

STUDY OF THE EFFECTS OF NEGLECTING OR CONSIDERING THE RAYLEIGH SCATTERING IN MONTE CARLO SIMULATION CODES APPLIED FOR THE MEASUREMENT OF LARGE VOLUME SAMPLES*

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Abstract. The Monte Carlo method is often used for the simulations of gamma spectrometric measurements that are typical for germanium detectors. In contrast, there are few simulations for the measurements of radioactive waste drums. When such simulations are applied they often rely on simplified models and approximations. For the measurement of a standard radioactive waste drum the detection efficiency is in the range $10^{-6} - 10^{-8}$. This means that a very large number of photons should be simulated in order to obtain a result with satisfactory uncertainty. The question arises if it is possible to limit the simulation only to a much reduced group of photons that contribute predominantly to the detector signal. For the case of radioactive sources of small sizes, up to several dm^3 , it has been shown that the Rayleigh scattering does not have a significant contribution to the detector signal, but the case of radioactive waste drums with typical sizes in the 200 dm^3 range has not been investigated yet. We investigate whether the Rayleigh scattering could be neglected in the calculation of detection efficiency for the measurement of waste drums. If the answer to this question is a positive one, then the simulation of emitted photons in every point inside the source can be reduced to the simulation of only those photons that are emitted inside the solid angle in which the detector is seen from the emission point. We investigate also the possibility to reduce the simulation time by avoiding the simulation of photons that are emitted towards the detector but suffer interactions before exiting the drum. In this hypothesis the transport of photons through the drum is possible to be described by using an attenuation factor that is calculated analytically based on the total attenuation coefficient of the photons.

Key words: waste drum, efficiency calibration, Rayleigh scattering

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1. INTRODUCTION

Recently the topic of waste management has become of prime importance as many human fields of activity rely on nuclear technologies that are inherently generating radioactive waste. The general accepted solution for such waste is long time storage in appropriate repositories. The storage procedure however can only be applied after a thorough characterization and sorting of the waste so that it can be assigned to the appropriate waste management stream. A powerful characterization technique for drums containing radioactive waste relies on gamma spectrometry since this non-destructive technique is easy to apply on a regular basis for many waste drums. A qualitative analysis of a waste drum is achieved fairly easy by employing an energy calibration to identify the contained radionuclides from the energy of emitted gamma rays. The quantitative analysis of the radionuclides content however is not so straight forward because one first needs to evaluate the amount and pattern of scattering and absorption the emitted photons suffer inside the waste drum. For experimental procedures that are applied, large volume reference sources are used to study the transport of the gamma radiation through the waste drum and the detector response. An experimental procedure can describe a limited number of cases of waste configurations inside the waste drum and previous studies have shown that the result of the measurement is very much dependent on the properties of the matrix encasing the waste and of the waste distribution inside the drum. To complement the experimental procedures, computational techniques based on Monte Carlo simulation codes are used as a flexible tool.

Powerful and affordable modern day computers and large data storage capacities allow almost anybody to use computer simulations for describing complex experiments. A computer simulation may be used to describe a real world experiment or a fictional one (*i.e.* simulations are used as a powerful and flexible design and testing tool). In both cases simulations are an effective way of studying a phenomenon without the need of actually producing it in laboratory conditions (providing of course that the nature of that particular phenomenon is known so that it can be described theoretically and implicitly in a simulation algorithm). In many cases the simulations are the only way one can evaluate the outcome of an experiment, since the costs of the real experimental setup might be prohibitive or the practical activities could be too hazardous to organize. In spite of their enhanced flexibility, the methods based on simulation techniques may present some drawbacks as they usually require long computing times in order to describe a given experimental setup.

2. THE RAYLEIGH SCATTERING IN MONTE CARLO SIMULATION CODES

The Monte Carlo method [1] is often used for the simulations of gamma spectrometric measurements that are typical for germanium detectors. In contrast, there are few simulations for the measurements of radioactive waste drums and when such simulations are applied they often rely on simplified models and approximations. At a very basic level there is no fundamental difference between the Monte Carlo simulations of measurements of common radioactive sources with a volume in the dm^3 range [2-6], as commonly encountered in the environmental radioactivity monitoring (a field in which the Monte Carlo method is often applied) and the simulation of measurements of waste drums with a volume in the 200 dm^3 range [7]. Therefore this is not the reason for which the Monte Carlo method is avoided in the measurement of such drums. The main reason is linked to the computation time that is required in order to obtain a result with satisfactory uncertainty. Indeed the very small detection efficiency, in the range 10^{-6} – 10^{-8} , requires the simulation of a very large number of photons that are emitted during disintegrations occurring inside the radioactive source in order to accumulate a sufficient number of signals, leading to a reasonable uncertainty. As the time required for the transport of a photon emitted from the source and also for the secondary radiations generated throughout the waste drum is long, the corresponding total time required for simulation will be very long.

The very small detection efficiency means that from all the emitted photons from disintegrations very few contribute to the detector signal. The great majority of photons are either absorbed in the drum, or are scattered and do not reach the detector or reach it with reduced energy and thus do not contribute to the photo peak signals. In this context the question arises if it is possible to limit the simulation only to a much reduced group of photons that contribute predominantly to the detector signal, thus gaining a considerable amount of time by avoiding the description of radiation transport for the great majority of the emitted photons. In the particular case when we are interested only in the detection efficiency and not the entire spectrum, it is enough to describe only the photons that do not lose energy in the drum; these photons either do not interact at all in the drum, or suffer only coherent scattering (Rayleigh), this scattering mode being the only interaction mechanism for photons with energy in the range of interest in which the photons do not suffer energy loss. For the case of radioactive sources of small sizes, up to several dm^3 , it has been shown that the Rayleigh scattering does not have a significant contribution to the detector signal, but the case of radioactive waste drums with typical sizes in the 200 dm^3 range has not been investigated yet.

It is very important to know whether the Rayleigh scattering could be neglected in the calculation of detection efficiency for the measurement of waste drums. If the answer to this question is a positive one, then the simulation of emitted photons in every point inside the source can be reduced to the simulation of only those photons that are emitted inside the solid angle in which the detector is seen from the emission point; all the other photons that are emitted in other directions can not reach the detector with un-modified energy since they should suffer Compton scattering and thus their energy would be reduced. Taking into account that the solid angle in which a detector (with the size of the order of cm) is seen from a distance of the order of tens of cm up to 1 m is a very small fraction from the total solid angle, it means that by eliminating from the simulation the photons that are emitted in other directions other than towards the detector would provide a considerable gain in simulation time.

To answer this question an accurate simulation program was developed in order to reproduce the response of HPGe detectors in the measurement of radioactive waste drums. Unlike other applications of the Monte Carlo method for the measurement of radioactive waste drums, the described program includes all the details of the detector with the corresponding shielding and all details of a typical waste drum. The program is based on the simulation code GEANT 3.21 [8, 9] and was applied for two gamma spectrometric systems installed at IFIN-HH, Bucharest, Romania, the MADERA and the ISOCART systems. In both cases simulations were done for several distributions of radionuclides inside the drum and for several energies. The simulations were performed for the case in which the Rayleigh scattering is considered and also for the case in which it is neglected. The dependence of the detection efficiency and of the spectral shape on the inclusion or not of the Rayleigh scattering in the simulation strategy was investigated. Obviously the simulations require a very long computation time and therefore they can not be applied on a regular basis, but are instead only reference results.

Another possibility to reduce the simulation time is to avoid the simulation of photons that are emitted towards the detector but suffer interactions before exiting the drum. Taking into account the size of the drum relative to the attenuation length of the photons, the amount of photons interacting inside the drum is a considerable fraction and if it would be possible to avoid simulating them then the simulation time would be considerably improved. This is possible to achieve by introducing an attenuation coefficient that is calculated analytically based on the total attenuation coefficient of the photons and by avoiding the simulation of the corresponding interactions. The question arises if the value of the total attenuation coefficient should include also the contribution of the Rayleigh scattering. This matter has been addressed too in the framework of this work by studying the spectra of those photons that do not lose energy in the drum for the cases in which the Rayleigh scattering is considered or neglected.

3. THE MADERA AND THE ISOCART SYSTEMS

The main components of the MADERA system are (Fig. 1):

- the acquisition system with the HPGe detector;
- the translation-rotation system;
- computerized MCA (Canberra PC board) with acquisition software Genie

PC.

For the MADERA system [10–14] in the geometry of the experimental setup a lead collimated vertical HPGe detector of 30% relative efficiency and 1.8 keV energy resolution (at 1332 keV) was used. The rectangular collimator window has the length 10 cm and the width 4 cm. The lead shield has the inner diameter 9 cm and the wall thickness 4.4 cm. The detector was located at 32.5 cm radially from the surface of the calibration drum.

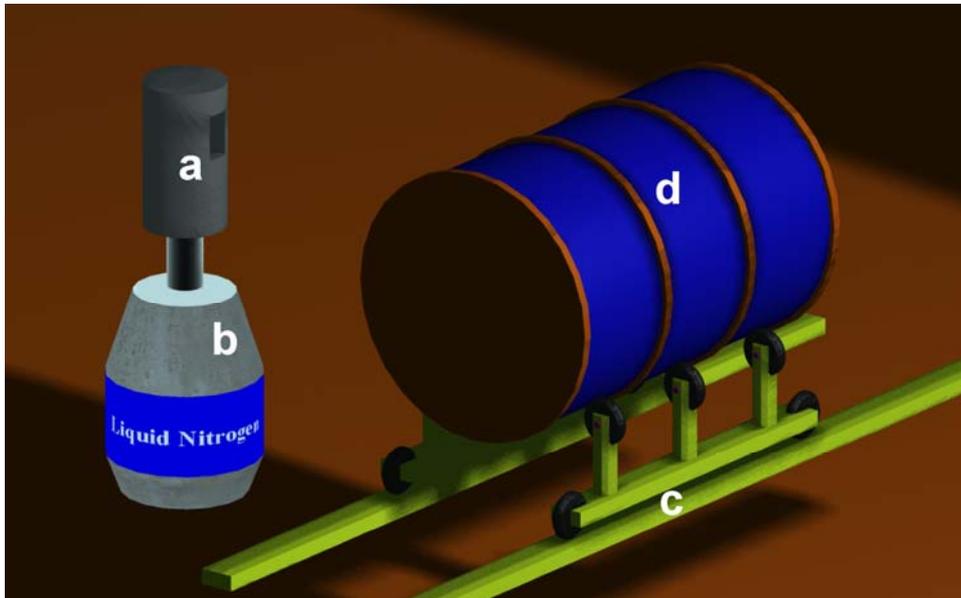


Fig. 1 – A 3D schematic representation of the MADERA system; several components are indicated: a) the detector shielding (Pb+Cu) the liquid nitrogen cooling unit; b) the drum support with the rolling mechanism; c) the calibration drum.

The main components of the ISOCART system are:

- the acquisition system with a lead-collimated horizontal HPGe detector;
- a specially-designed cart which carries all the components: the HPGe detector, the shield and the collimator, the digiDART™ high performance multichannel analyzer and a laptop computer; the cart provides the detector height adjustment and variable tilt adjustment and also the mobility of the acquisition system at different distances from the measurement drum (Fig. 2);

– 2 turntables for rotating the waste drums.

In the case of the ISOCART system [12,14] in the geometry of the experimental setup a lead-collimated horizontal HPGe detector was used, of 40% relative efficiency and 1.9 keV energy resolution at 1332 keV. The standard collimator is 1.6 cm thick, 20.3 cm long and 11 cm in diameter. It is composed of lead with a 2 mm inner copper liner. The detector was located at 100 cm radially from the surface of the calibration drum.

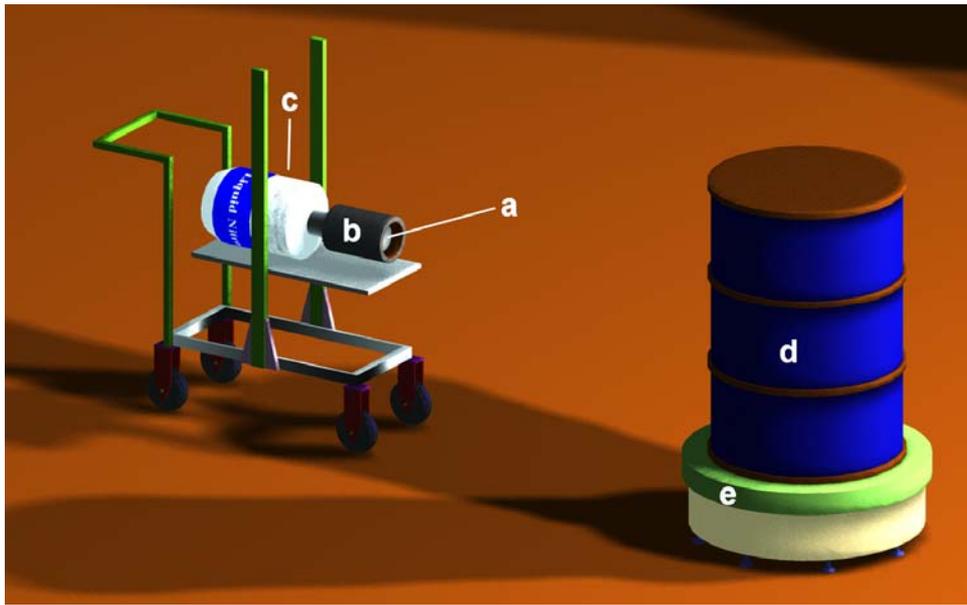


Fig. 2 – A three-dimensional schematic representation of the ISOCART system; several components are indicated: a) the aluminum detector casing; b) the detector shielding (Pb+Cu); c) the liquid nitrogen cooling unit; d) the calibration drum; e) the drum support with the rotating mechanism.

4. THE SIMULATIONS

As the simulation time for obtaining a low uncertainty is very long, in the simulation program many quantities were simultaneously evaluated. For each geometry, energy and case (with/without Rayleigh scattering) the spectrum of energy deposition in the germanium crystal was recorded. This corresponds to an ideal case, when the detector resolution is neglected. Simultaneously a realistic spectrum was recorded, by replacing each value of the energy deposited in the detector by a random value selected from a normal distribution with mean equal with the energy deposited and width corresponding to the actual energy resolution of the detector. The comparison of the ideal spectra for the two cases (with/without Rayleigh scattering) can reveal more sensibly the differences resulting from

neglecting the Rayleigh scattering, while the comparison of the real spectra for the two cases can provide a measure of the effect in the actual measurement conditions. Another spectrum was also recorded including only the signals produced in the detector by the photons that did not deposit any energy in the drum; in the case when Rayleigh scattering is neglected, these photons did not interact at all in the drum, while in the case when Rayleigh scattering is included, these photons did not interact at all in the drum or interacted through Rayleigh scattering. The comparison of the latter spectra in the two cases is useful for testing whether the analytic description of the attenuation in function of the total attenuation coefficient excluding Rayleigh scattering is acceptable.

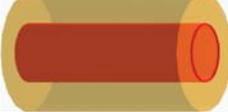
MADERA	Energy [keV]				
	122	611	661	1173	1332
 point source at one end of the drum	-	-	10×10^9 + 10×10^9	10×10^9 + 10×10^9	-
 uniformly distributed source	10×10^9 + 10×10^9	-	10×10^9 + 10×10^9	10×10^9 + 10×10^9	10×10^9 + 10×10^9
 ring shaped source (r=18 cm)	10×10^9 + 10×10^9	-	-	-	-
 tube source (r=12 cm)	10×10^9 + 10×10^9	-	10×10^9 + 10×10^9	-	-

Fig. 3 – This diagram presents the considered source geometries and energies that were simulated for the MADERA system. For each case the number of source photons traced is given.

Several tests have been performed for both the MADERA and ISOCART detector geometries. The configurations chosen for the radioactive source were: a point source placed at one end of the drum 1 cm deep under the surface, a uniformly distributed source inside the entire volume of the drum, a tube source and a ring source. While the tube and the ring shapes may represent approximations of sources with the same shape, they are also used to approximate linear sources and point sources that are rotated around the symmetry axis of the drum. Several energies were also considered for the gamma photons: 122 keV, 611 keV, 661 keV, 1173 keV and 1332 keV. Figures 3 and 4 present the simulated configurations. For every configuration 20×10^9 events have been simulated, of which half of the events were simulated considering the Rayleigh scattering while the other half were simulated without considering it. In all, the equivalent computing time for one CPU was ~ 6 months.

ISOCART		Energy [keV]				
		122	611	661	1173	1332
Source geometry						
	point source at one end of the drum	-	10×10^9 + 10×10^9	10×10^9 + 10×10^9	10×10^9 + 10×10^9	-
	uniformly distributed source	10×10^9 + 10×10^9	-	10×10^9 + 10×10^9	10×10^9 + 10×10^9	10×10^9 + 10×10^9
	tube source (r=12 cm)	10×10^9 + 10×10^9	-	10×10^9 + 10×10^9	-	10×10^9 + 10×10^9
	point source in the center of the drum	10×10^9 + 10×10^9	-	10×10^9 + 10×10^9	-	-

Fig. 4 – This diagram presents the considered source geometries and energies that were simulated for the ISOCART system. For each case the number of source photons traced is given.

5. RESULTS AND DISCUSSIONS

For the considered source geometries and for each energy the simulated photon spectra are compared for the case when the scattering is neglected and the case when it is considered. Such comparison is presented in Fig. 5 and Fig. 6 as example plots. The simple comparison of such spectra reveals no significant effect for the case of neglecting the scattering. A similar comparison of simulated spectra was possible to do only for the photons that did not deposit energy in the drum (Fig. 7 and Fig. 8). The case of these photons can be understood as follows: for the case of neglecting the Rayleigh scattering such photons did not interact at all in the drum, while for the other case when the scattering is simulated it means that either they did not interact at all, or they suffered only coherent scattering. Also in the case of this comparison there was no significant difference between the two cases. Because not all photons are selected to build these spectra they have considerably less events than in the case of all photon spectra.

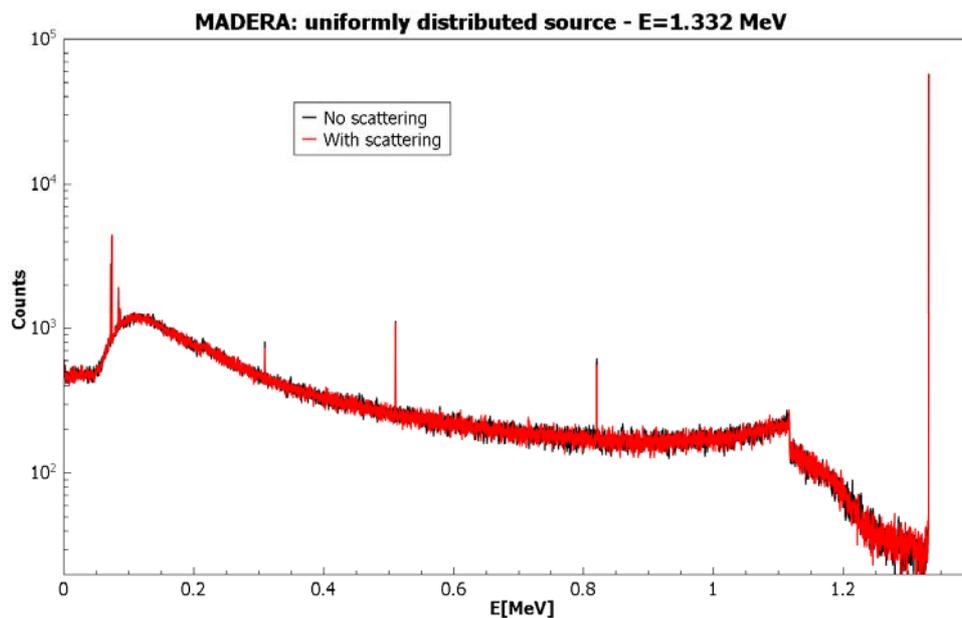


Fig. 5 – Spectra comparison for the two cases of neglecting or considering the coherent scattering (in black the case of no scattering and in red the case with scattering); at 0.511 MeV the annihilation peak is visible while at its right and left are the escape and double escape peaks.

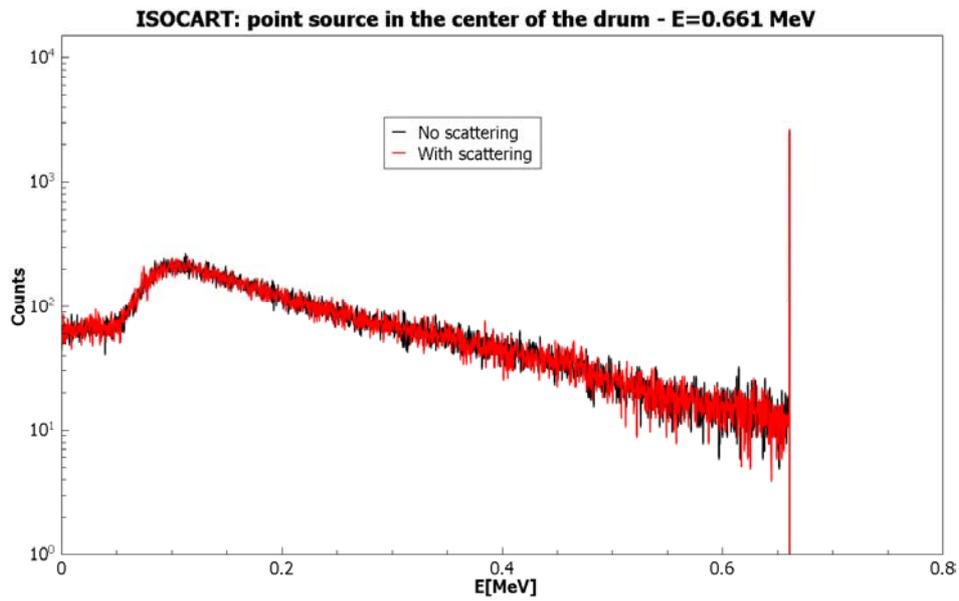


Fig. 6 – Spectra comparison for the two cases of neglecting or considering the coherent scattering (in black the case of no scattering and in red the case with scattering).

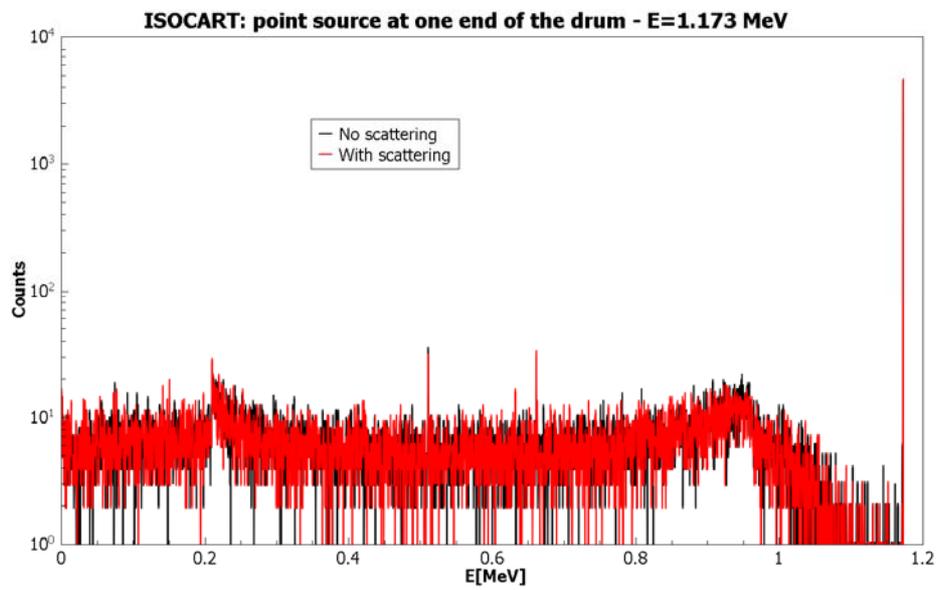


Fig. 7 – Spectra comparison for the two cases of neglecting or considering the coherent scattering (in black the case of no scattering and in red the case with scattering) only for those photons not losing energy inside the drum.

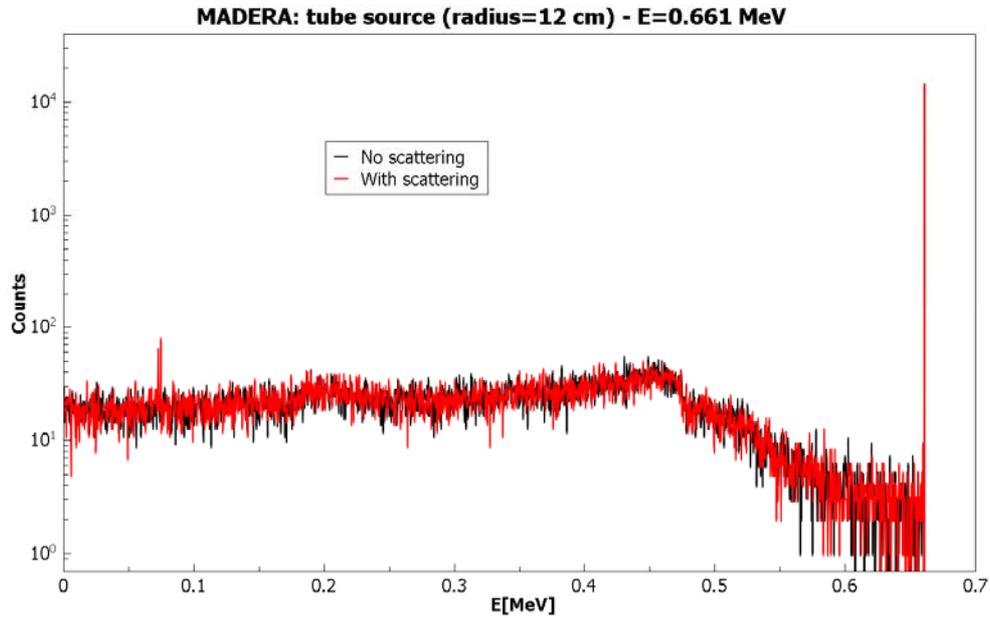


Fig. 8 – Spectra comparison for the two cases of neglecting or considering the coherent scattering (in black the case of no scattering and in red the case with scattering) only for those photons not losing energy inside the drum.

The peak efficiencies have also been calculated and compared for the two cases, as presented in Table 1. The differences are in general smaller than the associated statistical uncertainty of the results indicating that the effect of the Rayleigh scattering is very limited. Towards lower energies where the coherent scattering contributes more to the total cross section there is increased difference between the results in the two cases, but the statistical uncertainty also increases significantly. For lower energies the number of photons reaching the sensitive area of the detector decreases due to increased attenuation in the drum and therefore for the same number of initial photons we will record less events (resulting in increased statistical fluctuations).

This tests suggest that the effect of neglecting the Rayleigh scattering in Monte Carlo simulation codes is very limited and by ignoring it one can still give accurate descriptions of the gamma radiation transport through matter. This means that in fact the other competing effects to the total cross section can be effectively described by means of an analytical attenuation form. In a simulation code, such an analytical treatment would provide a very fast simulation with no expense of quality.

Table 1

Table showing a comparison between the calculated peak efficiencies for all considered geometries and energies

No.	Case Detector system/ Geometry of the source/ Photon energy	ϵ Peak efficiency average values (no Rayleigh scattering)	σ_{ϵ} Uncertainty	ϵ^R Peak efficiency average values (with Rayleigh scattering)	σ_{ϵ^R} Uncertainty	$(\epsilon^R - \epsilon)/\epsilon^R$ Relative difference
1	MADERA: Point source at one end of the drum E=661 keV	6.54503E-08	1.24791E-08	6.2933E-08	1.0659E-08	-0.040
2	MADERA: Point source at one end of the drum E=1173 keV	1.92285E-07	2.22595E-08	1.91897E-07	1.99043E-08	-0.002
3	MADERA: Uniformly distributed source E=661 keV	5.64151E-06	1.07638E-07	5.6356E-06	1.01394E-07	-0.001
4	MADERA: Uniformly distributed source E=1173 keV	5.44477E-06	9.56094E-08	5.46142E-06	1.04857E-07	0.003
5	MADERA: Uniformly distributed source E=1332 keV	5.529E-06	8.88818E-08	5.54169E-06	9.92871E-08	0.002
6	MADERA: Ring shaped source E=122 keV	8.03606E-07	3.73925E-08	8.02734E-07	4.14663E-08	-0.001
7	MADERA: Tube source E=122 keV	1.31481E-07	1.47284E-08	1.32062E-07	1.38129E-08	0.004
8	MADERA: Tube source E=661 keV	1.33282E-06	5.15745E-08	1.33204E-06	5.12610E-08	-0.001
9	ISOCART: Point source at one end of the drum E=611 keV	1.60431E-07	1.72892E-08	1.5985E-07	1.76898E-08	-0.004
10	ISOCART: Point source at one end of the drum E=661 keV	1.81053E-07	1.88004E-08	1.78052E-07	1.47582E-08	-0.017
11	ISOCART: Point source at one end of the drum E=1172 keV	4.30559E-07	2.56971E-08	4.34141E-07	2.7418E-08	0.008
12	ISOCART: Tube source E=122 keV	8.53952E-08	1.09278E-08	7.8521E-08	1.11605E-08	-0.088
13	ISOCART: Tube source E=661 keV	7.94989E-07	4.19405E-08	7.75238E-07	3.35825E-08	-0.025
14	ISOCART: Uniformly distributed source E=122 keV	7.01102E-06	1.10041E-07	7.00579E-06	1.02312E-07	-0.001
15	ISOCART: Uniformly distributed source E=661 keV	5.4651E-06	1.04158E-07	5.51787E-06	1.05733E-07	0.010
16	ISOCART: Uniformly distributed source E=1172 keV	4.74989E-06	9.02243E-08	4.72085E-06	9.79121E-08	-0.006
17	ISOCART: Uniformly distributed source E=1332 keV	4.59082E-06	9.50428E-08	4.62151E-06	9.99396E-08	0.007
18	ISOCART: Point source in the drum center E=122 keV	5.13146E-09	2.74028E-09	4.35690E-09	2.69672E-09	-0.177
19	ISOCART: Point source in the drum center E=661 keV	2.38661E-07	2.03176E-08	2.47859E-07	2.06009E-08	0.037

Taking into account the uncertainties associated to the simulation results the differences in the detection efficiency are not very important. It is possible that by producing some simulated samples for considerably more photons for the case of considering or neglecting the Rayleigh scattering differences could be recorded that are significant from the statistical point of view. If this should be the case then it

would be desirable that the simulations would consider the Rayleigh scattering. On the other hand, in order to reduce the computation time the number of simulated photons should be considerably smaller than the one simulated in the described tests and consequently the associated uncertainties would be significantly larger than in the case of neglecting the Rayleigh scattering. By neglecting the Rayleigh scattering in the evaluation of the detection efficiency a significant reduction of simulation time is achieved of the order of the ratio between the total solid angle and the solid angle in which the detector is seen from the emission point (by simulating only photons that are emitted in a narrow solid angle from the emission point towards the detector).

Additionally in this case it is also possible to avoid the random exclusion of photons on their track from the emission point towards the detector (interactions that would considerably and randomly reduce the number of photons reaching the detector) by considering a weight factor that is proportional with the attenuation which is calculated analytically based on the total attenuation coefficient (excluding the Rayleigh scattering) of photons through various materials. The results presented in this paper show that such approximations are appropriate. Their use leads to the creation of similar but much faster programs (*e.g.* GESPECOR) that can be applied on a regular basis for the treatment of a number of photons that is large enough for the associated statistical uncertainty to be much smaller than that obtainable in the case in which the Rayleigh scattering is included in the simulation. In this case the total uncertainty, including the statistical contribution as well as the one resulting from the neglect of the Rayleigh scattering, is much smaller than the total uncertainty (dominated by the statistical component) that is possible to obtain in a reasonable amount of time if the Rayleigh scattering is considered.

6. CONCLUSIONS

The tests described in this paper were purely Monte Carlo tests in which the importance of considering or neglecting the Rayleigh scattering in Monte Carlo simulation codes was evaluated. The GEANT simulation package was used to simulate gamma radiation transport through a drum and detector configuration. Two experimental setup cases have been considered for modeling in simulations: the MADERA and the ISOCART gamma spectrometric systems. Several geometrical configurations have been considered for the waste drum and source assemblies. The energy of the emitted photons was set to take a number of relevant values (covering a rather wide energy range) and also the shape of the source was changed to some specific geometries of interest (point source, ring or tube source, uniformly distributed source). The tests were performed twice for the same simulated setup, once neglecting the Rayleigh scattering and the second time by considering it. For every case the detection efficiency was calculated and also the energy deposit spectra in the detector were stored. The all photon spectra and the spectra generated

only by the photons that did not interact in the drum were then compared for the cases of neglecting the Rayleigh scattering and for the cases of considering it. The direct comparison between spectra shows no obvious effect coming from the Rayleigh scattering as the pairs of spectra do not show obvious differences. When comparing directly the calculated detector efficiencies for the two cases the differences are smaller than the associated statistical uncertainties. Overall, the tests show that the effects of considering the Rayleigh scattering are negligible and in fact the rest of the competing effects taking place in the drum for the photons that contribute to the peaks in the spectra can be described by means of an analytical attenuation form also neglecting the Rayleigh scattering. Neglecting Rayleigh scattering and describing photon interactions in the drum by means of an analytical attenuation factor increases simulation speed with no loss of simulation quality.

The results thus validate fast simulation procedures as those applied in simulation codes as GESPECOR.

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