Antineutrino Detection Techniques for Monitoring Long-Term Geological Repositories

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Abstract:

The radioactive nuclear waste produced in the past decades contains many significant quantities of plutonium, one of the key ingredients for manufacturing nuclear weapons. For the first few hundreds of years after discharge, the dominant radioactivity of the waste comes from long-lived beta-decaying elements, in particular ^{90}Sr and ^{137}Cs. In this paper, we discuss the prospects of safeguarding long-term geological nuclear waste repositories by detecting the low-energy antineutrinos emitted via beta-decays. We investigate whether these antineutrino measurements could be carried out with time projection chambers, for example using liquid argon. In contrast to the typical scintillation or water-Cherenkov detectors, this emerging technology could detect antineutrinos even below the inverse beta decay kinematic threshold, i.e. the typical energy range for radioactive waste emissions. Furthermore, due to their imaging properties, time projection chambers could also provide directionality information which may be used for background rejection and potentially also for indicating if and where a certain amount of nuclear waste has been diverted. We present a first feasibility study for employing liquid-argon detectors for safeguarding geological repositories. We consider a realistic repository layout as a study case and evaluate the detector performance in this context, from first principles.

Keywords: radioactive waste; geological repositories; safeguards; antineutrino detection

1. Introduction

As nuclear programmes become older, safeguarding the radioactive waste becomes more important.

The International Atomic Energy Agency (IAEA) evaluated that the global cumulative amount of spent fuel (SNF) was approximately 380,500 tonnes heavy metal at the end of 2014 [1]. Furthermore, based on the output of the 438 reactors in operation in 2014, the IAEA estimated that about 10,000 tonnes of spent fuel are discharged every year. This implies that, presently, more than 430,000 tonnes spent fuel are stored around the world.

The typical spent nuclear fuel composition depends on the reactor burnup. Table 1 shows the evaluated composition of SNF extracted from a light-water reactor considering a burnup of 50GWd/t [2]:

Material	Relative amount
Uranium (< 1% ^{235}U, mostly ^{238}U)	93.4%
Fission products (^{129}I, ^{90}Sr, ^{135}Cs, etc.)	5.2%
Plutonium	1.2%
Minor actinides (^{237}Np, ^{241}Am, ^{243}Cm, etc.)	0.2%

Table 1: Typical isotopic composition of spent nuclear fuel [2]

The Institute for Science and International Security estimated that, at the end of 2014, the amount of civilian irradiated (i.e. present in spent fuel) and unirradiated (directly usable for nuclear weapons) plutonium was approximately 2,400 tonnes, increasing by approximately 50 tonnes per year [3]. A breakdown of the civil plutonium stocks per country at the end of 2014 is given in Fig. 1. The amount of irradiated plutonium is several factors larger than the unirradiated quantity. While the unirradiated plutonium is more susceptible for proliferation, also spent fuel could in principle be diverted to separate the plutonium.

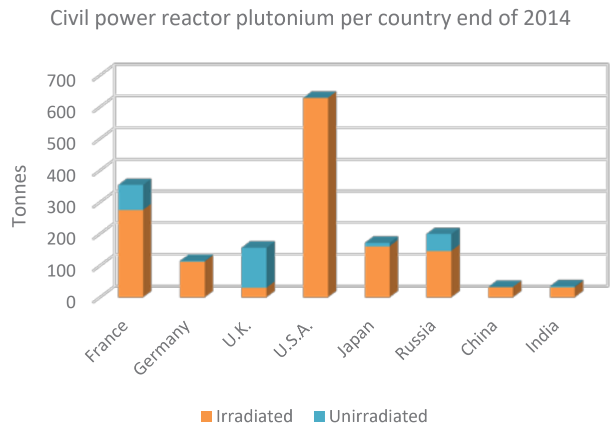

Figure 1: The amounts of irradiated and unirradiated plutonium held per country at the end of 2014. [3]

The spent fuel will eventually need to be stored in permanent geological repositories, which require safeguards to prevent diversion. In conceptualizing safeguards options, the IAEA currently focuses on the "early detection of unauthorized

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movement of fissile material" [4] which could occur in the form of e.g. unexpected excavations. Thus, the envisioned monitoring techniques include: (*i*) site inspections, (*ii*) aerial photography, (*iii*) satellite imagery and (*iv*) microseismic surveys [4].

A different approach that should be examined is the potential use of antineutrino monitoring in this context. If operated continuously, it could be a new tool for Continuity of Knowledge, providing redundancy to the foreseen containment and surveillance measures. A change in the antineutrino flux, once detected, serves as clear, difficult to fake evidence of clandestine material diversion. Furthermore, it could be used in a re-verification context, by comparing the detected antineutrino rate and spectra to what would be expected, either from measurements before the loss of Continuity of Knowledge, or from results of a repository simulation model built from information provided in the inventory declaration. Even a rough estimate of the potential diversion region may be possible.

In this paper, we discuss the potential use of antineutrino monitoring by presenting first results of a feasibility study that focuses on imaging detectors, which may provide significant advantages compared to other detector types. The paper is organized as follows: in section 2, we discuss the main properties of the antineutrinos emitted by the radioactive waste and the need for new detection techniques. The potential way forward, i.e. time projection chamber detectors, is presented in section 3 with a focus on liquid-argon-filled detectors. The first results of our feasibility study are summarised in section 4.

2. Antineutrinos for Monitoring Geological Repositories

The concept of antineutrino monitoring of spent nuclear fuel has been proposed by Brdar et al. [5]. In the first few hundreds of years, the dominant radioactivity of the waste stems from the beta-decaying fission products. While many isotopes have rather short half-lives (in the order of several hours or a few days), in particular ^{90}Sr ($T_{1/2}$ = 28.78 years) and ^{137}Cs ($T_{1/2}$ = 30.17 years) contribute also decades later.

Due to their weakly interacting nature, with cross-sections lower than 10^{-38} cm^2, the antineutrinos escape even large amounts of shielding and can propagate through the geological structures surrounding the repository. There is no means to deflect the antineutrinos' trajectories. At the same time, the prospects of using antineutrino measurements in a safeguards context will be analysed here in view of the potentially long measuring times due to their low cross-sections.

2.1 Properties of Antineutrino Emissions from Radioactive Waste

To evaluate the antineutrino emissions from radioactive waste, the operation of a pressurized water reactor (PWR) was simulated with SERPENT 2 [6], a Monte Carlo neutron transport and burnup calculation code. The reactor model was based on the description given in [7] and it was implemented with an enrichment of 4% and burnup of 33 MWd/kg.

Antineutrino emission spectra (Fig. 2) were computed at seven different time steps between 100 and 500 years from the considered moment of discharge by summing over the spectra for each beta-decaying nuclide.

As can be seen, the antineutrino emissions have a low energy range, i.e. from a few eV up to approximately 4.3 MeV. This constitutes a challenge for the antineutrino detection, since typical reaction cross-sections decrease at lower energies. However, it can also be observed that, even after more than 200 years since discharge, a significant number of antineutrinos, i.e. $\mathcal{O}(10^{12})$ for energies integrated below 2.3 MeV, are still emitted.

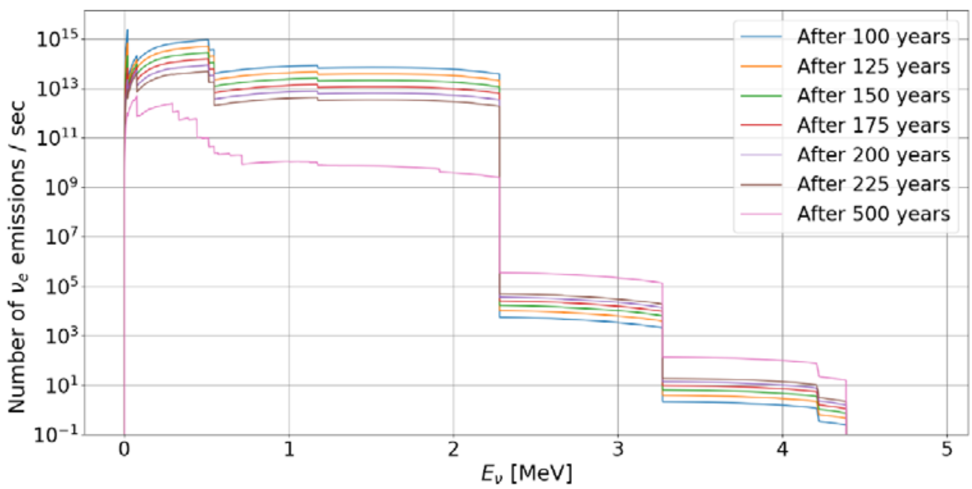

Figure 2: Antineutrino emission spectra from one fuel assembly of a simulated PWR reactor with a burnup of 33 MWd/kg and enrichment of 4% computed at seven different time intervals from the simulated discharge [8].

2.2 Motivation for New Antineutrino Detection Methods in the Safeguards Context

The use of antineutrino measurements for monitoring purposes has already been considered in the case of nuclear reactors, i.e. for determining reactor shut-down periods or estimating the plutonium content in the core. Since antineutrinos interact only weakly, the detector must provide a large mass and a high target density to achieve a reasonable detection time. Two research projects in this context are WATCHMAN [9] and CHANDLER [10].

In both the WATCHMAN and CHANDLER detectors (a water-Cherenkov and scintillation detector, respectively), the anti-neutrinos interact with the sensitive material by means of the inverse beta decay (IBD) reaction: $\bar{\nu}_e + p \rightarrow e^+ + n$, which is characterised by a kinematic threshold: it can only occur for antineutrino energies E_ν larger than 1.8 MeV. As can be seen from Fig. 2, relying exclusively on the IBD reaction, one could detect less than 20% of the antineutrinos coming from the described radioactive waste. Consequently, a sensitive material which would enable antineutrino reactions without an intrinsic energetic threshold would be desirable.

Furthermore, for safeguards purposes, a directionality capability would be helpful, especially since the antineutrino emission rate decreases with time: reconstructing the incoming direction of the antineutrino emissions could be used for background rejection and potentially also for indicating where a certain amount of nuclear waste has been diverted. While not impossible, it is very challenging to obtain directionality information from the IBD reaction – a large number of detected events ($\mathcal{O}(10^3)$) is required to locate the source with an angular resolution of a few degrees [11].

Recently, as the interest in the field of fundamental neutrino research is gradually encompassing the lower energy range of solar and supernova antineutrinos, new detection methods are proposed and/or prototyped. Antineutrino detectors based on time projection chambers filled with liquefied noble gases or organic-liquid scintillators seem to be particularly promising.

3. Time Projection Chambers for Antineutrino Detection

The idea of using time projection chambers (TPCs) with noble gases for neutrino detection was first proposed by Carlo Rubbia in 1977 [12]. It is only in the last decade that TPC prototypes have been realised and tested in the field of fundamental neutrino physics and dark matter detection.

3.1 Fundamentals of TPCs

The TPCs considered for antineutrino detection typically consist of a large volume of liquefied noble gas like argon or xenon. Argon constitutes approximately 1% of Earth's atmosphere, especially the ^{40}Ar isotope with an abundance of 99.6%, it is usually cheap to produce (and to liquify) and it is commercially available. The volume is encompassed by a high-voltage cathode on one side and an anode on the opposite surface which also contains several read-out wire planes. The uniform electric field realised between the cathode and the anode planes typically has a strength of 500 V/cm. To be liquid, the noble gas must be cooled to a very low temperature, e.g. 87K (-186.15° C) in the case of argon. More recently, organic-liquid scintillating materials like tetramethylsilane appear to be very promising, especially since they can operate at room temperature.

When an (anti-)neutrino interacts via charged or neutral current exchange with an atom in the sensitive material, i.e. either with the electrons or the nucleus itself, the emergent particles ionise and excite further atoms along their trajectory. The emitted free electrons drift in the liquid, under the force of the electric field, until they reach the read-out wires, in which they generate small currents. The wires are placed at very close distance to each-other, e.g. 3-5 mm, and constitute a very dense net. To obtain multi-dimensional information about the charged particles' tracks, several wire planes can be used, placed under different angles with respect to each other. In addition, the excited atoms also emit scintillation light which can be measured with photosensors (PMTs). The light signal can be used to trigger the signal acquisition. Liquid argon is an excellent scintillator, providing approximately $4{\times}10^4$ photons per MeV [13].

Thus, one of the main benefits of using time projection chambers stems directly from their mode of operation: unlike water or scintillator detectors, they are *imaging* detectors – providing a three-dimensional reconstruction of the tracks left by the charged particles emerging from an antineutrino interaction. It enables a reconstruction of the antineutrino energy and its incoming direction on an event-by-event basis, which is helpful for background rejection.

TPCs filled with an organic-liquid scintillator that can operate at 15-20°C would be well suited for monitoring geological repositories since they require no additional cryogenic infrastructure. However, more research and development work are required to demonstrate the feasibility and performance of these proposed sensitive materials.

In contrast, liquid-argon-based detectors (LArTPCs) require cooling but have already been built and tested in fundamental physics experiments like, e.g. ArgoNeuT [14] and MicroBoone [15]. Furthermore, they are considered for large-scale neutrino fundamental research experiments like DUNE [16]. Consequently, we will focus on liquid-argon TPCs in the rest of this paper.

3.2 Antineutrino Interactions in Liquid-Argon

In the standard water-Cherenkov or organic-scintillator-based detectors, antineutrinos mostly interact by means of the inverse beta decay process: $\bar{\nu}_e + p \rightarrow e^+ + n$. However, since there are no free protons available in liquid argon, this reaction cannot take place. The analogous reaction in liquid argon is the charged current absorption: $\bar{\nu}_e + {}^{40}\text{Ar} \rightarrow e^+ + {}^{40}\text{Cl}^*$. However, its energy threshold in the case of antineutrinos is even higher: $E_\nu > 7.5$ MeV, which makes it unfeasible for the antineutrino energy range relevant for safeguarding geological repositories (cf. Fig. 2).

In the elastic scattering reaction, the anti-neutrino scatters off electrons of the argon atoms: $\bar{\nu}_e + e^- \rightarrow \bar{\nu}_e + e^-$. This reaction is very important since, unlike IBD, it can provide the desired directionality information directly: the energy of the incoming antineutrino is usually larger than the rest mass of the electron, therefore, the latter carries away most of the antineutrino momentum and its direction is correlated to that of the incoming antineutrino.

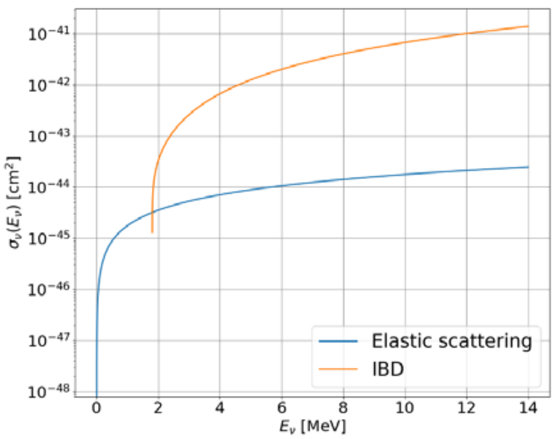

Figure 3: The energy-dependent antineutrino cross-sections f or the elastic scattering and IBD reactions, based on the analytical calculations described in [17] and [18], respectively.

However, the cross-section of the elastic scattering reaction is about two orders of magnitude lower than the one of IBD, as can be seen from Fig. 3. This clearly poses a detection challenge and is somewhat compensated by the higher target density provided by the liquid-argon. Nevertheless, it can also be seen that the reaction is kinematically allowed for the entire energy range relevant for the antineutrino emissions from radioactive waste.

A second reaction that can occur in liquid argon is the coherent elastic neutrino-nucleus scattering (CEνS). It occurs via weak neutral current and essentially refers to antineutrinos being scattered off entire argon nuclei. The cross-section of this reaction in liquid argon is expected to be approximately of the order of $\mathcal{O}(10^{-38})$ cm^2, i.e. about three orders of magnitude higher than the IBD cross-section.

The CEνS cross-section is presently being measured by experiments such as e.g. COHERENT [19]. Despite the high cross-section, the recoil energy of the nucleus is very low, in the order of a few tens of keV, and it remains to be seen whether this reaction is detectable at all in a LArTPC. Consequently, the CEνS interaction is not taken into account in the present study. Thus, the antineutrino elastic scattering off the electrons in the argon atoms is the interaction considered in the following.

Liquid argon (^{40}Ar) provides a target density comparable to water and scintillator materials, e.g. like the one used for the KamLAND neutrino detector [20]. Table 2 shows a comparison of several sensitive materials in terms of the number of expected events in 80 tonnes of material. For this comparison 500 tonnes of spent nuclear fuel (SNF) with a cooling time of 100 years have been considered and treated as a point source for the antineutrino emissions. A distance of 50 m between the detector and the emissions' source was assumed. Furthermore, only two signal interactions were considered: the elastic scattering off electrons in liquid argon and the inverse beta decay reaction for water and the scintillator materials, respectively. The reaction cross-sections and the expected number of targets in the sensitive material volume have been computed accordingly. For each material, a detection time of 100 days was assumed.

Detector type	Nr. Events in 80 t and 100 days
LArTPC	128
Water-Cherenkov	93
Scintillator (KamLAND [20])	204

Table 2: Comparison between liquid argon and other typical antineutrino sensitive materials in terms of the expected number of SNF antineutrino events in 80 tonnes of material. The computation assumes the SNF is 100 years old. The relatively high target density of liquid argon compensates the lower elastic scattering cross-section. The main advantage of the LArTPC technology, however, remains its directionality capability.

4. Prospects for Safeguarding Geological Repositories with Liquid-Argon Detectors

To evaluate the feasibility of monitoring or re-verifying long-term geological repositories by measuring antineutrino emissions with liquid-argon detectors, a study case was defined. Four storage areas in the eastern region ("Ost 5", "Ost 6", "Ost 7" and "Ost 8") of the proposed "B1" layout for the planned repository in Gorleben, Germany were considered [21]. Presently, Gorleben is no longer regarded as a potential location for a long-term geological repository and the scientific investigations were stopped in 2012. Nevertheless, the initial studies were detailed enough to form the basis for a simulation of the repository.

The study case repository comprises 1120 containers with 10 fuel assemblies each. While the different ages of the fuel assemblies were correctly taken into account, we assumed that only fuel assemblies from pressurised water reactors with 55 MWd/kg burnup are stored. This is a simplification, as a detailed model is beyond this paper's scope. The B1 repository layout foresaw that the storage caverns would be located at a depth of 870 m [21]. In the study case, it was envisaged that the antineutrino detector would be placed at the same depth as the repository, but 100 m away from the nearest containers and completely decoupled from the rest of the repository, i.e. in a cavern of its own. In view of the transport and deployment requirements, it was considered that the antineutrino detector would have the dimensions of a regular shipping container. 80 tonnes of liquid-argon constitute about 70% of the volume, while the remaining 30% are necessary for the thermal insulation which enables the argon to be maintained in a liquid form. For the preliminary evaluation discussed here, it was assumed that the entire sensitive volume is fully instrumented and that the detection efficiency is 100%.

4.1 Expected Detection Rate

The detection rate in the liquid-argon detector considered for the study case was calculated according to: $N_\nu(E_\nu) = \Phi_\nu(E_\nu) \cdot T \cdot \sigma(E_\nu)$, where Φ_ν is the energy-dependent antineutrino flux at the location of the detector, T is the number of targets in the detector active volume and σ is the differential cross-section for elastic scattering shown in Fig. 3. The antineutrino flux is expressed as the number of emissions per second and per cm^2 and it takes into account the distance between the detector and the emissions' source. The number of targets T was computed based on the target density expressed in number of electrons per cm^3. The detection rate was then scaled up to the detector size.

Table 3 shows the antineutrino detection rate for different times after 2020. For this study, an optimistic detection efficiency of 100% has been assumed since the efficiency of the LArTPC technology in the low energy range is presently being investigated.

Time elapsed since 2020 [years]	Number of antineutrino events detected within 3 months
100	1116
125	615
150	339
175	187
200	103
225	57
500	< 1

Table 3: Total detection rates of the antineutrino emissions (without background), based on a study-case layout of a geological repository comprising 11200 fuel assemblies of different ages and with a burnup of 55 MWd/kg. [8]

It can be seen from Table 3 that the expected detection rate diminishes significantly with time, as the amount of beta-decaying isotopes present in the radioactive waste decreases. Nevertheless, the number of observed antineutrinos is above 100 even after year 2220.

The background reduction efficiency was optimistically assumed to be 100% in this preliminary study. The largest background contributions stem from the solar neutrinos (120 counts per day in an 80 t LArTPC) and the cosmogenics, i.e. beta-decaying elements produced in detector materials via spallation induced by high-energetic cosmic muons (39 counts per day). This estimation is based on [22], [23] and it assumes an overhead of 4300 meters water equivalent. The expected number of background events will vary based on the depth and the geology of the specific repository location.

In this preliminary evaluation, the expected number of background events is almost a factor 10 larger than the number of signal events. To obtain a signal-to-noise ratio larger than 1, we estimate that the background reduction should have an efficiency of at least 92%. Methods of background reduction like e.g. using the directionality information to eliminate solar neutrinos and using a muon veto to reduce the cosmogenic background form the subject of our future research.

To understand the significance of the obtained detection rates, potential diversion scenarios were analysed.

4.2 Diversion Scenario

We assume that x % of the amount of radioactive waste was covertly diverted from the repository. At the same time, different data-taking times up to 24 months of continuous measurements were considered. The time required to detect a diversion of x % from the amount of radioactive waste with a certain confidence level was evaluated by hypothesis testing (the detection rate follows a Gaussian distribution, with a known standard deviation due to Poisson statistics).

The scenario was evaluated for two different times: year 2120 (Fig. 4) and year 2220 (Fig. 5).

It can be seen from Fig. 4 that, in year 2120, a 4% diversion can be detected with 95% confidence after about 12 months of data-taking. As expected, after another 100 years, the antineutrino detection rates decrease such that approximately a 14-15% diversion can be detected at a 95% confidence level.

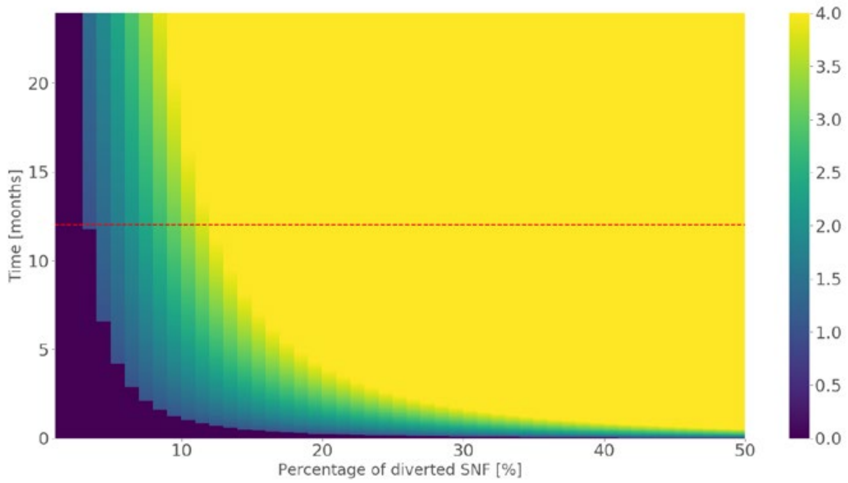

Figure 4: Results of the diversion hypothesis testing assuming that data-taking occurs in year 2120. The colour code shows the confidence level of the detection expressed in standard deviations. The dotted red line marks the detection time of 12 months.

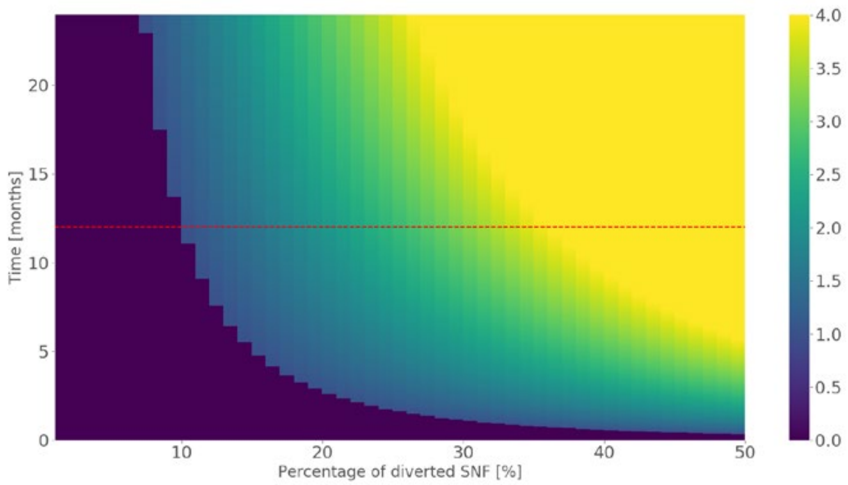

Figure 5: Results of the diversion hypothesis testing assuming the data-taking occurs in year 2220. The colour code shows the confidence level of the detection expressed in standard deviations. The dotted red line marks the detection time of 12 months.

The detection times can be reduced by choosing a larger detector volume. However, from layout considerations, detection times can be further reduced by deploying several - perhaps even smaller detectors - at different locations in the repository's proximity.

5. Conclusions and Outlook

As the quantity of radioactive waste increases worldwide, the potential risk of diversions in the context of unforeseeable societal and political events informs the necessity of safeguarding long-term geological repositories. The only messenger particles that could escape the confinement of the repository are the antineutrinos emitted decades later through the beta decays of isotopes like ^{90}Sr and ^{137}Cs still present in the waste. Presently, measuring the antineutrino emissions is the only approach that can offer direct information about the amount of radioactive waste present in the repository.

In this paper, we proposed the use of time projection chambers (TPCs) for measuring antineutrino emissions. For liquid argon, this technology is presently developed and validated by the neutrino physics community. More research and development work is required in order to demonstrate the feasibility of organic-liquid sensitive materials. Using time projection chambers to measure the antineutrino emissions presents several advantages in comparison to traditional detectors: (*i*) excellent track imaging capabilities – used for efficient background subtraction and locating where a potential diversion occurred, (*ii*) the absence of a kinematic threshold in the detection reaction which would otherwise limit the accessible antineutrino energy range and (*iii*) the relatively low production costs.

We performed a preliminary calculation of the expected event rate in LArTPCs and interpreted it in the context of a diversion scenario. We concluded that after 100 years since the closing of the geological repository a 4%

diversion can be detected within 12 months when using a single 80 t antineutrino detector. The required detection time increases with the age of the waste. Using several TPCs, placed at different locations in the vicinity of the repository, would significantly decrease the required detection time.

However, this evaluation assumes perfect detector efficiency and background rejection. A more realistic estimation of the detection and background rejection efficiencies could lead to an increased detection time. For a realistic feasibility study, the LArTPC performance should be studied in a full simulation that also takes the solar, cosmogenic and geoneutrinos background into account. This simulation together with a detailed treatment of the background forms the subject of our future studies.

6. Acknowledgements

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