period of time.

5.2 Simulation conditions

In this particular case an irradiation of a cooper target of 0.5 cm radius and 20 cm length is simulated. The irradiation profile consists of 1 week irradiation with a 120 GeV proton beam with an intensity of $8 \cdot 10^6$ particles, followed by 24h of cooldown over a total amount of 6 weeks. After another cooldown of 1 week the target is transported to the laboratory.

5.3 Steps of the simulation

First the irradiation is simulated in the full CERF geometry (Figure 12). The activities of the residual nuclei (in Bq), produced in the target, are scored after the cooldown period of 1 week. This output is then parsed into a readable format by the source routine. In this step the weighted average and statistical error of the activities, from several simulation runs, are calculated. This isotope inventory is then used in the laboratory geometry (Figure 13) as a source for radioactive decay. Now all required quantities can be scored in the lab geometry.
Figure 12: Full CERF geometry. Note: part of the shielding around the target is not shown so the target can be seen.

Figure 13: Full laboratory geometry. Note: the roof has been removed to see the inside.
5.4 Comparison to standard FLUKA methods

Because the geometry is not easily interchangeable in FLUKA, a case like the one described above can only be roughly estimated. This has the drawback of not having any spatial informations of the final geometry. Another way would be the simulation of a source consisting only of the main contributing isotope. This is a very rough estimate since small quantities of different isotopes can have a huge impact on the overall effective dose.

Figure 14 shows the comparison of the dose-equivalent (dose-eq) simulated with FLUKA in CERF and with the source routine in the LAB. As one can see around the target itself the dose-eq is the same within a small range of error. The farther one goes away from the target, the bigger the difference gets. This is exactly as expected, since the concrete walls also contribute to the overall dose-eq in the CERF geometry.

Figure 14: Ratio between the scored dose-eq of the FLUKA run and the source routine

The advantage of the source-routine over standard FLUKA methods can be seen in Figure 15. The shielding of the table can only be estimated with the source -routine in the lab geometry.
Figure 15: Spatial geometry information. **Left:** Standard FLUKA methods, no shielding from the table. **Right:** Source-routine, shielding of the table in the new geometry is considered.

5.5 Results for the Laboratory geometry

Figure 16 shows a complete dose map for the laboratory geometry. One can see the shielding effects of the walls and the doors. Also, an estimation for the dose outside the laboratory can be made. In this case the dose-eq is not higher than a few nSv/h in the hallway. Therefore no extra shielding is required. To get the same result with standard FLUKA methods is highly complicated.

Figure 16: Complete dose map for the laboratory geometry.
5.6 Error Estimation

In reality the produced isotopes in a target are not uniformly distributed, but have a shape that follows the build-up which depends on the beam direction and the target geometry. For targets that are small compared to the cross section of the beam, i.e. targets that are fully hit by the beam, the uniform distribution is a good approximation. In order to get a good estimate it is necessary to simulate an appropriate number of particles/decays to minimize the statistical error.

6 Conclusion

In this work a FLUKA source routine was written for simulation of randomly spatially distributed radioactive isotopes. The well known isotopes $^{137}$Cs and $^{60}$Co were simulated to test the sampling of a mixture of isotopes. The spatial distribution was tested using SimpleGeo as a visualization tool, and finally a use-case scenario was simulated for the CERF-Facility.

As seen in this work the spatial information is very important in making estimations for shielding and dose rate. This cannot be achieved using standard FLUKA methods. The second big advantage is the arbitrary mixture of isotopes. Since isotopes are usually always found in mixtures, this makes the simulation more similar to the real world scenario. The combination of these two features has numerous use-cases. One is the presented case of moving a irradiated object into another geometry, another important use-case is for example air activation.

6.1 Outlook

Of course there are some ways to further improve the program to get better estimations. The two main points are the implementation of the Bateman equations instead of the analogous approach. This would give an exact solution for the decay of the isotopes. The other, maybe even more crucial point, is the correct simulation of the spatial distribution of isotopes. A possibility would be to score the spatial information when scoring the isotopes. The disadvantage is the huge amount of data that comes with this approach.
References


