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Present and future potential of krypton-85 for the detection of clandestine reprocessing plants for treaty verification

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ABSTRACT

Burnup calculations are applied to determine the amount of krypton-85 that is produced during the irradiation of nuclear fuel. Since krypton-85 is most likely released into the atmosphere during reprocessing to separate plutonium, atmospheric transport modeling is used to calculate the worldwide distribution of krypton-85 concentrations stemming from emissions from declared reprocessing plants. The results are the basis for scenarios in which emissions from clandestine reprocessing facilities have to be detected against various background levels. It is concluded that today's background imposes heavily on the ability to detect small and medium plutonium separation rates; only high separation rates of 1 SQ per week and higher have a chance to be detected with feasible outlay. A fixed network of monitoring stations seems too costly; instead the high number of samples that are required rather calls for mobile sampling procedures, where air samples are collected at random locations over the world and are analyzed in regional laboratories for their krypton-85 concentration. Further, it is argued that krypton-85 emissions from declared reprocessing activities have to be significantly lowered to enable a worldwide verification of the absence of even smaller clandestine reprocessing. For each scenario the number of samples that have to be taken for probable detection is calculated.

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1. Motivation

One path to a nuclear weapon is to reprocess spent nuclear fuel and separate the contained plutonium from it. For this reason, the ability to detect clandestine nuclear reprocessing plants is of great relevance for nuclear arms control and nonproliferation. In past studies, krypton-85 has been identified as the most suitable environmental tracer to detect and quantify such nuclear reprocessing activities (von Hippel et al., 1986; Kalinowski et al., 2004). During the Cold War, the U.S. government has used this technique to estimate Soviet plutonium production (von Hippel et al., 1986; Goodman, 2007), and other governments may have monitored krypton emissions as well. In this paper, the prospects of using krypton-85 emissions to detect clandestine reprocessing plants are assessed based on both present technology and possible future developments while taking into account the existing atmospheric background resulting from reprocessing activities over the past 70 years. The importance of gaining confidence in the absence of undeclared reprocessing facilities may further increase with a Fissile Material Cutoff Treaty (FMCT) or other next-generation arms-control treaties.

Undeclared reprocessing facilities could be very challenging to detect. The only technical precursor needed for reprocessing is nuclear spent fuel. While large-scale reprocessing plants take years of planning, building, and testing to meet modern regulatory standards, experts have noted that common industrial facilities (e.g. winery, dairy, oil refinery) could be remodeled to function as "quick and simple" reprocessing plants (Ferguson, 1977). It may be possible to construct such a facility within one year.

A small reprocessing plant with a capacity to process 50 tons of heavy metal (tHM) per year could separate enough plutonium-239 for a single bomb in about a week. The IAEA defines a Significant Quantity (SQ) of nuclear material as "the approximate quantity of nuclear material in respect of which […] the possibility of manufacturing a nuclear explosive device cannot be excluded." For plutonium, this threshold is set at 8 kg for a first generation nuclear device including losses during manufacturing.

Krypton-85 is radioactive with a half-life of 10.76 years and decays by beta- and gamma-decay. Substantial amounts of this isotope have been released into the atmosphere since large-scale

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reprocessing activities started in late 1944. Since then, about 30 known reprocessing plants have released krypton-85 over various time periods and at different scales (Ahlswede et al., 2009). Negligible amounts are also released from nuclear explosions, nuclear power plants, and medical isotope production facilities (Ross, 2010). As krypton-85 is only removed from the atmosphere by its radioactive decay, a background has built up in the atmosphere.

This paper analyzes the technical feasibility of a global monitoring system to detect clandestine reprocessing plants based on their krypton-85 emissions. The number of air samples that need to be taken at random locations over the Earth's landmass for timely detection of plutonium production is calculated. The presented scenarios do not take into account that in some cases additional evidence, e.g. information provided by the intelligence community, may trigger a closer investigation of a certain area. Instead, only the environmental tracer krypton-85 is considered for an approach that does not rely on outside information.

In the following, the pathway of krypton-85 from its creation in nuclear fuel to its release from a reprocessing plant to its atmospheric detection is summarized.

2. Basics of monitoring reprocessing activities with krypton-85

Krypton-85 is produced in nuclear fuel during its irradiation in the reactor. While plutonium-239 is produced by neutron capture in uranium-238, krypton-85 is a fission fragment that is produced when materials such as uranium-235 fission. The exact amount of krypton-85 per amount of plutonium produced depends on the burnup and the type of the spent fuel. For this analysis, the buildup of transuranics and fission products (including krypton-85) during in-core fuel irradiation has been determined with infinite-lattice burnup calculations using MCODE (Xu et al., 2002), which is a linkage code combining the Monte Carlo particle transport code MCNP (Goorley et al., 2012) and the point-depletion code ORIGEN2 (Ludwig, 2002). Krypton-85 inventories have been corrected to account for decay while the discharged fuel is in storage. Fig. 1 shows the activity of krypton-85 per kg of produced plutonium versus burnup. Spent fuel from power reactors has a higher burnup and contains more heat-generating fission products. Thus, spent power-reactor fuel is usually stored and cooled for several years before being reprocessed to allow the short-lived products to decay.

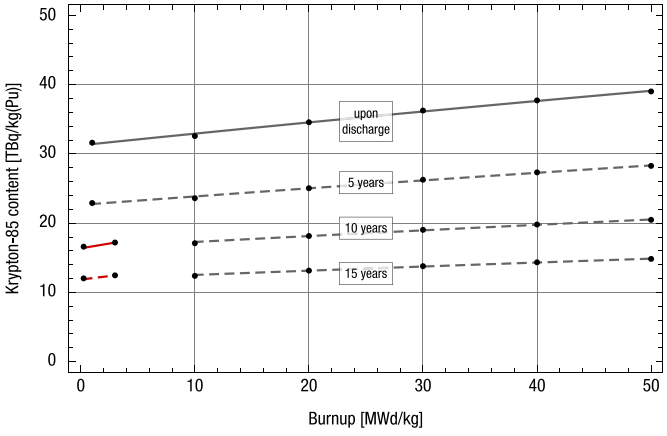

Fig. 1. Krypton-85 activity per produced kg of plutonium in spent fuel over fuel burnup. The gray lines represent power reactor fuel, upon discharge and various ages before opened for reprocessing. The red lines are for a plutonium production reactor (heavy water) and the dashed red line for 5-year-old spent fuel. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fuel from plutonium production reactors has a lower burnup, and reprocessing can therefore start much sooner.

Spent fuel rods are typically stored in spent fuel pools or in dry casks before being reprocessed. In addition to cooling requirements, backlogs of spent fuel can be another reason for extended storage periods. For example, in the reprocessing plant in La Hague, France, the amount of spent LEU fuel awaiting reprocessing at the end of 2010 was 12,000 tons; the average amount of fuel unloaded and the effective reprocessing rate are about 1000 tons per year (IPFM, 2015). If the oldest fuel is reprocessed first, an average storage period of 12 years can be assumed between discharge from the reactor and reprocessing at La Hague. Thus, with the krypton-85 half-life of 10.76 years, a significant part of the krypton-85 may already have decayed once spent fuel reaches the reprocessing stage.

Plutonium can be separated from spent nuclear fuel by the PUREX (Plutonium Uranium Extraction) process, which is the predominant method for reprocessing (Feiveson et al., 2014). The spent fuel rods are chopped up into small pieces and dissolved in nitric acid. The plutonium and uranium are extracted from the nitric acid solution in a light organic solvent and separated with centrifugal extractors. In this process, krypton-85 is released from the spent fuel and enters the off-gas stream of the facility. Krypton retention is technically challenging and expensive; as a noble gas it cannot be filtered with standard absorbent techniques as other particulates. The technique has therefore not been employed in commercial-scale facilities. At some point, krypton is released through the stack into the atmosphere and, once it is airborne, it is dispersed by the prevailing winds. Due to its noble-gas characteristics it is not subject to chemical loss processes during atmospheric transport; radioactive decay is the only loss process.

Whether it can be detected at a monitoring station downwind, and attributed to the releasing facility, depends primarily on the grade of dilution in the atmosphere and on the ability to discriminate the signal against normal fluctuations in the background. The determination of the krypton-85 concentration in air is not a straightforward task. The atmosphere contains about 1.14 ppm by volume of krypton. With current procedures, air samples of about 10 m^3 are typically taken over the course of one week (Bollhöfer et al., 2014); a common minimum air-sample size of 200 l has also been reported (Ross, 2010). After removal of water vapor and CO_2, the dry air sample is pumped through activated charcoal at liquid nitrogen temperature to allow the krypton (not only krypton-85) to be adsorbed onto the charcoal. This is done at a pressure below 500 hPa to prevent condensation of O_2 and N_2. The krypton is released from the adsorbent by heating it up to 300 °C, and pumped into a gas bottle (Bollhöfer et al., 2014).

Once separated, the krypton gas sample can be analyzed for its krypton-85 concentration. Current state-of-the-art detectors use proportional counters to measure the beta-decay activity of the sample. By also chromatographically determining the volume of the krypton sample and using the volume concentration of krypton in air, the krypton-85 activity concentration can be determined. The lower detection limit for such measurements is usually between 1 and 10 mBq/m^3 in the atmosphere. Typical for such sampling and detection methods are 1% statistical uncertainty and 3% systematic uncertainty (Bollhöfer et al., 2014).

Next-generation detectors will be based on an Atom Trap Trace Analysis (ATTA) (Yang et al., 2013; Kohler et al., 2014). This method uses a laser-based, magneto-optical trap. The frequency of the laser can be fine-tuned to the atomic transition frequency to isolate the desired isotopes. A two-dimensional magneto-optical trap cools and traps krypton atoms while providing isotope selectivity. The resulting stream of atoms is pumped into a three-dimensional magneto-optical trap, where the atoms are counted via their

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