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Feasibility study of detection of high-Z material in nuclear waste storage facilities with atmospheric muons

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Muon radiography is a well-established technique which is widely used in investigating the internal density structure of targets of different size and composition. Some examples of successful applications are the search for hidden chambers in archaeological sites and the monitoring of geological structures like volcanoes. The two main approaches to muon radiography are based on the effects of multiple Coulomb scattering and on absorption inside the target of atmospheric muons. The results of a Monte Carlo feasibility study of using muon radiography to investigate the presence of high-Z material (e.g. uranium) inside nuclear waste storage facilities using both the above mentioned techniques are presented. Albeit muon radiography has already been successfully applied to this kind of investigation in the past, this is the first time that it is benchmarked against the detection of cm-sized, high-Z samples inside building-sized storage facilities. For both multiple scattering and absorption approaches, preliminary results show that uranium samples of typical size greater than 5 cm can be detected inside a storage silo with a size of some meter filled with concrete, with a data taking period of several weeks. Smaller samples with size 2 cm are not detectable due to multiple scattering within the concrete matrix. The dependence of these results on the position of the samples and on the duration of data acquisition have been investigated and are reported as well, together with an estimate of the detection probability for fake signals.

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1. Introduction

Atmospheric muons are produced by the interaction of high-energy cosmic rays (mainly protons) with the upper layer of the Earth's atmosphere. The penetrating radiation that reaches the Earth's surface is mainly constituted of muons, which have a rate of about 1 particle per square centimeter per minute. The main interaction processes of muons with matter are energy loss through ionization of atomic electrons and multiple scattering from atomic nuclei. These processes can both be exploited in order to investigate the internal density structure of large target bodies. The pioneering works of George [1] and of Alvarez and collaborators [2] paved the ground for successive applications in different fields like geology and volcanology [3, 4, 5] and monitoring of legacy nuclear waste storage [6, 7]. Recently, two different measurement techniques based on multiple Coulomb scattering and on muon absorption, respectively, have been tested and compared on the damaged nuclear power plant in Fukushima (JP), showing a better sensitivity of the multiple scattering technique for both high-density core materials and low-density regions like air gaps [8, 9].

Recent interest has been put by the nuclear industry in searching small amounts of heavy nuclear material (of the order of some tens of cm^3) inside storage facilities with dimensions of some meters. Legacy containers for nuclear waste from the half of last century now pose significant challenges in terms of ensuring a safe disposal route as well as protection of the environment. In the past there perhaps wasn't the same safety culture and detailed record keeping procedures as there are now, which of course means that for these legacy waste containers there is a strong need to better characterise the contents of these containers. Non-invasive interrogation techniques have therefore to be developed in order to improve the knowledge about their actual content. To this extent, muon radiography shows interesting potential. However, the study case differs significantly from the successful applications of muon radiography mentioned in the previous paragraph. The relative size of the searched material and of the containing structure (some cm vs. some m) pose a severe question about the magnitude and the detectability of an eventual signal. Moreover, the size of the container lies in the intermediate region between small targets like canisters, where the multiple scattering provides the most sensitive results, and large ones like volcanoes, where absorption gives reliable and cost-effective results. It is not clear which measurement technique is best suited for this intermediate case.

This paper describes the first attempt to investigate the above described scenario by means of Monte Carlo simulations. The goal of this preliminary work is to assess the feasibility of detecting small, cm-sized samples of heavy nuclear material like uranium buried inside building-sized container silos filled with concrete, and the relative performance of two muon radiography techniques based respectively on multiple scattering and muon absorption. Results from this kind of study can help to nail down the size of the (eventual) expected signal and to identify the possible optimizations in both measurement techniques and data analysis algorithm.

In the following sections a brief review of the two techniques is given, followed by a description of the simulation setup and a discussion of the results.

2. Muon radiography

2.1 Multiple Coulomb scattering

Charged particles traversing a given material are scattered by electromagnetic interactions with atomic nuclei. The mean scattering angle increases with the atomic number of the target nuclei, thus making the probability of large scattering angles higher for heavy target materials. With a pair of tracking detectors it is possible to determine the particle's trajectory before entering and after exiting the target volume, and to reconstruct the scattering angle and the position of the scattering point within the target. The associated scattering density λ is an observable which depends on the atomic number Z of the target material and which grows larger for high values of Z [10].

To reconstruct the scattering density of the target, the volume is divided in voxels. The size of the voxels can influence the resolution of the scattering density map, so in principle this parameter should be optimized. However, when interrogating a large volume like the one studied here, computing power is the limiting factor. In this work, the voxels are cubic volumes with size 2.5 cm, unless otherwise specified. The scattering density maps are reconstructed iteratively from simulated muon data using an optimized implementation of the MLEM algorithm [11]. For each voxel traversed by each muon the scattering likelihood is computed assuming for the scattering density the value determined at previous step, and from the median of the likelihood over all muons the updated scattering density is then computed. For the first step, the value of λ for a homogeneous background material, i.e. concrete in this case, is used for every voxel. Upon convergence, the signal-to-background ratio (S/B) for each voxel is computed as the ratio of the obtained λ over the λ of the background. The S/B provides a normalized contrast parameter which measures the discrimination of the over-density in each voxel with respect to the background concrete.

2.2 Muon absorption

A different approach to muon radiography leverages the energy loss due to ionization of atomic electrons. A muon traversing a target body will lose its kinetic energy and eventually be stopped if the energy loss is sufficiently high. The amount of lost energy increase with the density of the traversed material, so more dense regions will stop more muons. By comparing the amount of detected muons with the expected rate in the hypotheses of a homogeneous body, a map of the density fluctuations with respect to the mean hypothesized density can be constructed. This investigation method is preferred over the multiple scattering approach when interrogating large structures like volcanoes, since it needs only one detector and thus allows for greatly enhanced geometric acceptances and reasonable data taking periods. The reduced cost of producing only one detector instead of two is another advantage of the absorption method.

When compared to very large targets, a typical meter-sized muon track detector is point-like, so it cannot produce a three-dimensional density map, being sensible only to the integrated column density of the target. However, the dimensions of a legacy storage silo are comparable to the size of the detector: each point of the target silo is seen from different angles by different parts of the detector, and this "stereoscopic view" can be leveraged in order to build a full three-dimensional tomographic scan of the target density. The reconstruction method used in this work is described in details in [12], and is based on the back-projection of detected muon tracks on a plane intersecting the target volume. A two-dimensional histogram (called the "back-projection map") counting the number of intersection points of detected muon tracks with the back-projection plane is computed and then compared with the expected histogram for a homogeneous target. Regions of the back-projection maps overlapping with a positive density fluctuation of the target will show a deficit in the count of intersection points. This deficit is the searched for signal, so these regions are called the "signal regions" of the back-projection map. When moving the back-projection planes, the size and the signal intensity of the signal regions will increase and decrease, respectively, due to a focusing effect very similar to the well-known one in classical optics. Thus the position of the signal region on the back-projection layer which minimizes the size of the region and maximizes the signal gives the full three-dimensional position of the density fluctuation inside the target. A normalized parameter describing the statistical significance of a density fluctuation is the signal-to-noise (S/N) parameter, computed for each bin (or "pixel") of a back-projection map as the difference of counts for the real target and for the hypothesized homogeneous one divided by its statistical error.

3. Monte Carlo simulations

A full Monte Carlo simulation of the storage silo scenario has been implemented using the Geant4 toolkit [13]. The simulated flux of atmospheric muons is derived from ground measurements taken with a magnetic spectrometer [14], in order to minimize the systematic error associated to this parameter. The storage silo is a 4 m high cylinder with radius 3.5 m filled with concrete. The detector is made of 2 vertical planes, each one made of scintillating elements arranged in two orthogonal X-Y views, separated by 50 cm and with a size of 2x2 m^2. It is placed 50 cm away from the silo and facing the silo itself, laying on the ground. For the multiple scattering simulations there is another identical detector placed upstream of the silo, at a height of 3 m. A sketch of the detectors arrangement is given in fig. 1. The layers of the detector for absorption are monolithic: the muon impact position is taken from the Monte Carlo truth, and a gaussian smearing of 0.3 cm is applied to mimick the response of a real detector. For multiple scattering the layers are segmented in 1000 scintillating fibers of 0.2 cm pitch.

Different cubic uranium samples of different sizes (20, 10, 5 and 2 cm) are placed inside the silo. For the multiple scattering simulations, one sample at a time is placed at the center of the silo, while for absorption many different samples are placed in different places in the same simulation run, to save computing time and study the dependence of the detection capability on the position of the sample. Additionally, multiple scattering simulations have been conducted with variable amounts of effective concrete thickness (modulated by varying the concrete density) in order to assess the effect of additional multiple scattering in the concrete on the final results.

4. Results

The data analysis for this preliminary study has taken advantage of the known positions of the simulated uranium samples. While this is clearly not corresponding to a real world case it is appropriate for the purpose of this study, which is to assess the presence of a signal and the detection capability of two different measurement techniques. The implementation of a signal-finding algorithm and other refinements of the analysis procedures are beyond the scope of this work.

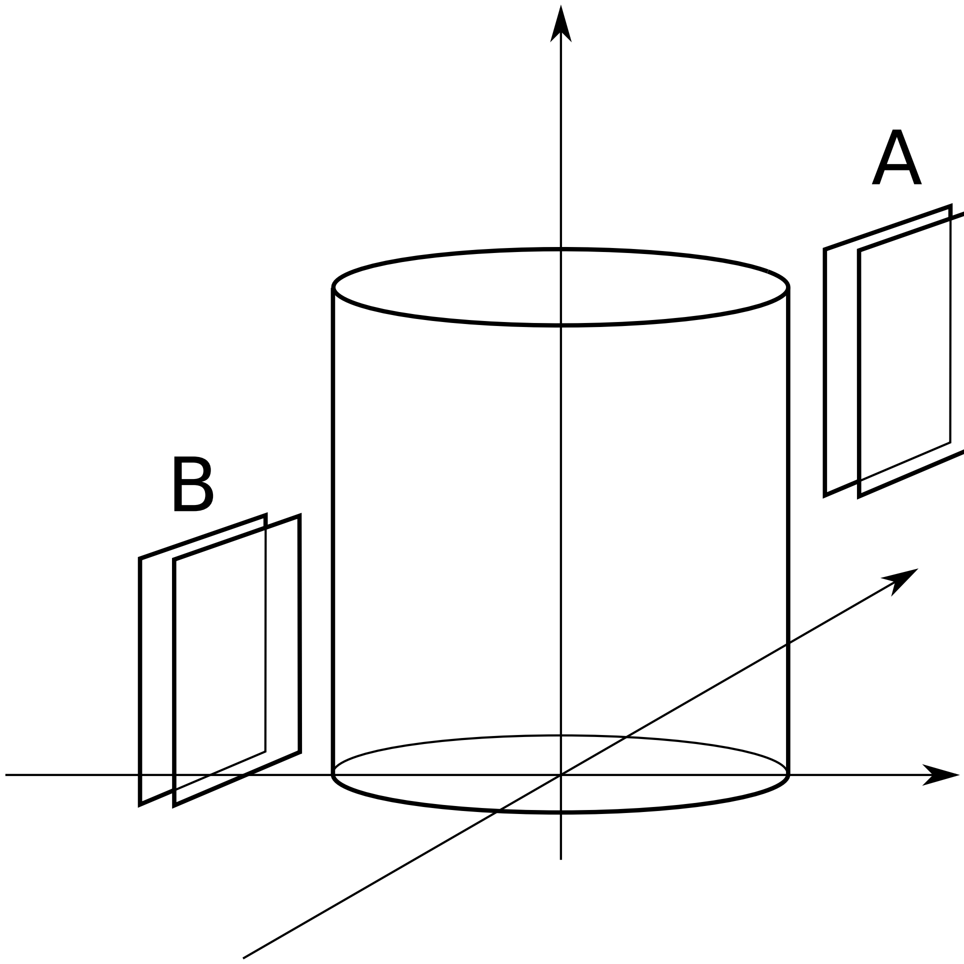

Figure 1: Arrangement of the detectors with respect to the silo. In the absorption simulations only detector B is present. Detected muons goes from right to left.

4.1 Multiple Coulomb scattering

The results of the multiple scattering analysis are shown in fig. 2. The S/B ratio shows a clear dependence on the size of the uranium sample and on the effective concrete thickness; however, both the 20 cm and the 10 cm samples are always detected, with S/B values exceeding the background ones by at least 30% with negligible statistical error. The situation is more mixed for smaller samples: the 5 cm one is detected only for an effective concrete thickness less than 2 m, for which an excess of about 20% is observed, while the 2 cm sample is never detected. Various hypotheses have been tested to explain these results, from the large separation between the detectors which impairs the detection of large scattering angles to the size of the voxels which in the case of the 2 cm sample contain also some concrete. The most significant explanation is that the contribution of multiple scattering in the surrounding concrete reduce the capability of identifying small-sized uranium samples.

The dependence of S/B on the data acquisition period is less pronounced than that on concrete thickness but still appreciable. For a 2 m concrete thickness the big sample is detectable with a very short data taking period, while the 10 cm one and the 5 cm require at least 6 and 20 weeks respectively to be identified, the latter with a S/B value of only 1.2. The big statistical errors and the decreasing trend with increasing time shown for large samples at small times is due to the contribution of large scattering angles from the tails of the scattering distribution [11].

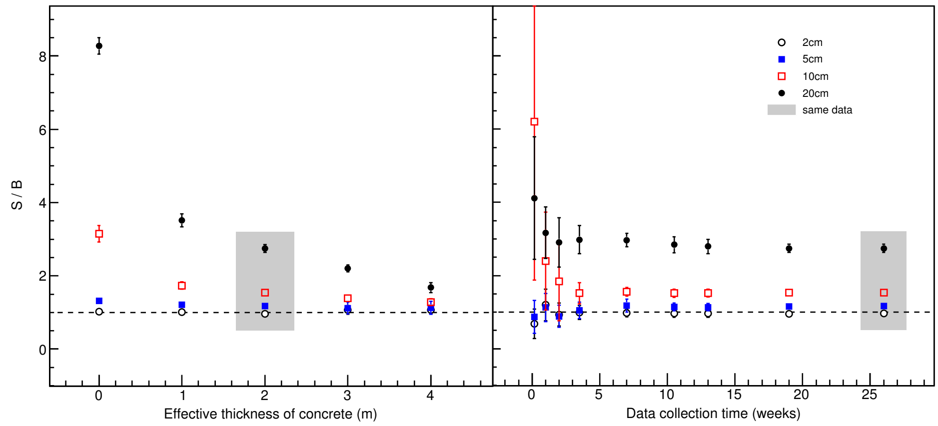

Figure 2: Results of the multiple scattering analysis. The left panel shows the dependence of the S/B level for the different uranium samples on the effective concrete thickness for a data acquisition time of 26 weeks. The right panel shows the dependence of the same variables on the time acquisition period (100% efficiency and duty cycle are hypothesized) for an effective concrete thickness of 2 m). The shaded boxes mark the same data sets in the two plots for the sake of an easier comparison.

4.2 Muon absorption

The absorption analysis is centered on the S/N variable, which gives a measurement of how large the signal is with respect to the expected background fluctuations. In order to fix the detection threshold, the probability of having a S/N greater than a fixed value in at least one of the pixels of the difference map has been computed, taking into account the fact that the signal of a single pixel follows a Skellam distribution (being the difference of two Poisson random variables) and the look-elsewhere effect. It turn out that there is only a 0.08% probability of having a S/N > 5 due to a pure statistical fluctuation over the whole difference map, so this S/N value has been taken as the detection threshold.

As for the multiple scattering results, the detection of uranium samples with absorption measurements shows a dependence on both the size of the sample and the thickness of the concrete surrounding it. The 10 cm sample has been detected when placed at the center of the silo, but not when placed at the bottom of the silo. This is due to the reduced statistics given by the less intense muon flux at large polar angles. In a similar way, the 5 cm sample at the center was not detected but it was when placed at the top of the silo. Again, this is due to the increased statistics since the line-of-sight connecting the detector and the sample is at a lower polar angle (and thus sees a higher flux), but also due to the reduced amount of concrete and thus of spurious multiple scattering along that line-of-sight. The 2 cm sample was never detected, regardless of its position, due to the multiple scattering inside the concrete. Fig. 3 shows the S/N ratios for the two detected samples: a data acquisition time of about 5 weeks is needed for the 10 cm sample, while about 10 weeks are necessary for the 5 cm one.

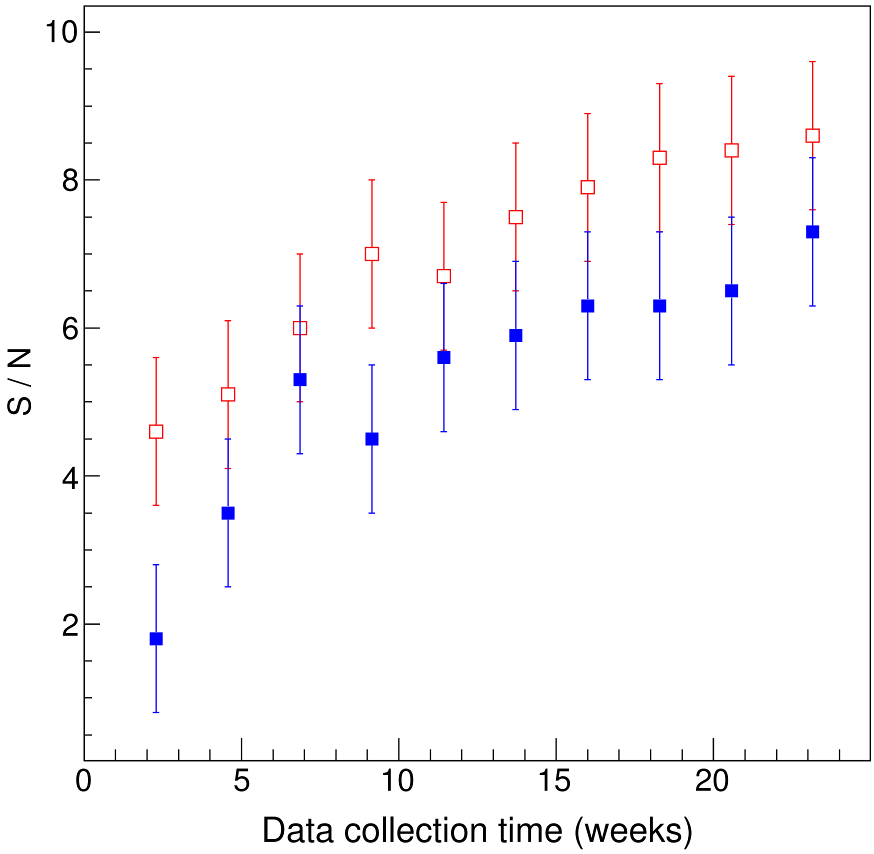

Figure 3: Variation of the S/N ratio for the 5 cm (full boxes) and 10 cm (open boxes) uranium samples with data acquisition time. Error bars are statistical errors. Details about the positions of these samples are given in the text.

5. Discussion

While based on different approaches and different estimators for the signal, the two muon radiography techniques gave very consistent results. The detection of small-sized uranium samples inside large storage silos made of concrete depends mainly on the size of the sample and the thickness of the concrete. It is possible to detect properly positioned uranium samples down to 5 cm size with a reasonable data taking period. Smaller samples seems to be not detectable in any case: multiple scattering in the concrete constitute the main source of background which completely wash away the faint signal coming from the 2 cm samples.

The results obtained in this preliminary study shows that there is potential for a successful application of the muon radiography technique to the monitoring of legacy nuclear waste storage sites. Dedicated work is however needed to improve the robustness of these results when taking into account real-world factors like non-homogeneous concrete, systematic effects of the Monte Carlo simulation, additional background contributions due to enlarged acceptances and surrounding buildings etc.

6. Acknowledgements

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