

## Antineutrino Monitoring of Spent Nuclear Fuel

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Military and civilian applications of nuclear energy have left a significant amount of spent nuclear fuel over the past 70 years. Currently, in many countries worldwide, the use of nuclear energy is on the rise. Therefore, the management of highly radioactive nuclear waste is a pressing issue. In this paper, we explore antineutrino detectors as a tool for monitoring and safeguarding nuclear-waste material. We compute the flux and spectrum of antineutrinos emitted by spent nuclear fuel elements as a function of time, and we illustrate the usefulness of antineutrino detectors in several benchmark scenarios. In particular, we demonstrate how a measurement of the antineutrino flux can help to reverify the contents of a dry storage cask in case the monitoring chain by conventional means gets disrupted. We then comment on the usefulness of antineutrino detectors at long-term storage facilities such as Yucca mountain. Finally, we put forward antineutrino detection as a tool in locating underground “hot spots” in contaminated areas such as the Hanford site in Washington state.

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### I. INTRODUCTION

With carbon-dioxide-induced climate change and the scarceness of fossil fuels becoming imminent problems for humanity, nuclear energy is undergoing a renaissance. However, nuclear technology comes with a number of intrinsic problems, such as the limited availability of nuclear fuel, the danger of proliferation of nuclear weapons technology, the risk of major accidents, and the management of highly radioactive waste. As a result, nuclear energy is relatively expensive compared to many other energy sources.

In this paper, we, in particular, focus on the waste issue: We argue that a measurement of the antineutrino flux emitted by beta-decaying isotopes can be a unique component in a multifaceted approach to monitoring and safeguarding nuclear-waste repositories. The unique advantage of antineutrinos is that they penetrate the shielding surrounding the repository and thus offer a direct method for remotely probing the nuclear material inside. Other probes like gamma rays or neutrons (see, for instance, Ref. [1]) are heavily attenuated by the materials they need to traverse on the way to a detector.<sup>1</sup> Therefore, relating their measured fluxes to the actual content of the repository requires a sophisticated propagation model, which in turn relies on an accurate knowledge of the contents of the repository. This cyclic dependence on information is one of

the major limitations of conventional monitoring methods. On the downside, the very fact that antineutrinos are not attenuated even by a whole mountain implies that antineutrino detection has to deal with very small cross sections  $\lesssim 10^{-41}$  cm<sup>2</sup> [3]. Any meaningful flux measurement thus requires the deployment of a large detector with at least several tons of active material for a time period of the order of months.

Nevertheless, thanks to advances in detector technology, this appears feasible at a comparatively reasonable cost. In fact, practical applications of antineutrino detectors in the nuclear industry have been discussed for a long time, mostly in the context of monitoring power reactors [4–10]. Several detectors have been built to demonstrate the feasibility of such applications [11,12], and further studies are planned in current and future experiments [13].

In the following, we first compute the antineutrino flux and spectrum emitted by spent nuclear fuel and then consider several scenarios in which antineutrino detectors can be used in the context of radioactive waste repositories.

### II. ANTINEUTRINO EMISSION FROM SPENT NUCLEAR FUEL

For the first 1000–10 000 yr after discharge from a reactor, the total activity of spent nuclear fuel is nearly exclusively caused by beta decays (and the associated gamma emission). Therefore, a large number of antineutrinos are produced. However, detection by inverse beta decay,  $\bar{\nu}_e + p \rightarrow n + e^+$ , the main detection reaction for electron antineutrinos, requires antineutrino energies of at least 1.8 MeV. The lifetime of a beta-decaying nucleus

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<sup>1</sup>For a recent study on muon radiography applied to the problem at hand, see Ref. [2].

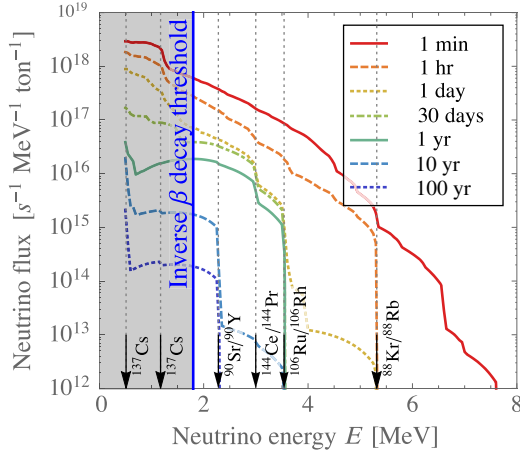


FIG. 1. The spectrum of electron antineutrinos emitted by spent nuclear fuel as a function of the time after discharge from the reactor. We also indicate in gray the area below the threshold for inverse beta decay, the dominant antineutrino-detection process, at 1.8 MeV. The data underlying this plot are available in Supplemental Material [20].

scales roughly like  $Q^5$ , where  $Q$  is the energy released in the decay. Therefore, the detectable antineutrino signal for most fission fragments decays within hours to days after fission ends. There are, however, a handful of isotopes that have a two-stage decay, where the first decay has very small  $Q$ , and thus a resulting long lifetime, followed by a fast decay with  $Q > 1.8$  MeV. The most notable example is strontium-90, which decays with a half-life of 28.90 yr to yttrium-90, which in turn decays within hours to the stable zirconium-90 with  $Q = 2.22801$  MeV [14]. Strontium-90 is produced in around 5% of all fission events [15–17]. The isotopes with the next-longest lifetimes with antineutrino emission above 1.8 MeV in their decay chains are ruthenium-106 (371.8 days [18]) and cerium-144 (284.91 days [19]). As a result, the detectable antineutrino emission of spent nuclear fuel after more than a few years is entirely given by strontium-90. It is worth noting that strontium-90 (like all other fission fragments) remains in the high-level waste resulting from reprocessing using the widely employed plutonium uranium redox extraction (PUREX) process. In Fig. 1, we plot the number of electron antineutrinos emitted per second, per MeV, and per ton of spent nuclear fuel as a function of the antineutrino energy for fuel elements of different ages. We assume a burnup<sup>2</sup> of 45 GW days. As expected, we observe a softening of the spectrum over time, as short-lived isotopes with large  $Q$  values decay away. Note, however, that even after 100 yr a

<sup>2</sup>Burnup is a measure of how much energy per unit mass has been extracted from nuclear fuel. It is directly proportional to the total number of fissions and thus to the strontium-90 content and the antineutrino emission rate.

nonzero flux remains above the energy threshold of 1.8 MeV for inverse beta decay.

### III. DRY-CASK STORAGE FACILITIES

As long-term storage facilities for spent nuclear fuel are becoming available only slowly, temporary storage solutions have become a necessity. Once fuel elements have been allowed to cool in a spent fuel pool for approximately 10 yr [21,22] after discharge from the reactor, they are typically transferred to dry storage casks, large shielded steel cylinders several meters tall, each of them holding approximately 14–24 tons of spent nuclear fuel elements with a uranium content of 10–17 tons [22–24]. The layout of a typical dry storage facility is shown in Fig. 2. Even though safety and security measures are in place to protect such facilities, manipulations are imaginable. The core of the International Atomic Energy Agency’s methodology for spent fuel is so-called continuity of knowledge (CoK): The amount and type of fuel loaded into a cask is monitored and recorded, the cask is closed, and a tamper-proof seal is applied. As long as the seal is intact and the records are available, the resulting CoK allows one to infer with a great deal of certainty the contents of the cask. However, even during routine operations it is conceivable that records are inaccurate or lost or that seals are compromised. Several methods based on neutron or gamma-ray detection are under development to restore CoK in this case; see, for instance, Ref. [1].

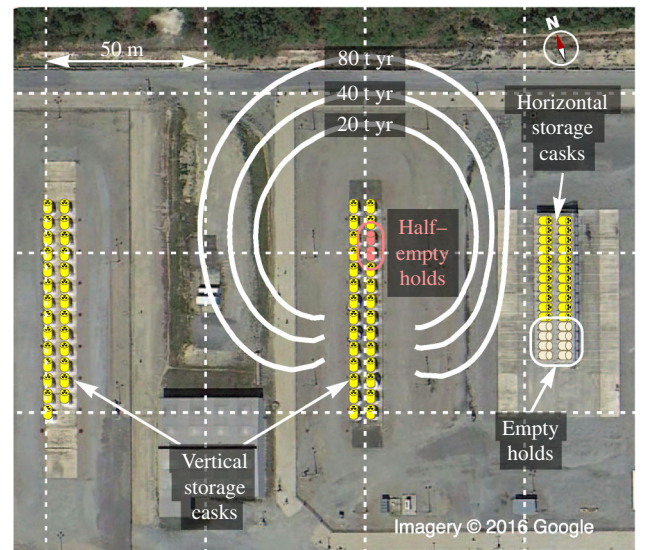


FIG. 2. The dry storage facility at the Surry Nuclear Power Plant in Virginia, USA [23]. Filled storage casks, highlighted in yellow, contain 9–16 MTU each. In the benchmark scenario discussed in the text, we assume that 50% of the spent fuel in two 15-MTU casks (marked in red) has gone missing. Colored contours indicate the exposure (in ton years) required to establish the loss of nuclear material at the 90% confidence level.

Here, we envision instead the deployment of an antineutrino detector, with a fiducial target mass<sup>3</sup> of the order of approximately 20 tons, close to the storage casks for several months. Using as an example the storage facility at the Surry Nuclear Power Station in the U.S., where casks hold 9–16 metric tons of uranium (MTU), we assume that 50% of the radioactive material from two of the 15-MTU casks (colored in red in Fig. 2) goes missing. This roughly corresponds to removing 3% of the total amount of nuclear waste stored at Surry. We make no claim that an actual diversion case would have any similarity to this scenario nor that this could occur as part of routine operations; it merely serves to indicate the general level of sensitivity we might expect from antineutrino monitoring.

To determine what it takes to discover such an anomaly, we simulate the expected number of detected antineutrino events as a function of the detector position for the two hypotheses “all storage casks full” ( $F$ ) and “50% of nuclear material missing in two casks” ( $M$ ). We use the antineutrino spectrum given by the blue dashed curve in Fig. 1 (10 yr after discharge) and the inverse beta-decay cross sections from Ref. [3]. Neutrino oscillation effects, though small, are taken into account, with the oscillation parameters given in Ref. [25]. The rate of antineutrino events per ton of fiducial detector mass and per MTU of source mass is

$$N_\nu = 5.17 \text{ yr}^{-1} \text{ ton}^{-1} \text{ MTU}^{-1} \times \left(\frac{10 \text{ m}}{d}\right)^2, \quad (1)$$

where  $d$  is the distance between the source and the detector (both treated as pointlike). This number depends mildly on the time after discharge and is, for instance, reduced by approximately 5% one year later. In the following, we always assume measurement campaigns lasting one year or less and therefore neglect this small effect.

The irreducible background to the measurement includes antineutrinos from running nuclear reactors with an expected event rate of

$$N_{\text{bg}} = 359 \text{ yr}^{-1} \text{ ton}^{-1} \text{ GWth}^{-1} \times \left(\frac{\text{km}}{d}\right)^2. \quad (2)$$

For the 5.2 GWth (thermal power) reactor in Surry, located  $d \sim 1$  km away from the envisioned 20-ton detector, this leads to approximately 37 300 antineutrino events per year. We take this background into account in our simulations. Backgrounds from other power stations and from radioactive decays in Earth (geoneutrinos) are smaller by at least a factor of about  $10^{-4}$ , and we therefore neglect them.

The dominant reducible backgrounds arise from radioactive decays and cosmic-ray interactions mimicking an antineutrino signal. With current single-volume liquid

<sup>3</sup>The fiducial detector mass is the effective mass, after accounting for fiducial volume cuts and efficiency factors introduced in the event reconstruction and analysis.

scintillator detectors like Double Chooz, RENO, and Daya Bay, when deployed at the surface, these backgrounds would be a factor of 10–10 000 larger than the anticipated antineutrino signal. Current detectors identify signal candidates by looking for a delayed coincidence between a primary particle and a delayed neutron capture. However, they are not able to exploit the spatial correlations between the primary and delayed signals, nor can they tell whether the primary particle is a positron, as in inverse beta decay, or a photon or electron, as in most background events. Fortunately, these shortcomings could be overcome in a detector with sufficient spatial resolution to tag positrons by resolving the two 511-keV x rays from their annihilation [26]. Prototypes of detectors with this capability exist and have been successfully operated in particular by the SoLid and CHANDLER Collaborations [13]. Currently, an improvement of the signal-to-background ratio by a factor of 1000 is achievable, and further improvements appear feasible with improved shielding and an increased concentration of neutron capture targets like lithium-6. It thus appears plausible that within a few years even the low rate of antineutrinos from nuclear waste will become detectable in surface detectors. In the following, we assume that this has been achieved by the time the proposed measurements are carried out, and we neglect reducible backgrounds.

Events are divided into 0.2-MeV-wide energy bins. Denoting the number of signal events expected under the two alternative hypotheses by  $F_i$  and  $M_i$  and the number of background events by  $B_i$ , we define the test statistic

$$\chi^2 \equiv 2 \sum_i \left\{ F_i - M_i + (M_i + B_i) \log \left[ \frac{M_i + B_i}{F_i + B_i} \right] \right\}, \quad (3)$$

which follows a  $\chi^2$  distribution.

The results of the analysis are represented by the contours in Fig. 2, which indicate where the antineutrino detector should be placed in order to establish the flux deficit at the 90% confidence level with 20, 40, and 80 ton yr of exposure, respectively. We see that the detector needs to be placed within approximately 50 m of the affected casks in order to collect the approximately 4000 events needed for the measurement.

#### IV. APPLICATION TO LONG-TERM STORAGE FACILITIES

Above-ground storage of spent nuclear fuel, while widely used, is only a temporary solution, and the long-term goal must be to establish underground repositories that can keep radioactive material out of the biosphere for  $10^4$ – $10^6$  yr [27]. The usefulness of antineutrino detectors at such geological repositories is limited by the low antineutrino fluxes after strontium-90 has decayed away (half-life 28.8 yr). Moreover, in order not to disturb the repository, the construction of antineutrino detectors seems



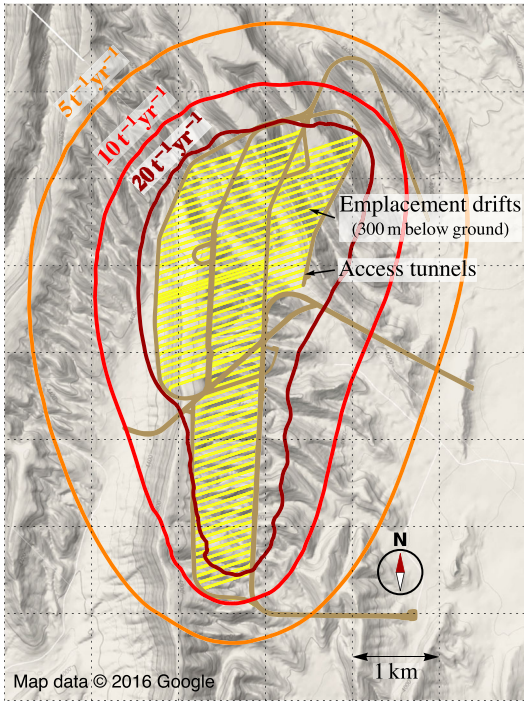


FIG. 3. The planned long-term storage facility at Yucca mountain. The yellow grid indicates the drifts holding the radioactive material at a depth of 300 m below the surface, while red and orange contours show the expected antineutrino count rates for a detector at the surface.

feasible and useful only at distances of the order of 100 m or larger.

To illustrate the prospects of detecting antineutrinos from a geological nuclear-waste repository, we show in Fig. 3 the signal event rates expected at the proposed Yucca mountain repository in Nevada, which would hold 70 000 MTU of radioactive material, stored approximately 300 m underground. We see that even a small detector (approximately 10 tons) located at the surface would see an appreciable event rate. With a kiloton-scale instrument like KamLAND [28] or the planned JUNO experiment [29], count rates would be significantly larger, especially when such a detector is placed in an underground location closer to the repository. Even then, however, it would be possible to detect only cataclysmic disruptions of the repository. More typical (but nevertheless highly dangerous) failure scenarios that involve the leakage of only a small amount of nuclear material into the surrounding soil would not be detectable. This may change, however, once detector technology with better directional sensitivity becomes available (see below).

## V. LEAKAGE OF NUCLEAR MATERIAL AT THE HANFORD SITE

Sometimes, nuclear oversight agencies are faced with the challenge to secure or decommission a nuclear-waste repository in which the contents, and perhaps even the

underground location, of storage casks are not known. An example is the Hanford site in the state of Washington (USA), where plutonium for military purposes was produced from 1944 to 1987. At Hanford, a major problem is the leakage of storage containers for high-level nuclear waste, leading to radioactive contamination of ground water [30,31].

Consider first a scenario where the location of storage tanks is known, but their precise content is not. We focus on one particular array of storage tanks at Hanford, the T tank farm [30], which consists of 16 tanks, arranged in a  $4 \times 4$  grid measuring approximately  $120 \text{ m} \times 80 \text{ m}$  and originally containing between 0.2 and 5 MTU of spent fuel each. We assume the nuclear material in the tanks was discharged from a reactor 50 yr ago. With a detector placed 30 m from the most massive (5-MTU) tank and taking into account background antineutrinos from other storage tanks and from the Columbia nuclear power plant (30 km away, 3.5 GW thermal power), the amount of material in that tank can be measured with an uncertainty of  $\pm 2.1$  MTU for an exposure of 20 ton yr (35 signal events) and with an uncertainty of  $\pm 1.1$  MTU for an exposure of 80 ton yr (140 signal events). The age of the nuclear material (i.e., the time after discharge) can be determined to lie between 44 and 54 yr with 80 ton yr of exposure, assuming the true age is 50 yr.

Assume now that a fraction of the radioactive material in the most massive tank is slowly leaking out. We model this situation by reducing the inventory of the tank and introducing a secondary point source containing the leaked material 20 m below its original location. Detecting such a leakage seems unfeasible with established detector technologies but requires antineutrino detectors that measure not only energy but also the direction of incoming antineutrinos. Some preliminary efforts in this direction have been undertaken [26,32,33], but a working detector is still far off. As one of the goals of the present study is to motivate further R&D in this field, we in the following assume the availability of a compact detector with an expected angular resolution down to  $\mathcal{O}(10)$  degrees [26]. We bin events in the cosine of the zenith angle  $\cos \theta$  (five bins) and the azimuth angle  $\phi$  (nine bins). We use a highly simplified model of angular smearing in terms of a Gaussian with a width of  $20^\circ$ . For simplicity, we integrate over energy, assuming that the time of discharge and thus the antineutrino energy spectrum are known already. We estimate that, by deploying a directionally sensitive 20-ton detector at a distance of 30 m from the damaged tank, a leakage of 55% of the tank's content can be discovered at 90% C.L. after 12 months (30 signal events). With an 80-ton detector, the detection of 25% leakage is possible.

## VI. RADIOACTIVE SPILL AT HANFORD BUILDING 324

As a further application scenario, we consider an actual spill of radioactive material that happened in October 1986 in a radiochemical plant at Hanford known as building 324. At the time, a large amount of strontium-90 and caesium-137,

with a total activity of 1.3 MCi, was released from a hot cell. Half of it leaked into the ground and is now presumed to be located within a  $10\text{ m} \times 10\text{ m}$  area about 2 m below ground level [31]. In 2010, a pit was excavated approximately 20 m from the spill to drive long steel pipes into the affected area, thus allowing the deployment of temperature and activity sensors. This method has the disadvantage that it allows moisture to enter the contaminated soil, which may ultimately allow radioactive material to seep further into the ground, possibly reaching ground-water levels. For future incidents of this type, we therefore consider the deployment of antineutrino detectors for remote sensing. Modeling the spill as a point source at a depth of 2 m and assuming the availability of an 80-ton antineutrino detector with angular sensitivity located 30 m away at the same depth, we find that a further downward shift of the nuclear material by 3.5 m is detectable at the 90% C.L. after one year of exposure.

## VII. LOCALIZING NUCLEAR-WASTE CONTAINERS

Let us now turn to a more speculative scenario where neither the exact location nor the contents of storage casks are known. This could happen, for instance, when documentation is lost and localization using other methods like ground-penetrating radar is not feasible, for instance, in a scenario where many casks are buried underground but only a few contain high-level nuclear waste. We envision successive or simultaneous deployment of 80-ton antineutrino detectors on a two-dimensional grid with a spacing of 250 m. We again assume angular sensitivity, but, since the distance between detectors and sources is large, the zenith-angle measurement is irrelevant and can be discarded. Figure 4 illustrates the outcome of such an analysis for four randomly placed storage casks of unknown content and for

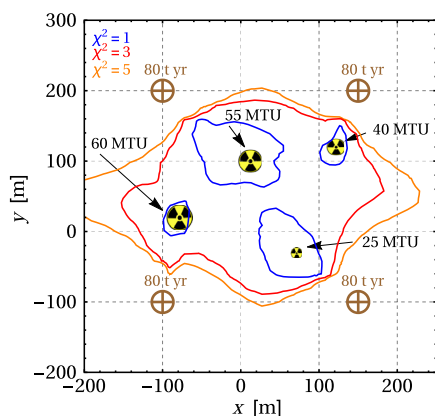


FIG. 4. Using antineutrino detectors (brown  $\oplus$  symbols) to localize nuclear-waste storage casks (radioactive hazard symbols). Colored contours indicate the accuracy with which casks can be localized (see the text for details). We assume an exposure of 80 ton yr per detector, and we use the antineutrino spectrum expected 50 yr after discharge from a reactor.

an exposure of 80 ton yr per detector. Using a stochastic optimization method, we fit the positions  $(x_j, y_j)$  and activities  $m_j$  of the four sources. Colored contours show the dependence of the test statistic  $\chi^2$  [defined in analogy to Eq. (3)] on the fit values of  $(x_1, y_1)$ , with the other  $(x_j, y_j)$  as well as all  $m_j$  allowed to float. We see that storage casks can be localized to within tens of meters. For a further refinement of their position (for instance, in order to guide cleanup efforts), the procedure can be repeated with detectors moved closer to the source positions determined in the initial scan. With an approximately 30-m spacing between detectors, the position of each source can be determined to  $\mathcal{O}(m)$  accuracy.

## VIII. SUMMARY

We calculate the antineutrino flux and spectrum from spent nuclear fuel and use these results to outline possible applications of antineutrino detectors in monitoring and managing nuclear-waste repositories. We show that, in a specific diversion scenario at a dry-cask storage facility, the installation of an antineutrino detector could allow oversight agencies to remotely detect a leakage or theft of the stored nuclear waste. Further study is needed to assess the applicability of the method to other diversion scenarios. At long-term geological repositories, a significant antineutrino flux is expected, but detecting realistic anomalies such as a leakage of a small amount of radioactive material would require advanced detector technologies with angular sensitivity. Such detectors could also help in the decommissioning of nuclear installations like the Hanford site, where they would allow for the localization of nuclear material and for the characterization of spills.

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