Hybrid Monte Carlo Method for Standard Source Geometry

3.1 Introduction
3.2 Attenuation Correction Factor
   3.2.1 Measurement of linear attenuation coefficient (μ)
   3.2.2 Calculation of attenuation correction factor (k_{att})
   3.2.3 Aim of this work
3.3 Hybrid Monte Carlo Method
3.4 Programs
   3.4.1 Cylindrical geometry
   3.4.2 Box geometry
   3.4.3 Spherical geometry
   3.4.4 Disc geometry
3.5 Validation of the Hybrid Monte Carlo approach
   3.5.1 Theoretical validation
   3.5.2 Experimental validation
3.6 Conclusion
3.1 Introduction

As discussed in Chapter 1, the gamma rays while passing through a medium may interact by any of the three interaction modes and may be scattered or absorbed in the medium. This can be more clearly understood by considering a cylindrical sample placed on the detector surface as shown in Figure 3.1. Due to the voluminous nature of the sample, a fraction of the photons generated in the sample volume may interact in the sample itself as shown in the figure. This results in loss of photons in the sample and hence reduction in the number of photons that would have reached the detector in the absence of such interaction. The probability of such interaction becomes important as the sample becomes more and more voluminous or the density of the sample increases. As emphasized in Chapter 1, the net effect is the loss of proportionality between the measured count rate of a radionuclide and its amount in the sample. The use of calibration standard is difficult due to diversity of sample matrix encountered in NDA [Reilly et al. (1991)] and the problem can only be solved by correcting the measured count rate for self-attenuation as given in equation 1.7.

Figure 3.1 Possible gamma ray interactions within the sample volume before reaching the detector.
3.2 Attenuation Correction Factor

The attenuation correction factor \(k_{\text{att}}\) is defined as:

\[
k_{\text{att}} = \frac{RR(\mu = 0, \text{specified shape})}{RR(\mu \neq 0, \text{real shape})}
\]  

(3.1)

where \(RR(\mu = 0, \text{specified shape})\) is the count rate for totally non-attenuating \((\mu = 0)\) sample having a specified shape. This specified shape may not necessarily be the actual shape of the sample and can be approximated to a simplified shape like a point or a line depending upon the sample-detector configuration. \(RR(\mu \neq 0, \text{real shape})\) is the measured count rate for the sample. For computational purpose, the \(k_{\text{att}}\), as defined above, can be written as [Parker (1991)]:

\[
k_{\text{att}} = \frac{\int \rho I \varepsilon(r) dV}{\int \rho I \varepsilon(r) \exp(-\mu t(r)) dV}
\]  

(3.2)

where \(\rho\) = spatial density of the isotope being assayed (g/cm\(^3\)),

\(I\) = emission rate of the assayed gamma ray (\(\gamma\)/g-s),

\(\varepsilon\) = absolute full energy detection efficiency,

\(\mu\) = linear attenuation coefficient of the sample,

and \(t\) = distance the gamma ray travels within the sample.

The integration is to be performed over the volume of the sample. From equation 3.2, it can be seen that, for obtaining \(k_{\text{att}}\), linear attenuation coefficient of sample matrix is required as an input. The measurement of attenuation correction factor is a two step process. First step is to obtain linear attenuation coefficient of the sample under study.
The next step is to choose an appropriate model to obtain the sample-detector geometry dependent attenuation correction factor. The two steps involved are discussed in detail below.

3.2.1 Measurement of Linear Attenuation Coefficient ($\mu$)

The linear attenuation coefficient ($\mu$) describes the fraction of a beam of X-rays or gamma rays that are absorbed or scattered per unit thickness of the absorber. For a narrow beam of monoenergetic photons (Figure 3.2), the change in beam intensity ($dI(x)$) while passing through a small thickness ($dx$) of the absorber can be expressed as:

$$dI(x) = -I(x)n\sigma dx$$  \hspace{1cm} (3.3)

Where, $n$ is the number of atoms/cm$^3$ and $\sigma$ is the proportionality constant and is a measure of the probability of gamma ray interaction per absorber atom.

![Diagram showing transmission of a gamma ray through an attenuating medium](image)

**Figure 3.2** Transmission of a gamma ray through an attenuating medium present in between the source and the detector.

On integrating this equation over the thickness of the absorber,

$$I = I_o \exp(-n\sigma x)$$  \hspace{1cm} (3.4)
where, $I$ and $I_o$ are the intensities of gamma ray with and without the absorber, $x$ is the distance traversed by the gamma ray through the absorber. The number of atoms/cm$^3$ ($n$) and the proportionality constant ($\sigma$) are usually combined to yield the linear attenuation coefficient ($\mu$). Therefore the equation becomes:

$$I = I_o \exp(-\mu x)$$

(3.5)

The linear attenuation coefficient determines the mean free path ($\lambda$) of a photon in a medium i.e. $\lambda=1/\mu$ where, $\lambda$ characterizes the average distance a photon will traverse before undergoing a collision in a medium. If there is a precise knowledge of the composition and density of the sample, the linear attenuation coefficient of the sample can be obtained theoretically by taking the literature mass attenuation coefficient ($\mu_m$) at the monitored energy for the given medium. The $\mu_m$ values for different matrices have been compiled in Hubbell (1982). Due to the diversity of samples encountered in NDA work, these details of sample composition and density may not be always available. Thus it is a common practice to measure $\mu$ experimentally by transmission technique. The experimental set-up for this is already shown in Figure 3.2. Here, the transmission of a collimated beam of gamma ray from an external source is measured with and without the sample. Then, the linear attenuation coefficient is obtained using equation 3.5. The choice of external source is important and depends upon the radionuclides monitored. In principle, one should use a transmission source that emits a gamma ray of energy as close as possible to the energy of the gamma ray emitted by the sample and should be preferably monoenergetic.
3.2.1 Calculation of Attenuation Correction Factor \((k_{att})\)

Once the \(\mu\) of the sample is known, one has to choose an appropriate model to obtain the sample-detector geometry dependent attenuation correction factor. This is done by simplifying the general equation 3.2. The detector in this case is considered to be a point detector. For homogeneous sample, the factor \(\rho I\) will remain constant in both numerator and denominator and will cancel out. Since the parameters \(\varepsilon\) and \(t\) are function of the co-ordinate of the point at which gamma ray originates within the sample, they cannot be taken out of the integral and hence they will not cancel out. For majority of geometrical configurations, the integrals cannot be evaluated analytically, and numerical integration is required. There is extensive literature on evaluation of \(k_{att}\) for different sample shapes [Moens et al. (1981), Debertin and Jianping (1989), Chilton et al. (1984), Croft (1993), Dixon (1951), Dickens (1972)]. Based on sample-to-detector distance, two general approaches have evolved.

(I) Far-field Geometry

A great simplification occurs, if \(\varepsilon\) is considered to be independent of position within the sample. This idealization is practically realized if the distance between sample and detector is large enough so that the gamma rays reaching the detector are essentially parallel and \(\varepsilon\) can be assumed to be independent of point of origin of the gamma ray within the sample and can be taken out of integrals. Then \(\varepsilon\) in the numerator and denominator get cancelled and the integration can be performed over the sample volume. This makes the integrals relatively easy to compute. In such a case, the size of the sample can be considered to be negligible compared to the source-to-detector distance. This
situation is called far-field geometry and $k_{att}$ in such a case becomes independent of the
detector size, shape and sample-to-detector distance and is only influenced by the sample
characteristics like its size, shape and matrix. For far-field cases, analytical expressions
are available in literature for some common sample geometries [Croft et al. (2005)] viz.
box-shaped (rectangular parallelepiped), cylindrical [Owen and Fage (1921)] and
spherical [Francois (1974), Thirring (1912)].

(II) Near-field Geometry

In practice, it is not always possible to count the sample far away from the detector. For
example, in environmental samples, activity is usually low, and the sample is required to
be counted close to the detector to maximize the count rate. In such a case, the far-field
formulae cannot be applied as they assume detector to be a point detector and $\varepsilon$ is
assumed to be independent of point of origin of gamma ray in the sample. In literature,
separate numerical models are available for calculating attenuation correction factors for
the near-field case. These models are based on certain assumptions to simplify the
sample-detector geometry. For example, in $k_{att}$ calculation for cylindrical samples in near-
field geometry [Parker (1991)], only the circular cross-section of the sample is considered
and the detector is assumed to be a point detector. Moreover, there are restrictions about
sample-detector dimensions.

Gunnink and Niday (1972) and Cline (1978) developed semi-empirical methods to find
the peak efficiency of co-axial Ge(Li) detectors including implicit or explicit corrections
for gamma ray attenuation. Points, disks, cylindrical and spherical sources were
considered. These techniques, however, are based on approximating mathematical
models, representing the detector [Gunnink and Niday (1972), Cline (1978)] and the source [Gunnink and Niday (1972)] as physical points. Aguiar et al. (2006) and Aguiar (2008) also proposed an analytical approach for cylindrical geometry to get full energy peak efficiency taking into account the self-attenuation in the sample. The approach assumes detector as a point inside the detector volume with its position determined empirically using point sources.

A semi-empirical procedure to calculate absolute detector efficiency for a point, disc and cylindrical samples has also been described by Moens et al. (1981). The method takes care of gamma ray attenuation within the sample and in any interjacent absorbing layer. The method has been devised for cylindrical geometry including co-axial Ge(Li) detectors and is free of any simplification in the sample-detector geometry. The peak efficiency of any investigated configuration (ε_{p,x}) is calculated from the measured efficiency of a reference point source (ε_{p,ref}) positioned on the detector axis at large distance from the detector using the formula:

\[ ε_{p,x} = ε_{p,ref} \frac{Ω_x}{Ω_{ref}} \] (3.6)

where \( Ω_{ref} \) and \( Ω_x \) are the effective solid angles of the point source and the sample respectively. \( Ω_{ref} \) takes into account the attenuation within the sample. The effective solid angle is related to the peak efficiency (ε_p) as:

\[ ε_p = \frac{1}{4π} \frac{P}{T} \frac{1}{Ω} \] (3.7)

where \( P/T \) is peak to total ratio for the detector for the given gamma ray. The method involves numerical integration for the evaluation of \( Ω_x \), and requires detector dimensions.
as input. Debertin and Jianping (1989) proposed a point detector model for the calculation of detector efficiency from attenuating samples.

Alternative to all these approaches is a full scale Monte Carlo calculation of $k_{att}$ [Sima and Arnold (1996), Sima (1996), Pérez-Moreno et al. (2002), Vargas et al. (2002)]. The advantage of this approach is that it can reproduce any sample-detector geometry and minimize experimental work. The limitation of this method is that it can be used only when the bulk density and the elemental composition of the samples are known accurately. It also requires a detailed description of sample-detector geometry. Monte Carlo calculation of $k_{att}$ is computationally very time consuming.

Recently, a hybrid Monte Carlo method has been used to calculate detector efficiency [Yalcin et al. (2007)] and self-attenuation correction factors [Badawy et al. (2000)] where analytical expressions are used in Monte Carlo simulation. This leads to considerable reduction in computation time. García-Talavera and Peña (2004) also proposed a Hybrid approach that combines the experimental measurements and MC simulations can be applied to any unknown composition.

### 3.2.3 Aim of this Work

From the above discussion, it can be concluded that although there are several approaches available in the literature but none is valid at all sample-detector geometries. Also it is not easy to decide which formula to use as it not only depends upon the sample-to-detector distance but also on the sample-detector dimensions. For eg. a distance of 10 cm may be a far-field for 20 ml vial but not for a 200 L drum. Therefore, in spite of so many approaches available in literature, for laboratories where attenuation correction is a part
of routine analysis, a simple, convenient method is still very much required which can be applied without bothering much about sample-detector geometry.

In this work, a Hybrid Monte Carlo (HMC) method has been developed for obtaining attenuation correction factors. This method avoids any simplified assumption used for near or far-field geometry formulae. Moreover, unlike the far-field and near-field approaches, the detector is not considered to be a point detector. This method does not require detailed information about detector dimensions excepting the radius of the detector crystal (considered to be a cylinder). In the present work, this method has been used to obtain attenuation correction factors for the three sample geometries i.e. cylinder, sphere and box at different sample-to-detector distances as a function of transmittance and these results are compared with the analytical expressions available in the literature. MCNP calculations [Briesmeister (2000)] have also been performed to validate the prediction of our calculation for samples of varied transmittance and sample-to-detector distance. This method has been further validated by comparing the results of calculation with the experimentally obtained $k_{att}$’s. The experiment was done for three sample geometries – 1) cylindrical sample with axis collinear with the detector axis (disc geometry), 2) cylindrical sample with axis perpendicular to detector axis (cylindrical geometry) and 3) box shaped sample. In all the cases, the centre of gravity of the sample was fixed on the detector axis. All the three geometrical set-ups are shown in Figure 3.3.
3.3 Hybrid Monte Carlo Method

In equation 3.2, as already emphasized above, all the factors cancels out except $\varepsilon$ and the exponential term. As mentioned before, $\varepsilon$ is the efficiency of the detector for particular sample-detector geometry and so is inversely proportional to $1/r^2$ where $r$ is the distance the gamma ray travels from the point of origin in the sample to the detector surface. So, attenuation correction factor for a particular path followed by a gamma ray can be simplified as:

$$k_{att} = \frac{1/r^2}{\exp(-\mu t)/r^2}$$

(3.8)

where $t$ is the distance the gamma ray travels in the sample. The exponential term in the denominator corrects for the gamma ray attenuation in the sample. Both $\varepsilon$ and $t$ will vary depending upon the point of origin of the gamma ray in the sample and also on its path followed to reach the detector. In the case of an attenuating container wall, an additional
factor in equation 3.8 in the exponential will come ($\mu c t c$, where $\mu$ and $t_c$ are the linear attenuation coefficient and path length of the gamma ray in the sample wall respectively).

As demonstrated below, $t_c$ can be found out by the same procedure as $t$. Two representative paths of gamma ray originating in a cylindrical sample whose axis is perpendicular to the detector axis are shown in Figure 3.4. Consider the origin is fixed at the centre of gravity of the sample, which lie on the detector and sample axis. Suppose a random point is generated within the sample having co-ordinate $(a, b, c)$ and another random point $(a1, d, c1)$ on the detector surface which is at a distance $d$ from the origin. Suppose the line connecting $(a, b, c)$ and $(a1, d, c1)$ intersect the sample cylinder at $(x, y, z)$. Then $r$ and $t$ for this representative path is given by:

$$r = \sqrt{(a1 - a)^2 + (d - b)^2 + (c1 - c)^2}$$  \hspace{1cm} (3.9)

$$t = \sqrt{(x - a)^2 + (y - b)^2 + (z - c)^2}$$  \hspace{1cm} (3.10)

Figure 3.4 Representative paths of two gamma rays from sample to the detector surface.
From equation 3.9, \( r \) can be calculated since the point of gamma ray origin and the point at which it reaches the detector is known. But for calculating \( t \), the co-ordinate of the point of intersection \((x, y, z)\) of the line connecting \((a, b, c)\) and \((a_1, d, c_1)\) and the sample cylinder need to be known. The procedure for calculating \((x, y, z)\) is given below. The equation of the straight line joining \((a, b, c)\), \((x, y, z)\) and \((a_1, d, c_1)\) is given by:

\[
\frac{x-a}{a_1-a} = \frac{y-b}{d-b} = \frac{z-c}{c_1-c}
\]  

(3.11)

The value of \((x, y, z)\) can be obtained from the point of intersection of this straight line and the circle defined by the equation

\[
x^2 + y^2 = r_{\text{sample}}^2
\]

(3.12)

where \(r_{\text{sample}}\) is the radius of the cylindrical sample. Similarly, \(r\) and \(t\) can be obtained for any pair of points lying on the detector surface and in the sample volume. In the present numerical approach, a large number of pair of random points are generated which are lying on the detector surface and in the sample volume, the actual \(k_{\text{att}}\) for the sample will be given by the following equation:

\[
k_{\text{att}} = \frac{\frac{1}{n} \sum_{i=1}^{n} 1/r_i^2}{\frac{1}{n} \sum_{i=1}^{n} \exp \left(-\mu t_i/r_i^2\right)}
\]

(3.13)

\[
= \frac{<1/r_i^2>}{<\exp \left(-\mu t_i/r_i^2\right)>}
\]

(3.14)
where <> sign represents the average of the quantity concerned. The approach is hybrid since Monte Carlo principle is combined with the analytical method.

3.4 PROGRAMS

In order to calculate attenuation correction factors, separate FORTRAN programs were written for different sample geometries. The description of the programs written for common sample geometries is given below:

3.4.1 Cylindrical Geometry

For cylindrical geometry, the attenuation correction program ‘CYL.FOR’ is given as program 1 in the Appendix II of this thesis. In this program, the linear attenuation coefficient (amu or μ) is kept fixed and transmittance of the gamma ray is given as an input in 'transm.txt'. The input of the program can be changed as required. An example of the input file is given in the input as Input 1 in the Appendix II of this thesis. This file gives the list of transmittance of the gamma ray for which $k_{att}$ values are to be calculated. The radius of the cylindrical sample corresponding to the given transmittance values given are calculated in the program itself. The diameter of the detector surface, which is an input in the calculations, was given as 5 cm. This corresponds to the manufacturer supplied diameter of the detector used in the experiments. The length of the crystal does not enter in the calculations. The function ‘aphasa’ is used to generate random numbers in both the sample volume and on the detector surface. For example, for a cylindrical sample (with radius and half-height of the cylinder as $r$ and $h$ respectively), three random
numbers \((a, b, c)\) are first generated within a cuboid of dimensions \(2f \times 2f \times 2h\). Then all the points which lie outside the cylinder are rejected by imposing the condition:

\[
a^2 + b^2 \leq f^2
\]  

(3.15)

Similarly, to generate random point on the circular detector surface of radius \(g\), first random point is generated in a square of length \(g\) and then all the random points which lie outside the circle of radius \(g\) are rejected. These co-ordinates are then used to get the distance traveled by the gamma ray to reach the detector as given by equation 3.9. These co-ordinates are also then used to find the intersection point of the gamma ray with the sample surface i.e. \((x, y, z)\) as given in Section 3.3 of this thesis. Once \((x, y, z)\) is known then the distance traveled by the gamma ray in the sample \((t)\) is calculated using equation 3.10. These are then used to get \(k_{\text{att}}\). The output is stored in the file 'cylinder.out'. The calculations were performed by generating approximately \(10^5\) particles, using a desktop PC. This program was run separately for different sample-to-detector distances.

### 3.4.2 Box Geometry

For a box shaped sample \((2f \times 2f \times 2f)\), random points were generated within the dimension of the box. No additional conditions are required. The distance traveled by the gamma ray within the sample before reaching the detector \((t)\) can be obtained by solving Eq. (3.11) and the equation defining the plane surface of the box from which the gamma ray is emerging:

\[y = f\]  

(3.16)

\(k_{\text{att}}\) can then be obtained from the equation 3.14. The FORTRAN program for box shaped sample ‘BOX.FOR’ is given as program 2 in Appendix II. The symbols and
functions used in this program are similar to those used in program 1. The input and the output files are ‘transm.txt’ and ‘box.out’ respectively. Here also the attenuation correction factors have been calculated as a function of transmittance and sample-to-detector distances. The transmissions were varied by keeping the linear attenuation coefficient ($\mu$) constant and varying the half-thickness of the box ($yy$ or $f$).

3.4.3 Spherical Geometry

The random points in a spherical sample of radius $f$, are generated by first generating random points ($a$, $b$, $c$) within a cube of edge $2f$ and then rejecting all the points which lie outside the sphere. This is accomplished by imposing the following two conditions:

\[ \frac{a^2 + b^2}{f^2} \leq 1 \]  
\[ \frac{a^2 + c^2}{f^2} \leq 1 \]  

The distance traveled by the gamma ray to reach the detector is then obtained by using equation 3.9. The distance traveled by the gamma ray within the sample before reaching the detector ($t$) can be obtained by solving the equation 3.11 and the following equation defining the spherical surface:

\[ x^2 + y^2 + z^2 = f^2 \]  

$k_{att}$ can then be obtained from equation 3.14. The FORTRAN program for spherical sample ‘SPHERE.FOR’ is given as program 3 in Appendix II. The description of this program is also similar to that of the previous two programs. The input and the output files are ‘transm.txt’ and ‘sphere.out’ respectively.
3.4.4 Disc Geometry

For the disc geometry, the random points are generated in the sample in a way similar to cylindrical geometry i.e. first random points are generated in a box of same dimensions and then all the points lying outside the disc are rejected by imposing the following condition:

\[ a^2 + c^2 \leq f^2 \]

(3.20)

where \( f \) is the radius of the disc. Then, the distance traveled by the gamma ray within the sample before reaching the detector can be obtained by solving the equation 3.11 and the following equation:

\[ y = h \]

(3.21)

where \( h \) is the half-thickness of the disc. \( k_{\text{att}} \) can then be obtained from equation 3.14. The FORTRAN program for disc geometry ‘DISC.FOR’ is given as program 4 in the Appendix II. In this program, proper correction for the gamma rays reaching the detector from the walls of the disc has been incorporated. Here, the diameter of the cylindrical detector was 6 cm which was chosen for the calculations to match with the experimental detector dimensions. Separate runs were taken for each sample-to-detector distance.

3.5 Validation of the Hybrid Monte Carlo Approach

3.5.1 Theoretical Validation

The hybrid Monte Carlo approach was validated by comparing its results with MCNP calculations and geometrical formulae available in literature. The comparison was done at different sample-to-detector distances and for different extents of attenuation. Three most
commonly used sample geometries namely sphere, box and cylindrical were considered. Since the calculation of attenuation correction factor by both the HMC method and far-field and near-field formulae require linear attenuation coefficient as an input, therefore firstly linear attenuation coefficients were measured experimentally.

**Measurement of linear attenuation coefficient (µ)**

To measure linear attenuation coefficient experimentally, uranium was chosen as the attenuating matrix. This was done owing to the high Z of uranium. The linear attenuation coefficients were obtained for four uranyl nitrate samples (100-390 mg/ml) at the energies of interest. These samples were prepared by dissolving the required amount of U₃O₈ in concentrated nitric acid, followed by dilution with 2M HNO₃. The µ (cm⁻¹) were then obtained experimentally by transmission measurement of a collimated beam. For this, the transmission source was counted with and without the sample. The schematic of experimental set-up has already been shown in Figure 3.2. The ¹⁶⁹Yb was used as the transmission source as it has gamma rays close to the ²³⁵U gamma rays used in our measurement. Table 3.1 gives the ¹⁶⁹Yb and ²³⁵U gamma ray energies monitored. Here, the unattenuated count rate was obtained by replacing the sample with 2M HNO₃ solution.

The transmission curve as a function of gamma ray energy is shown in Figure 3.5. The highlighted points in the figure shows the gamma ray energies of ²³⁵U monitored. The transmittance for ²³⁵U energies was obtained by linear interpolation between the transmittance for ¹⁶⁹Yb energies. The linear attenuation coefficient was then obtained from equation 3.5.
Figure 3.5 Transmission curve as a function of gamma ray energy using $^{169}$Yb as transmission source.

Table 3.1 Gamma ray energies of $^{169}$Yb and $^{235}$U.

<table>
<thead>
<tr>
<th>Gamma ray energy (keV)</th>
<th>$^{169}$Yb</th>
<th>$^{235}$U</th>
</tr>
</thead>
<tbody>
<tr>
<td>130.5</td>
<td>143.8</td>
<td></td>
</tr>
<tr>
<td>177.2</td>
<td>163.4</td>
<td></td>
</tr>
<tr>
<td>198.0</td>
<td>185.7</td>
<td></td>
</tr>
<tr>
<td>307.7</td>
<td>205.3</td>
<td></td>
</tr>
</tbody>
</table>
Comparison of HMC results with Far-field results

As a first step, all the calculations were done by keeping the sample-to-detector distance sufficiently large so that the results can be compared with the far-field correction factors available in the literature. To calculate attenuation correction factors for a wide range of transmittance, the linear attenuation coefficient was fixed at 0.5 cm\(^{-1}\) which corresponds to the value at 186 keV for one of the uranyl nitrate sample. The transmittance was varied by changing the dimension of the sample along the detector axis. For cylinder and sphere, the radius of the sample was changed and for a box, its length was changed. In the case of cylinder, the height was of the order of the diameter of the detector so that the path of the gamma ray reaching the detector from the top or bottom surface could be neglected. But practically, if the height of the sample to be analyzed is such that there is a finite probability of gamma rays reaching the detector from the top or bottom of the sample, then the corrections can be easily incorporated in the required program. Table 3.2 gives the results of the present \(k_{\text{att}}\) calculations for the three geometries using a sample-to-detector distance of 100 cm. It also gives the corresponding \(k_{\text{att}}\) values calculated from the following far-field analytical expressions [Parker (1991)]:

\[
k_{\text{att(cylinder)}} = \frac{1}{2} \frac{\mu D}{[I_1(\mu D) - I_2(\mu D)]}
\]  
(3.22)

\[
k_{\text{att(box)}} = \frac{\mu x}{[1 - \exp(-\mu x)]}
\]  
(3.23)
where $D$ is the diameter of the cylinder and sphere, $x$ is the half-thickness of the box, $I_1$ and $L_1$ are modified Bessel and modified Struve functions of order 1. The data in Table 3.2 show a good agreement between the present calculations and the literature for all geometries, but deviations upto 5% are observed at very low transmittance. This validates the method of present calculation of $k_{att}$ at far-field geometry.

**Table 3.2** Results of $k_{att}$ calculations by the present method and comparison with values available in the literature for samples of different geometries.

<table>
<thead>
<tr>
<th>T</th>
<th>Cylinder</th>
<th>Box</th>
<th>Sphere</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$k_{att}$</td>
<td>$k_{att}^*$</td>
<td>$k_{att}$</td>
</tr>
<tr>
<td>0.01</td>
<td>3.74</td>
<td>3.79</td>
<td>4.43</td>
</tr>
<tr>
<td>0.09</td>
<td>2.29</td>
<td>2.30</td>
<td>2.60</td>
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<td>0.17</td>
<td>1.90</td>
<td>1.91</td>
<td>2.11</td>
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<td>0.25</td>
<td>1.68</td>
<td>1.69</td>
<td>1.84</td>
</tr>
<tr>
<td>0.33</td>
<td>1.53</td>
<td>1.54</td>
<td>1.65</td>
</tr>
<tr>
<td>0.41</td>
<td>1.42</td>
<td>1.42</td>
<td>1.51</td>
</tr>
<tr>
<td>0.57</td>
<td>1.25</td>
<td>1.26</td>
<td>1.31</td>
</tr>
<tr>
<td>0.73</td>
<td>1.14</td>
<td>1.14</td>
<td>1.17</td>
</tr>
<tr>
<td>0.89</td>
<td>1.05</td>
<td>1.05</td>
<td>1.06</td>
</tr>
<tr>
<td>0.97</td>
<td>1.01</td>
<td>1.01</td>
<td>1.02</td>
</tr>
</tbody>
</table>

*From Parker (1991)
three geometries were performed at varying sample-to-detector distance. These results were compared with the results from literature formula. Figure 3.6(a) shows the ratio of $k_{att}$ obtained by the present calculations to the $k_{att}$ calculated from the far-field expressions at different sample-to-detector distances for cylindrical geometry. The results agree within 2\% at almost all transmittance at higher distance (> 50 cm). The deviation starts at lower distance, particularly at lower transmittance. The lower value of $k_{att}$ than predicted by far-field expression is expected at closer distance as the geometric efficiency becomes dependent on the position of emission of gamma ray within the sample.

**Comparison of HMC results with MCNP results**

In order to further confirm the result of the present calculations, Monte Carlo simulation for the given sample-detector geometry were performed using MCNP code. For this purpose, company supplied dimension of the detector was used. In this case also uranium solution with experimentally determined $\mu = 0.5 \text{ cm}^{-1}$ at 186 keV was chosen as the matrix and its density was given as an input in the calculations. The transmittance of the solution was varied by changing the radius of the cylinder and sphere and length of the box. The unattenuated count rate was obtained by replacing the sample solution by air. The $k_{att}$ was calculated from the ratio of unattenuated to attenuated count rates. The results of the present calculations relative to MCNP calculations for cylindrical geometry are shown in figure 3.6(b). It is observed that the ratio no longer shows the kind of systematic distance dependence as observed in figure 3.6(a). This shows that the result of our calculations is applicable at all sample-to-detector distance. However, the ratio in figure 3.6(b) shows about 10\% deviation at lowest transmittance independent of sample-
to-detector distance. This may be due to inherent uncertainty involved in the MCNP calculations.

![Figure 3.6](image)

**Figure 3.6** Ratio of attenuation correction factors at different distances as a function of transmittance for cylindrical geometry. (a) $k_{att}^h/k_{att}^*$

(b) $k_{att}^s/k_{att}^s$

$^h$ Present calculation

$^s$ MCNP calculation

$^*$ From Reilly et al. (1991)

**Comparison of HMC results with Near-field results**

To see whether the present calculation converges with the prediction of near-field formula, the HMC results along with the far-field and MCNP results, were compared
with the results of near-field expressions (two dimensional model) as given below [Parker (1991)] for cylindrical geometry:

\[
k_{\text{att}} = \frac{\sum_{m=1}^{M} \sum_{n=1}^{N} \Delta A(m,n) / L^2(m,n)}{\sum_{m=1}^{M} \sum_{n=1}^{N} \{\exp[-\mu_t(m,n)] \Delta A(m,n) / L^2(m,n)\}}
\]  \hspace{1cm} (3.25)

where \(\Delta A(m,n)\) is the area element of a circular cross-section of the sample, \(L(m,n)\) is the distance of the area element from the point detector, \(t(m,n)\) is the distance traveled by the gamma ray in the sample as shown in Figure 3.7.

**Figure 3.7** Two dimensional model for computing \(k_{\text{att}}\) for a cylindrical sample.

Figure 3.8(a) shows the two dimensional model \(k_{\text{att}}\) relative to \(k_{\text{att}}\) values obtained from far-field expressions for cylindrical geometry. Here also, it was observed that the near-field formula match reasonably well at longer distances at all transmittance with the far-field values and as expected, the deviation starts at shorter sample-to-detector distance at
lower transmittance. The increasing importance of geometric efficiency at less sample-to-detector distance is apparent from the increasing deviation of far-field formula from near-field values as the distance decreases.

![Graphs showing the ratio of attenuation correction factors at different distances as a function of transmittance for cylindrical geometry.](image)

**Figure 3.8** Ratio of attenuation correction factors at different distances as a function of transmittance for cylindrical geometry. (a) $k_{att}^\# / k_{att}^*$

(b) $k_{att}^\# / k_{att}^\$

(c) $k_{att}^\# / k_{att}^\&$

^# Two-dimensional model [Parker (1991)] calculation

^$ Present calculation

* From Parker (1991)

& From MCNP calculations
Chapter 3

The comparison of the present result with the two dimensional model has been shown in Figure 3.8(b). It is observed that the values match within 2% at all transmittance even at 10 cm sample-to-detector distance. The deviation observed at shorter distance may indicate the failure of the near-field formula at very close sample-to-detector distance and needs further investigation. For quantitative comparison, the $k_{att}$ values at 10, 5 and 3.5 cm sample-to-detector distance, calculated based on two-dimensional model, present calculations and MCNP calculations are given in Table 3.3.

Table 3.3 Results of $k_{att}$ calculations by the present method and comparison with values available in the literature and MCNP values for cylindrical sample.

<table>
<thead>
<tr>
<th>T</th>
<th>$d = 10$ cm $k_{att}$</th>
<th>$d = 5$ cm $k_{att}$</th>
<th>$d = 3.5$ cm* $k_{att}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$k_{att}$</td>
<td>$k_{att}$</td>
<td>$k_{att}$</td>
</tr>
<tr>
<td>0.01</td>
<td>3.09</td>
<td>3.17</td>
<td>3.05</td>
</tr>
<tr>
<td>0.09</td>
<td>2.16</td>
<td>2.19</td>
<td>2.10</td>
</tr>
<tr>
<td>0.17</td>
<td>1.84</td>
<td>1.86</td>
<td>1.80</td>
</tr>
<tr>
<td>0.25</td>
<td>1.65</td>
<td>1.66</td>
<td>1.61</td>
</tr>
<tr>
<td>0.33</td>
<td>1.51</td>
<td>1.52</td>
<td>1.49</td>
</tr>
<tr>
<td>0.41</td>
<td>1.41</td>
<td>1.42</td>
<td>1.39</td>
</tr>
<tr>
<td>0.57</td>
<td>1.25</td>
<td>1.25</td>
<td>1.24</td>
</tr>
<tr>
<td>0.73</td>
<td>1.14</td>
<td>1.14</td>
<td>1.13</td>
</tr>
<tr>
<td>0.89</td>
<td>1.05</td>
<td>1.05</td>
<td>1.05</td>
</tr>
<tr>
<td>0.97</td>
<td>1.01</td>
<td>1.01</td>
<td>1.01</td>
</tr>
</tbody>
</table>

5 From 2D model
6 From present calculations
* From MCNP calculations
* These calculations were done with a higher $\mu$ so as to reduce the radius of the sample so that sample does not overlap with the detector.
As seen from the table, there is excellent agreement of the results of all the three calculations but for lowest transmittance values where agreement is seen between the present calculation and MCNP. Thus, it is seen that the present method is independent of any approximations regarding sample-to-detector distance for calculating $k_{att}$ for cylindrical geometry.

Similar calculations were performed for box and spherical geometry. Figure 3.9(a) shows the comparison of the present calculation with far-field expressions for box geometry. Again, the deviation from the literature expression is observed at lower transmittance and at shorter distance. The corresponding results with respect to MCNP simulation are shown in Figure 3.9(b). The distance dependence of the deviation, as seen from figure 3.8(a) disappears in Figure 3.9(b). Also, the predictions of MCNP agree within 5-8% with our results at all distance and at all transmittance. Also, there is no near-field formula available for box shaped geometry.

Similar comparisons are shown in Figure 3.10(a) and (b) for spherical geometry. In this case, the results of HMC calculations and far-field formula agree within 1% for all transmittance at all distance except at highest transmittance and 5 cm distance. The MCNP results also show the similar trend. It appears that the far-field formula for spherical geometry is applicable at all sample-to-detector distance in the transmittance range considered in the present work. It is thus seen that the $k_{att}$ values for common sample geometries, at all sample-to-detector distance, and in the transmittance range considered, can be calculated using the common approach developed in the present work. The MCNP calculations also provide a good estimate of the $k_{att}$. However, the calculations are involved, time consuming and require full description of the detector
geometry and exact sample composition which is not always available. The present
calculations are simple, rapid and require only $\mu$, detector diameter and sample
dimensions as input. The calculations can be extended to disc-shaped and other common
sample geometries.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figures/figure3.9.png}
\caption{Ratio of attenuation correction factors at different distances as a function of transmittance for box-shaped sample. (a) $k_{att}^\# / k_{att}^*$
(b) $k_{att}^\# / k_{att}^5$
\textsuperscript{#} Present calculation
\textsuperscript{*} From Parker (1991)
\textsuperscript{5} MCNP calculation}
\end{figure}
3.5.2 Experimental Validation of the Hybrid Monte Carlo Approach

In order to validate the theoretical computations, attenuation correction factors were also determined experimentally for cylindrical, disc and box geometry.

I. Cylindrical and disc geometry

For cylindrical and disc geometry, uranium solution was chosen as the attenuating matrix. For this purpose, the four 20 ml standard uranyl nitrate solutions prepared for the
theoretical validation study were used. The cylindrical and disc geometry as explained above in Figure 3.3 was realized by counting the same set of uranyl nitrate samples in a horizontal and a vertical HPGe detector. Both the detectors had a relative efficiency of 20% and a resolution of 2.2 keV at 1.33 MeV. The 143 and 186 keV gamma rays emitted by $^{235}\text{U}$ were monitored. The experimental disintegration rates ($dps_{\text{expt}}$) of a particular uranyl nitrate sample at the energy of interest (143 and 186 keV) was obtained by correcting the measured count rate of the sample with the efficiency at that energy and the intensity of the corresponding gamma ray, as given by:

$$dps_{\text{expt}} = \frac{CR_y}{\varepsilon_y a_y}$$  \hspace{1cm} (3.26)

where, $CR_y$, $\varepsilon_y$, $a_y$ is the count rate from sample, efficiency and abundance of the gamma ray concerned. For efficiency calibration, $^{152}\text{Eu}$ and $^{133}\text{Ba}$ were used in the same geometry. The experimental disintegration rates will not be constant for a particular sample and will show the increasing effect of attenuation as the gamma ray energy decreases. The experimental $k_{\text{att}}$’s at a particular energy was then determined by taking the ratio of the actual disintegration rate of the sample and the experimental disintegration rate at that energy:

$$k_{\text{att(expt)}} = \frac{dps_{\text{calc}}}{dps_{\text{expt}}}$$  \hspace{1cm} (3.27)

These measurements were also carried out at different sample-to-detector distances so as to check the applicability of the current approach over a wide range of sample-to-detector distances.
Chapter 3

Comparison of HMC results with Experimental, Near-field and MCNP results

To calculate the attenuation correction factors from the HMC method, the sample dimensions and sample-to-detector distance were given as an input in the calculations. In all the calculations, the sample diameter and height were taken to be 2.54 cm and 4 cm respectively. The detector diameter was 5 cm for horizontal detector (cylindrical geometry) and 6 cm for vertical detector (disc geometry). Along with this, the linear attenuation coefficients obtained experimentally at the energies monitored for different samples were given as an input in the program. These calculations were also performed by generating approximately $10^5$ particles, using a desktop PC.

To further confirm the result of the present calculations, Monte Carlo simulation for both the geometries was performed using MCNP code. In these calculations, apart from the precise information about the sample-detector geometry, the concentration of uranium in the sample was given as an input. The unattenuated count rate was obtained by replacing the sample solution by air. The $k_{att}$ from MCNP was calculated from the ratio of unattenuated to attenuated count rates. Figure 3.11 shows the theoretical $k_{att}$ values calculated from the given method compared with the experimental and MCNP results for cylindrical geometry at $d = 2.5$, 4.5 and 20 cm as a function of transmittance. It is seen from the figure that at $d = 4.5$ and 20 cm, the experimental and the MCNP $k_{att}$ values matches well with the present method $k_{att}$ and with the near-field formula. But at $d = 2.5$ cm, the near-field values shows a deviation from the experimental and the MCNP $k_{att}$ values but the present approach values matches well. This shows that the near-field formula is not valid at very near sample-to-detector distances while the present calculation is valid at all sample-to-detector distances.
Figure 3.11 The attenuation correction factors computed using numerical approach for cylindrical geometry as a function of transmittance for different sample-to-detector distances \(d\)

(a) \(d = 2.5\) cm (b) \(d = 4.5\) cm (c) \(d = 20\) cm.

Figure 3.12 shows the comparison of theoretical, experimental and MCNP obtained \(k_{att}\) values for disc geometry at three sample-to-detector distances, \(d = 2, 4, 5\) cm. Here also, the theoretical values match reasonably well with the MCNP and experimental \(k_{att}\) values at all the distances. This validates the numerical approach explained above over a wide range of sample-to-detector distances. Figure 3.12 also shows the \(k_{att}\) values calculated from a near-field one dimensional model given in Parker (1991):
\[ k_{\text{att}} = \left[ \frac{D}{d(d+D)} \right] \sum_{i=1}^{N} \frac{\exp[-\mu(I-0.5)\Delta x]}{[d + (I-0.5)\Delta x]^2} \] (3.28)

Here \( \Delta x = D/N \) where \( D \) is the sample depth, \( N \) is the number of intervals for the numeric integration and \( d \) is the distance from the sample surface to the detector surface. The model approximates the sample to be a line sample of depth ‘\( D \)’ and detector to be a point detector.

Figure 3.12 The attenuation correction factors computed using numerical approach for disc geometry as a function of transmittance for different sample-to-detector distances (\( d \)) (a) \( d = 2 \text{ cm} \) (b) \( d = 4 \text{ cm} \) (c) \( d = 5 \text{ cm} \).
As seen from the figure, the near-field formula values deviates significantly at very close sample-to-detector distance i.e. when the sample is almost in the touching configuration with the detector. This implies that the near-field formula although said to be valid at close sample-detector geometry, starts working only after certain sample-to-detector distance, whereas the numerical approach is found to be applicable at all sample-to-detector distance.

II. Box geometry

For box shaped samples, lead acetate solutions were used as the attenuating matrix, keeping in mind the high $Z$ of lead and high solubility of lead acetate. These solutions in the concentration range of 10 - 400 mg/ml were prepared by dissolving required amount of lead acetate in water. The linear attenuation coefficients of these samples were measured by using $^{57}$Co as the transmittance source by the method described above. To get experimental attenuation correction factors, known amount of $^{57}$Co activity was added to the samples. Known amount of activity was also added to a water sample which was chosen as a blank. These samples along with the blank were then counted in a 20% HPGe with a resolution of 2.2 keV at 1332 keV.

Figure 3.13 shows the experimental count rate per gram for $^{57}$Co activity as a function of lead acetate concentration for box samples. The straight line in the figure shows the expected count rate per gram as a function of lead acetate concentration. Here also, due to gamma ray attenuation, there is increasing deviation from linearity with increasing lead acetate concentration increases.
**Chapter 3**

**Figure 3.13** Count rate per gram of $^{57}$Co activity at 122 keV as a function of lead acetate concentration. The straight line shows the expected count rate per gram as a function of lead acetate concentration.

**Comparison of HMC results with Experimental, Near-field and MCNP results**

For box samples, experimental $k_{att}$ were determined by taking the ratio of count rate per gram of blank to the count rate per gram of sample. These $k_{att}$ were compared with the far-field $k_{att}$ as given by equation 3.23 and with the $k_{att}$ calculated from present approach. Figure 3.14 shows the experimental, far-field and present method $k_{att}$ for box-shaped sample at sample-to-detector distances, $d = 2.6$, 5.0 and 20.1 cm. As expected, at close distance i.e. at $d = 2.6$ and 5.0 cm, the experimental $k_{att}$ deviates significantly from far-field values but matches quite well with the present approach $k_{att}$. When the sample-to-detector distance is 20.1 cm, all the three attenuation correction factors match well. This again shows that the present approach is valid at all sample-to-detector distances.
Figure 3.14 The attenuation correction factors computed using numerical approach for box geometry as a function of transmittance for different sample-to-detector distances, (a) $d = 2.6$ cm (b) $d = 5.0$ cm (c) $d = 20.1$ cm.

3.6 Conclusion

A hybrid Monte Carlo method has been developed for calculation of attenuation correction factors for samples of varied geometries. The method has been theoretically and experimentally validated. It was observed that the near-field formulas available in literature are applicable only after certain sample-to-detector distance. The results of the
present calculation show the possibility of using a simplified common approach to calculate the $k_{att}$ for the simple geometries considered, over wide range of transmittance. The advantage of the HMC method is that it is free of any assumptions regarding sample-detector geometry. Also, the present approach is common to all sample geometries and sizes. The calculations will be particularly useful for attenuation corrections at shorter sample-to-detector distance where analytical formulae are not available. Also, these calculations will be useful when collimator geometry is involved for which no analytical expressions are available. The present calculation can also be extended to other geometries.