Remote detection of undeclared reprocessing

Michael Schoepner
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Michael Schoepfner
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About the IPFM

The International Panel on Fissile Materials (IPFM) was founded in January 2006 and is an independent group of arms control and nonproliferation experts from both nuclear-weapon and non-nuclear-weapon states.

The mission of the IPFM is to analyze the technical basis for practical and achievable policy initiatives to secure, consolidate, and reduce stockpiles of highly enriched uranium and plutonium. These fissile materials are the key ingredients in nuclear weapons, and their control is critical to achieving nuclear disarmament, to halting the proliferation of nuclear weapons, and to ensuring that terrorists do not acquire nuclear weapons.

Both military and civilian stocks of fissile materials have to be addressed. The nuclear-weapon states still have enough fissile materials in their weapon stockpiles for tens of thousands of nuclear weapons. On the civilian side, enough plutonium has been separated to make a similarly large number of weapons. Highly enriched uranium is still used in civilian reactor fuel in many locations. This material could be used to make Hiroshima-type bombs, a design well within the potential capabilities of terrorist groups.

The Panel has been co-chaired since 2015 by Alexander Glaser and Zia Mian of Princeton University and Tatsujiro Suzuki of Nagasaki University, Japan. Previously, it was co-chaired by Jose Goldemberg of the University of Sao Paolo, Brazil (2006–2007), R. Rajaraman of Jawaharlal Nehru University, New Delhi, India (2007–2014), and Frank von Hippel of Princeton University (2006–2014).

Its members include nuclear experts from 15 countries: Brazil, Canada, China, France, Germany, India, Iran, Japan, Norway, Pakistan, Russia, South Africa, Sweden, the United Kingdom, and the United States. This group of countries includes seven nuclear-weapon states and eight non-nuclear-weapon states.

IPFM research and reports are shared with international organizations, national governments, and nongovernmental groups. It has full panel meetings once a year in capitals around the world in addition to specialist workshops. These meetings and workshops are often in conjunction with international conferences at which IPFM panels and experts make presentations.

Princeton University’s Program on Science and Global Security provides administrative and research support for the IPFM.

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Overview

All of today’s nine nuclear-weapon states have produced plutonium for use in their nuclear arsenals. Efforts to remotely detect plutonium production and separation date back to the early years of the Cold War when the United States was collecting intelligence about the Soviet nuclear-weapon program. Detecting clandestine plutonium production and separation is important today for verifying compliance of non-nuclear-weapon states with the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) and, in the future, will be central to verifying compliance of nuclear-weapon states with a possible Fissile Material Cutoff Treaty (FMCT) banning production of plutonium and other fissile materials for use in nuclear weapons.

This report assesses how krypton-85, a non-reactive radioactive gas with a half-life of 10.8 years, could be used as an airborne tracer for monitoring the covert reprocessing of spent nuclear fuel to separate out the plutonium produced in it while the fuel was in a reactor. Detecting krypton-85 from plutonium separation should be possible since significant amounts of the gas are likely to be released into the atmosphere during reprocessing and because it is hard to capture krypton-85 efficiently. More than seven decades of reprocessing in a handful of countries around the world have led to a high background concentration of krypton-85 in the atmosphere, however, limiting the utility of krypton-85 monitoring for detecting at a distance small-scale clandestine reprocessing.

Two scenarios for clandestine reprocessing are considered in this report: undeclared reprocessing in a known plant and a clandestine reprocessing plant at an unknown location. Known facilities can best be monitored with safeguards at the reprocessing plant, such as the on-site measures, including inspections, used by the International Atomic Energy Agency (IAEA). An additional measure would be remote detection of krypton-85 in air samples. Due to strong dilution on environmental pathways through air, water and soil, and the resulting uncertainties, wide-area environmental sampling methods are generally unsuitable to monitor accurately plutonium separation rates at a nearby plant. However, such methods can help verify the shutdown status of a reprocessing plant.

Case studies of the reprocessing plants at Dimona in Israel and at Yongbyon in North Korea show that for either of these sites two or three down-wind krypton-85 monitoring stations located in the most frequent wind directions would be sufficient to monitor and verify a shutdown of reprocessing activities. Monitoring stations within the country and close to the reprocessing plant in question would greatly increase the monitoring efficiency.
A small clandestine reprocessing plant at an unknown location is challenging to detect since the limited krypton-85 releases are quickly masked by the current high atmospheric background. If such a plant is to be detected by fixed monitoring stations, it would require a high-density network of stations. An alternative approach analyzed in this report is to take air samples at random locations for later analysis of the krypton-85 content.

It appears large-scale random air sampling could detect significant clandestine reprocessing activities within the time it takes to separate a quantity of plutonium considered sufficient by the IAEA to build a first generation nuclear weapon. This approach can be challenging, however, for large target areas and if a very high probability of successful detection is set as a verification goal. About 50 air samples per day at random locations would be necessary to reliably monitor an area of 10 million square kilometers ($\text{km}^2$) for the absence of reprocessing activities with 90 percent certainty – for comparison, the United States and China each have an area of almost 10 million $\text{km}^2$ and Russia is the only country with an area larger than this.

Small reprocessing plants – those that separate only about one or two nuclear weapons worth of plutonium over a year – would be detectable with only about 50 percent probability because their krypton-85 footprint vanishes quickly into the current background. About 150 air samples at random locations are required for reliable detection (90 percent certainty) of such activities. The numbers of required air samples could be reduced by various methods and search strategies, such as reducing the total search area by excluding areas without infrastructure.

Improving the effectiveness of remote monitoring of known reprocessing plants and of the detection of clandestine reprocessing plants could be achieved by reducing the krypton-85 background in the atmosphere by ending civilian reprocessing. This would allow the high background level to stabilize and then decline over the next few decades as the krypton-85 decayed away. The number of samples required to detect covert reprocessing activities over a given region would be dramatically reduced. Thereafter, the detection of clandestine reprocessing would continue to become easier.

A future Fissile Material Cutoff Treaty which ended reprocessing not only for nuclear weapon purposes but also for civilian use would have a greater chance of remotely detecting possible clandestine reprocessing plants.
Plutonium and reprocessing

The world’s first nuclear weapon test was carried out by the United States on 16 July 1945. It was fueled by plutonium that had been produced and separated only a few months earlier at the Hanford Site on the Columbia River in Washington state. About three weeks after the test, a similar plutonium bomb was used to destroy Nagasaki. Today, every nuclear-weapon state is believed to use plutonium in its nuclear arsenal.

At least two of the four nuclear weapon states that are not parties to the Nuclear Non-Proliferation Treaty (NPT) – India and North Korea† – chose the plutonium pathway for the fissile material of their first nuclear weapons.‡ Historically, plutonium has been easier to produce than highly enriched uranium (HEU). The production of HEU by gaseous diffusion, the method used first by the five NPT nuclear-weapon states, was very costly, but it made possible the gun-type design, a design so simple that the first-generation device used to bomb Hiroshima was not even tested.† Producing plutonium in a nuclear reactor and separating it through reprocessing is less costly, and plutonium has a lower critical mass than HEU. However, the implosion design required for plutonium-based weapons is more difficult to accomplish than the gun-type design.

Figure 1 gives an overview of the production process of plutonium. Plutonium is created by neutron irradiation of uranium. By absorbing a neutron without fission, uranium-238 becomes uranium-239, which decays via the short-lived isotope neptunium-239 to plutonium-239. With ongoing neutron irradiation, the newly created plutonium-239 can absorb more neutrons to become plutonium-240 (or -241 or -242). Thus, the plutonium that is created during the irradiation of uranium can have different percentages of plutonium-239. The irradiation process takes place inside the uranium-bearing fuel rods in a nuclear reactor.

Plutonium can be separated from spent nuclear fuel with the PUREX (plutonium-uranium extraction) process. The spent-fuel rods are chopped into small pieces and dissolved in nitric acid. The plutonium and uranium are extracted from the nitric-acid solution in a light organic solvent, which is then separated with centrifugal extractors. Most reprocessing today is based on the PUREX process.

Another reprocessing technique being pursued in connection with breeder reactor programs is pyroprocessing, in which metal fuel is dissolved in molten salt and the plutonium and uranium are separated out electrochemically. The choice of reprocessing method has little effect on the release of fission products from a reprocessing plant.
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, the IAEA has set this threshold at 8 kilograms (kg) for a first-generation nuclear device, including losses during manufacturing. The irradiated uranium in spent light-water power reactor fuel contains about 1 percent plutonium. A small reprocessing plant with a capacity to process 50 tons of heavy metal (tHM) of power reactor fuel per year therefore could separate enough reactor-grade plutonium for one bomb in about a week.

Reprocessing programs

Production of plutonium for the first nuclear weapons started in 1944 in the B Reactor at the Hanford Site; reprocessing commenced shortly after at the same site. Today, China, France, India, Israel, Japan, North Korea, Pakistan, Russia, the United Kingdom and the United States have reprocessing programs (see Table 1).

All nuclear-weapon states had military reprocessing programs to build their weapon arsenals, but all NPT nuclear-weapon states have ceased to separate plutonium for that purpose. Russia stopped producing weapon-grade plutonium in the 2000s but has continued to separate plutonium from power reactor fuel for its breeder program since 1977. Production of military plutonium continues in India, Pakistan, North Korea and, it is believed, Israel.

Today, France, Russia, the United Kingdom, Japan, and India operate civilian reprocessing facilities. The United Kingdom and France launched civilian reprocessing in the 1960s and 1970s. The United States operated a civilian reprocessing plant only briefly, from 1966 to 1972, and today maintains the capability only for special operations. Globally, civilian reprocessing has not proven to be economical and has faced numerous technical problems. Germany has abandoned its reprocessing program, and the United Kingdom is in the process of doing so. France and Japan still have full civilian reprocessing programs. Russia and India are reprocessing for civilian purposes on much smaller scales, while China is operating a pilot civilian reprocessing facility.
<table>
<thead>
<tr>
<th>Country/Facility</th>
<th>Type</th>
<th>Status</th>
<th>Safeguards</th>
<th>Capacity (tHM/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CHINA</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Jiuquan</td>
<td>Civilian</td>
<td>Operating</td>
<td>no</td>
<td>50</td>
</tr>
<tr>
<td>FRANCE</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>La Hague UP2</td>
<td>Civilian</td>
<td>Operating</td>
<td>yes</td>
<td>1,000</td>
</tr>
<tr>
<td>La Hague UP3</td>
<td>Civilian</td>
<td>Operating</td>
<td>yes</td>
<td>1,000</td>
</tr>
<tr>
<td>INDIA</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Trombay</td>
<td>Military</td>
<td>Operating</td>
<td>no</td>
<td>50</td>
</tr>
<tr>
<td>Tarapur-I</td>
<td>Dual</td>
<td>Operating</td>
<td>no</td>
<td>100</td>
</tr>
<tr>
<td>Tarapur-II</td>
<td>Dual</td>
<td>Operating</td>
<td>no</td>
<td>100</td>
</tr>
<tr>
<td>Kalpakkam</td>
<td>Dual</td>
<td>Operating</td>
<td>no</td>
<td>100</td>
</tr>
<tr>
<td>ISRAEL</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dimona</td>
<td>Military</td>
<td>Operating</td>
<td>no</td>
<td>40 –100</td>
</tr>
<tr>
<td>JAPAN</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rokkasho</td>
<td>Civilian</td>
<td>Starting up</td>
<td>yes</td>
<td>800</td>
</tr>
<tr>
<td>Tokai</td>
<td>Civilian</td>
<td>Reprocessing shut down</td>
<td>yes</td>
<td>200</td>
</tr>
<tr>
<td>NORTH KOREA</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yongbyon</td>
<td>Military</td>
<td>Operating</td>
<td>no</td>
<td>100 –150</td>
</tr>
<tr>
<td>PAKISTAN</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nilore</td>
<td>Military</td>
<td>Operating</td>
<td>no</td>
<td>20 –40</td>
</tr>
<tr>
<td>Chashma</td>
<td>Military</td>
<td>Under construction</td>
<td>no</td>
<td>50 –100</td>
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<tr>
<td>RUSSIA</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RT-1</td>
<td>Dual</td>
<td>Operating</td>
<td>no</td>
<td>400</td>
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<tr>
<td>EDC</td>
<td>Civilian</td>
<td>Starting up</td>
<td>no</td>
<td>250</td>
</tr>
<tr>
<td>UNITED KINGDOM</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>B205</td>
<td>Civilian</td>
<td>To be shut down in 2018</td>
<td>yes</td>
<td>1,500</td>
</tr>
<tr>
<td>THORP</td>
<td>Civilian</td>
<td>To be shut down in 2020</td>
<td>yes</td>
<td>1,200</td>
</tr>
<tr>
<td>UNITED STATES</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>H Canyon, SRP</td>
<td>Converted</td>
<td>Special operations</td>
<td>no</td>
<td>15</td>
</tr>
</tbody>
</table>

Table 1. **Reprocessing plants worldwide.** The list includes facilities under construction, preparing for operations, or temporarily shut down. Capacity is measured in tons of heavy metal (primarily uranium) processed per year.

Currently, about 10,500 tHM of spent fuel are discharged annually, with most of it coming from nuclear power plants and only a small fraction from research reactors. About 2,000 tHM/yr of the total generated spent fuel is being reprocessed. Due to the substantial amounts of plutonium in spent fuel and the resulting proliferation risks, the IAEA monitors spent fuel in storage and reprocessing in NPT non-nuclear-weapon states and also, along with Euratom, civilian reprocessing in France and the United Kingdom. It uses remote surveillance systems and inspectors to monitor the transfer, storage, and sealing of fuel.\(^{a}\)
Proliferation risks from known reprocessing facilities

The NPT provides a safeguards regime to monitor the declared facilities, materials, and activities of member states. The Model Additional Protocol introduces measures to increase the chances of detecting undeclared materials and activities. These measures include the possibility, if approved by the IAEA Board of Governors, of environmental sampling outside nuclear facilities. This would allow soil, water, and air samples to be taken over wide areas and analyzed for radionuclide tracers. The detection of clandestine fissile-material production remains a challenge, however.

Most reprocessing plants are in nuclear-weapon states and have not been designed and constructed for safeguards. Only Japan, France, and the United Kingdom have their reprocessing plants under safeguards – Euratom safeguards in the case of the latter two (see Table 1). Reprocessing facilities in the other nuclear-weapon states are not under international safeguards and could be used for the separation of plutonium for weapons. In the absence of international monitoring, a country could divert plutonium in a declared facility from civilian to military purposes.

Reprocessing for weapons purposes is expected to be prohibited under a future Fissile Material Cutoff Treaty (FMCT). The United Nations Conference on Disarmament in Geneva has been seeking to begin talks on this treaty since 1995. Proposed drafts of a possible FMCT focus on obligations with regard to verification rather than detailing procedures.

Some countries may insist civilian reprocessing, which is neither economic nor necessary, be permitted under an FMCT. As concluded in this report, this would hinder the detection of clandestine reprocessing activities worldwide. Thus, it would be beneficial to ban reprocessing for all purposes. Furthermore, a ban on HEU for all purposes would also be easier to verify than a ban on HEU production only for weapon purposes.

When a state commits not to separate plutonium for weapons under the NPT or a future Fissile Material Cutoff Treaty (FMCT), the international community may wish to verify the accuracy of civilian material declarations or the absence of undeclared reprocessing or diversion in known facilities.

Because spent fuel in NPT non-nuclear-weapon states is placed under safeguards, a country wishing to secretly separate plutonium would need to divert spent fuel from safeguards or operate a clandestine reactor to irradiate uranium. A country also could try to take advantage of the approximately one percent measurement uncertainties to divert a significant amount of plutonium from the throughput of on the order of 10 tons a year in a large reprocessing plant.

As seen from these different pathways to reprocessing, it is desirable to have a range of options to monitor the reprocessing status in known facilities or to verify a shutdown.
Proliferation risks from clandestine reprocessing facilities

Unlike reactors, which have distinct visual, thermal, and logistical signatures during construction and operation, reprocessing plants do not have characteristic signatures to distinguish one from other industrial facilities. Typically located at a reprocessing site are indoor pools to store spent fuel, big concrete reprocessing “canyons,” tall stacks for emissions, and storage tanks for radioactive waste. Also, an existing non-nuclear industrial facility could be improvised into a “quick and simple” reprocessing plant and operated without a distinctive thermal signature. Industries that process wine, dairy products, or oil could provide necessary stainless-steel tanks and basic equipment to handle nuclear waste (see Figure 2). Such a facility could, in theory, be built within four to six months. Radiation safety in such an improvised facility would not be up to the standards of official reprocessing plants and could result in high occupational radiation exposures for its workers. According to one design study, however, it could be used to process one light-water reactor fuel assembly containing about 500 kg of uranium per day and to produce up to one SQ of plutonium per week (up to 1.1 kg of plutonium per day).

A newly built but undeclared reprocessing plant with conventional layout might be detected during construction with satellite imagery due to its thick walls for radiation shielding. An improvised facility would have no visible signatures to distinguish it from other industrial buildings in satellite imagery.

Of course, reprocessing in a clandestine facility requires a source of irradiated uranium. NPT non-nuclear-weapon states could divert some of their domestically irradiated uranium or import it from foreign sources. Alternatively, the country could also operate a clandestine reactor to produce unsafeguarded irradiated uranium. Countries that are not parties to the NPT but have a nuclear reactor have easier access to unsafeguarded spent fuel.

In conclusion, there is a gap in the detection capability and a need for remote detection of clandestine reprocessing plants.
Figure 2. A quick and simple reprocessing plant that could be hosted in a remodeled industrial plant. Such a facility would show no visible signatures to distinguish it from other industrial facilities and could separate one SQ of plutonium per week. Source: Unmaking the Bomb: A Fissile Material Approach to Nuclear Disarmament and Nonproliferation.
Remote detection of reprocessing activities

The production of plutonium requires the irradiation of uranium in a reactor, and its separation requires a reprocessing plant. Nuclear reactors are usually identifiable with satellite imagery in the visible and thermal spectrum. However, as noted in the previous section, reprocessing plants could be indistinguishable from industrial facilities. This makes remote detection especially challenging. Thus, even for a country with a declared nuclear program, it is not possible to exclude the existence of a clandestine reprocessing program. If a country’s reactors and its spent fuel are declared and safeguarded, it could still undertake clandestine reprocessing by building and operating an undeclared reactor or through clandestine imports of spent fuel.

Environmental sampling is a common tool to detect nuclear activities. Toward the end of World War II, the United States sent planes over Germany to take air samples and analyze them for radioactive xenon, a fission product, as an indicator of nuclear activities. Early nuclear monitoring efforts also were looking for two other fission products – iodine-131, which has a half-life of eight days, and krypton-85, which has a half-life of 10.8 years. However, iodine can be retained by a filter in a facility’s off-gas stream and even if not retained, is prone to precipitation and being washed out of the atmosphere. Iodine isotopes still enter the biosphere through plants and can be absorbed by livestock. Accumulated iodine in livestock’s thyroids can become detectable at some point, but this is not a timely and reliable detection method.

From 1945 to 1958, an Anglo-American intelligence collaboration sought to confirm the existence of a Soviet nuclear program and then quantitatively assess Soviet plutonium production and stockpiles. This effort largely focused on atmospheric sampling. Its activities included benchmarking trials in which large quantities of radioisotopes were released from the U.S. Hanford plant in 1949 (Operation Green Run for airborne radioactive products, particularly iodine-131, and Operation Bluenose, which specifically targeted krypton-85); interviews with German scientists returned from the Soviet Union to learn about Soviet nuclear facilities (Operation Dragon Return); and overflights over the Soviet Union in cooperation with British intelligence (Operation Nomination, later known as Music Programme).

Operation Nomination and Music Programme included that Canberra B6 airplanes (Figure 3) based in Australia were specially equipped for long-range missions and sent on secret sampling missions over the Soviet Union usually at altitudes of more than 6 km. The planes had a compressor driven by engine air that was built into the bomb bay and filled up special pressure bottles with air samples. Upon return, the bottles were shipped back to the United Kingdom to be analyzed for their krypton-85 content.
It was soon realized that krypton-85 was the most suitable environmental tracer for reprocessing activities. As a noble gas, it is difficult to retain in the facility, is not subject to precipitation or chemical reactions in the atmosphere and can be found in air samples from higher altitudes where spy planes cannot be shot down. Vertical transport of industrial emissions up to these altitudes can take about a week. Therefore, former releases of krypton-85 are already well mixed at that height and are not influenced by recent emissions that would dominate the total concentration at lower altitudes. Since only manmade activities produce significant amounts of krypton-85 and nuclear activities were started in 1944, only low background concentrations were present in the atmosphere during these missions.

Music Programme continued until 1958 when the United Kingdom and the United States concluded that the Soviet Union had reached nuclear sufficiency – that is, a stockpile of weapons large enough to carry out virtually any war plan. After Music Programme was formally concluded, similar airborne sampling missions over the Soviet Union were conducted, first by the Royal Air Force and later by U.S. Air Force U-2 airplanes well into the 1960s. During that same time period, a network of stations was established to operate and maintain sampling units deployed at consulates and other U.S. facilities abroad.

During the 1980s, independent U.S. scientists derived total past Soviet plutonium production from published measurements of the average atmospheric krypton-85 concentration in the Northern Hemisphere. At that time, the total krypton-85 concentration in the atmosphere was largely due to U.S. and Soviet plutonium separation efforts and only to a small degree due to reprocessing in other states. By subtracting the contribution of the known U.S. plutonium separation and correcting for decay, independent scientists were able to estimate the total Soviet plutonium stockpile from the history of the average atmospheric krypton-85 concentration.
In the mid- to late 1990s, Russian intelligence cooperated with U.S. counterparts to bring detection equipment provided by the CIA into the Russian embassy in Pyongyang, North Korea. The Russians received training and equipment to support efforts to determine whether the reprocessing plant at the nuclear facility in Yongbyon was separating plutonium. It has been assumed that the deployed sensing equipment was measuring krypton-85 concentrations.  

In the late 1990s, the IAEA published a study called STR-321 on the detectability of undeclared nuclear activities through wide-area environmental sampling (WAES). WAES can include sampling of air, soil and water. The report includes estimates of source terms from several types of fissile-material production facilities and analyzes the feasibility of detection. The results were supported by two trials around reprocessing plants to examine the application and logistics of environmental air sampling in a WAES verification framework. With regard to undeclared reprocessing, the study considered krypton-85, strontium-90, iodine-129, cesium-134, and cesium-137 as atmospheric tracers and concluded that, in most cases, atmospheric sampling is more likely to yield successful detection compared to soil and water sampling. Concentrations in soil samples are usually too low to be detectable. Detectability in water samples comes with large uncertainties depending on the geographical location, the release pathways, and operation of the reprocessing plant.

The size of a network of monitoring stations for krypton-85 would depend on the region to be covered and target detection probabilities for given reprocessing rates. As will be discussed below, however, it became clear that, for larger monitoring areas, such a network would be prohibitively costly.

More recent studies have examined the feasibility of standoff monitoring of known reprocessing plants. Fixed monitoring stations at a distance of up to a few tens of kilometers in the most common wind directions from the plant would detect releases of krypton-85 associated with plutonium separation. Since emission patterns and wind direction give rise to large uncertainties, this method is not suitable for monitoring levels of plutonium production against declared values, but rather the monitoring of a shut-down reprocessing plant.

The detectability of krypton-85 for this purpose has been drastically reduced since its first use, however. Today, the signal of krypton-85 plumes from a reprocessing plant of interest must be distinguished from the high atmospheric background that has accumulated since the start of industrial reprocessing in 1944 and the krypton-85 noise due to current emissions of large industrial reprocessing plants.

**Environmental releases from reprocessing**

The Model Additional Protocol of 1997 in principle allows the application of WAES for NPT safeguards.

Spent fuel contains fission products and actinides resulting from neutron absorption by the uranium fuel. The opening of spent fuel rods for reprocessing and the subsequent
chemical separation allow some gaseous and volatile fission products to enter the waste streams of the facility. Appropriate filters will retain most of the airborne particulates and gases. However, depending on the chemical element, a certain fraction can escape into the environment. These can potentially be used as environmental tracers to indicate the presence of reprocessing activities. In the absence of a background, radioactive isotopes can be detected more easily due to their radioactive signatures.

The suitability of a radionuclide as an environmental tracer depends on its presence in the spent fuel, the release fraction from the reprocessing plant, the atmospheric transport mechanisms, the background concentration in the atmosphere, and the capabilities for detecting the radionuclide. Selected radionuclides that can be expected to be released during routine operation of a reprocessing plant are listed in Table 2.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Total inventory</th>
<th>Atmospheric release during routine operation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>grams</td>
<td>Becquerel (Bq)</td>
</tr>
<tr>
<td>Krypton-85</td>
<td>7.50</td>
<td>1.1E+14</td>
</tr>
<tr>
<td>Tritium</td>
<td>0.01</td>
<td>4.3E+12</td>
</tr>
<tr>
<td>Iodine-129</td>
<td>2.00</td>
<td>1.3E+07</td>
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<tr>
<td>Plutonium-239</td>
<td>7.600</td>
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<td>400</td>
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<td>Antimony-125</td>
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<td>16</td>
<td>6.9E+13</td>
</tr>
<tr>
<td>Ruthenium-106</td>
<td>14</td>
<td>1.8E+15</td>
</tr>
<tr>
<td>Strontium-90</td>
<td>180</td>
<td>8.9E+14</td>
</tr>
<tr>
<td>Americium-241</td>
<td>1.10</td>
<td>1.3E+11</td>
</tr>
</tbody>
</table>

Table 2. Radionuclide inventory in spent fuel and atmospheric releases during routine operation of a reprocessing plant. The quantities represent the amount that is in irradiated fuel containing 1 SQ of weapon-grade plutonium (8 kg). The values are based on natural uranium irradiated in a Magnox reactor with a burnup of 0.55 MWd/kg and a cooling period of one year; other reactors and irradiation conditions can produce different values. Krypton-85 data for other fuel types and cooling periods are shown in Figure 4. 
Source: IAEA, IAEA Use of Wide Area Environmental Sampling, 1999.

During routine operation, non-noble gases in the off-gas stream are mostly captured in filter systems. Thus, only a fraction of the original inventory is released into the atmosphere, allowing for detection only short ranges from the facility. Additionally, after release, radionuclides other than noble gases are subject to dry and wet deposition adding further uncertainty to their detectability in atmospheric air. Gaseous tritium in atmospheric air can be detected at distances of up to about 10 km from facilities that reprocess more than 15 SQ annually. Soil sampling for the detection of radioactive tracers was considered in STR-321, but concentrations and probabilities were found to be too low.
Radioactive tracers also can find their way into the environment through aquatic releases. About 70 percent of the tritium from spent fuel is released as part of the liquid waste from a reprocessing plant.\textsuperscript{22} The tritium from spent fuel usually enters the aquatic waste stream in the form of tritiated water. Other nuclear facilities that release tritium into the environment are light-water reactors and tritium production reactors.

The release of waste into the aquatic environment can be handled very differently at different reprocessing plants. Sampling river or lake water can be used to find evidence of a suspect nuclear program in a certain region.\textsuperscript{27} For example, the La Hague reprocessing plant releases about 30 grams (g) per year – 10,000 terabecquerels (TBq) per year – of tritium into the sea while the Sellafield plant releases about 8 g/year (3,000 TBq/year).\textsuperscript{26} Ordinary water usually carries a tritium concentration of 0.1–0.9 Bq/liter.\textsuperscript{29} Seawater around the Cotentin Peninsula, where La Hague is located, has been found to contain a tritium concentration of 3–10 Bq/liter. Samples taken from the Rhone River downstream from the Marcoule Nuclear Site in southern France showed concentrations of 1–10 Bq/liter and even 20–50 Bq/liter in the immediate vicinity of the facility.\textsuperscript{30} Americium-241, strontium-90, and plutonium isotopes also were detected in the Rhone River downstream from Marcoule.\textsuperscript{31}

Radioactive xenon isotopes are not suitable for detecting reprocessing plants. They can be used as an indicator for other nuclear activities such as nuclear tests, nuclear power plants, or the extraction of short-lived fission products for medical uses.\textsuperscript{32} In the case of reprocessing, their half-lives of a few days or less are so short that they will decay before the fuel rod is opened for reprocessing.

Due to its significant fission yield and suitable half-life, krypton-85 is the radionuclide most likely to be detected from routine operation of a reprocessing plant. Accidental gaseous releases during the handling and filtration of off-gases have the potential of much larger releases into the environment resulting in a greater detection range. This could also make radionuclides other than krypton-85 suitable for detection. However, for the discovery of clandestine reprocessing activities, a reliable detection scheme should not rely on accidental releases but aim for detection during standard operation.

The amount of krypton-85 being released depends on the type of fuel, its burnup and the subsequent cooling time. Typical light-water power reactor fuel at low burnup contains at least 32 TBq of krypton-85 per kilogram of plutonium upon discharge. Higher burnups will increase this ratio. Low-burnup heavy-water reactor fuel for production of weapon-grade plutonium contains only about 17 TBq of krypton-85 per kilogram of plutonium.\textsuperscript{33}

Spent fuel from power reactors has a higher burnup and contains more heat-generating fission products. To allow the short-lived products to decay, it is usually stored and cooled for several years before being reprocessed. Fuel from plutonium production reactors has a lower burnup, and reprocessing can therefore start much sooner. More detailed data on how the amount of krypton-85 per kilogram of plutonium varies with burnup and cooling time are shown in Figure 4.
For the following assessment of the detectability of clandestine reprocessing it is assumed that at least 128 TBq of krypton-85 are released for each SQ of plutonium. This represents spent fuel with low burnup from a plutonium production heavy-water reactor after one year of cooling, or typical spent fuel from a power reactor after about 10 years of cooling. It is possible that a proliferator would aim for a somewhat smaller krypton-85 content, but except for waiting for several half-lives to let the krypton-85 decay, there is nothing that would significantly change the detectability assessment. It is also possible that emissions could be higher and lead to better detectability.

The frequency and magnitude of releases depend on the size and operational schedule of the reprocessing plant. Industrial facilities work on fixed schedules and apply off-gas treatment before atmospheric releases. This can lead to delayed releases. For larger facilities, releases of krypton-85 can be approximated as continuous.

A quick and simple reprocessing plant, as described earlier, probably would not employ sophisticated air filter systems. Depending on the schedule of chopping up spent-fuel rods, krypton-85 could be released multiple times per day or only once over the course of several days. However, in the absence of a krypton retention system, at some point all the krypton-85 that comes with the production of plutonium must be released.
Krypton retention

Reprocessing produces large volumes of contaminated air that are usually released into the atmosphere. On-site storage of the total gas volume would require high-pressure tanks, increasing the risk of accidental releases of large quantities over short time periods. Technically, however, krypton-85, as the most suitable tracer for reprocessing, could be removed from the gas before it is released.

In the treatment of off-gas streams from nuclear activities, particulates and chemically active or soluble radioisotopes are removed with filters before release through the stack into the atmosphere. Noble gases such as krypton-85, however, are chemically inert and thus difficult to retain. Known methods for krypton retention are based on either cryogenic methods or physical adsorption in molecular sieves, metal-organic frameworks, or porous organic polymers. These methods cannot distinguish different isotopes and separate all krypton isotopes from the off-gas stream, including stable krypton, which is present naturally in the atmosphere at a level of about 1 part per million (ppm) by volume.

Cryogenic distillation is the most developed krypton separation technology and is also used to separate xenon from air. Experimental applications of cryogenic distillation have been reported at reprocessing plants in the United States and Japan but have not been implemented for routine operations. The separation process is based on different boiling points of gases and can capture between 99 percent and 99.9 percent of krypton. Due to the technically challenging nature of the process, it is not always possible to reach these values. Ozone can accumulate during operation, and its explosive nature at elevated temperatures or fast warming to the boiling point can be safety hazards.

Fluorocarbon absorption and carbon dioxide absorption can be applied at higher temperatures and pressures. The separation of krypton from the off-gas is based on its higher solubility in a column of solvent. The explosive hazards are smaller than for cryogenic distillation, but capital and operational costs are comparable. The process still requires cooling, but the higher temperatures and widely available solvents lower the operating costs. The operation at higher pressures both offsets these savings and increases the chances of accidental releases. These methods have been developed and tested on a laboratory scale with reported krypton separation efficiencies of up to 99 percent but have not been deployed and tested on industrial scales and schedules.

Selective physical adsorption is simpler and less costly to operate compared to the cryogenic and absorption methods. Noble gases such as xenon and krypton are adsorbed in a selective sorbent where the sorbent capacities can be adjusted by either lowering the temperature or increasing the pressure. Once the krypton has been adsorbed, it can be transferred into storage containers by increasing the temperature and pumping. The most common adsorbent, activated charcoal, presents a fire hazard during the heating process, however. Zeolites with suitable adsorption capacities are potential alternatives without the fire hazard. Selective adsorption is used for smaller air volumes in noble-gas detection systems, but it has not yet been applied on the scale that would be required for the off-gas volumes created by a reprocessing facility.
Stable xenon isotopes are present in the reprocessing off-gas at concentrations about 10 times higher than those of krypton. Krypton separation methods also capture xenon, but both compete for the same capture capacities. Therefore, it may be advisable to separate xenon before krypton (see Figure 5).

Thus, the technologies to separate noble gases exist, but operational application may be challenging and comes with risk of accidental releases of large quantities of krypton. In the past, when a large growth in commercial reprocessing was foreseen, requirements for capture were considered. Krypton-85 emissions are not considered a health issue today, however, because the radiological doses to individuals downwind are low. In the future, some governments may introduce national regulations to restrict the emissions. But today, systems to capture krypton are not believed to be deployed in existing facilities, as they are costly and are not required by any regulations.

In summary, a noble-gas retention system at an undeclared reprocessing plant is technically possible. However, since there is not much experience with its large-scale application and no off-the-shelf solution exists, any noble-gas retention system would have prototype status and be prone to accidental releases. Additionally, it could become a bottleneck and possibly slow down reprocessing.

**Atmospheric dispersion**

The filtered off-gas stream, including krypton-85, is released through the stack into the atmosphere. Thereafter, the plume disperses according to wind patterns and turbulence, and the krypton-85 concentration becomes diluted over time.

Immediately after release, the plume is compact and dense. Its vertical and horizontal dispersal depends mainly on the local atmospheric stability and wind direction and speed. Further movement of the plume is subject to the movement of the surrounding air masses whose motions are determined by gravity, pressure gradients, and the Coriolis effect, which control the general circulation of the atmosphere.
Horizontal and vertical transport times can vary greatly; vertical transport times depend strongly on the season, the latitude, and the climate. Typical horizontal and vertical time scales are shown in Figure 6. In the midlatitudes of the troposphere, wind speeds from west to east are typically on the order of 10 meters per second (m/s), resulting in a transport time around the globe of a few weeks. Transport mechanisms from the midlatitudes toward the equator produce north-south wind speed components of only about 1 m/s. Thus, the exchange of air masses between the middle latitudes and the tropics usually happens on a time scale of one to two months. The exchange of air between the hemispheres takes significantly longer with a time scale of about one year.  

Vertical movements of air masses are caused by an imbalance between gravity and vertical pressure gradients. The troposphere stretches from ground level to the tropopause whose height varies from 7 km in polar regions to 17 km in midlatitudes and to 20 km in tropical regions. Basically, all weather is happening within the troposphere. The lowest part of the troposphere is the planetary boundary layer, where friction with the surface influences the motion of the air. The height of the layer varies between a few hundred meters and 2 km, depending on latitude, type of land, and time of day.

Figure 6. Characteristic time scales for horizontal and vertical transport. Top: Expectations of typical horizontal transport times in the troposphere. Middle latitudes often have high-velocity horizontal winds from west to east, while closer to the equator, vertical air movements dominate. This leads to a slow mixing of air masses between the hemispheres. Bottom: Typical time scales for vertical transport. The lowest layer of the troposphere is the planetary boundary layer (PBL), which is between a few hundred meters and 2 km high. Source: Jacob, Introduction to Atmospheric Chemistry, Chapter 4.
Particles that are released on or near ground level can mix through the height of the layer within one or two days depending on turbulent air movement and surface conditions. Mixing into the troposphere can take about one week (see Figure 6).

In the higher part of the troposphere, the atmospheric flows are almost geostrophic along the isobars – that is, perpendicular to pressure gradients. For transport beyond the tropopause – that is, into the stratosphere – the vertical exchange mechanisms are significantly slower; the temperature inversion above the tropopause inhibits vertical movement. Due to the long half-life of krypton-85, however, it is well mixed with air in the troposphere and even the stratosphere.

Detection systems and monitoring efforts

The inertness of krypton-85, which makes it difficult to remove from reprocessing off-gases, also makes it challenging to recover from atmospheric air. Atmospheric air contains not only krypton-85, but other, mostly stable isotopes of krypton. Taking into account all krypton isotopes, the atmosphere contains about 1.14 parts per million (ppm) of krypton by volume.

Current krypton-85 detection systems require a minimum air sample size of about 200 liters. During past monitoring efforts by the German Federal Office for Radiation Protection, air samples of about 10 cubic meters (10,000 liters) were processed to increase the accuracy of the measurement. These air samples usually were taken over the course of one week.

During preprocessing of the air sample, water vapor and CO₂ are removed. The dried air sample is then pumped through a column of activated charcoal at a liquid nitrogen temperature of 77 K and the krypton is adsorbed by the charcoal. A low pressure of 500 hPa (one half an atmosphere pressure) during this process prevents condensation of O₂ and N₂. After the krypton has been isolated in the column of activated charcoal, the temperature is increased to room temperature. At this point, the charcoal releases the krypton, which can be pumped into a gas bottle for storage, transport, and further analysis.

The recovered gas sample can be analyzed in a proportional counter to measure its beta-decay activity and thereby the concentration of krypton-85 in the krypton sample. The volume concentration of krypton-85 in air is determined by the volume concentration of krypton in the sample and the krypton-85 activity. This detection method typically results in 1 percent statistical and 3 percent systematic uncertainty and allows krypton-85 detection at air concentrations as small as 1–10 millibecquerels per cubic meter.

Next-generation detectors are currently under development and have demonstrated improved performance. They are based on an Atom Trap Trace Analysis (ATTA) employing a laser-based, magneto-optical trap that can be used to isolate even single atoms. The trap can separate specific isotopes by fine-tuning the laser frequency to the isotope's
atomic transition frequency. Potentially all krypton-85 atoms in a sample – not just those atoms that decay during the measurement period – can be trapped and counted with this setup. This will allow the use of smaller sample volumes of about 1–2 liters and measurement times of the detector on the order of only a few hours.

**Historic development of the atmospheric krypton-85 background**

Human-caused fission is the only significant source of krypton-85 in the atmosphere and, since krypton is chemically inert, there are no chemical or physical removal processes in the atmosphere. Radioactive decay is the only reduction process. As long as krypton-85 is released, the total inventory in the atmosphere will adjust until an equilibrium between the release and decay rates is reached. As seen in Figure 7, the total atmospheric content of krypton-85 seems to have reached equilibrium at about 2005 with a slight reduction since 2010.

![Figure 7. Global development of annual krypton-85 emissions (bars) and its total content in the atmosphere (line). The global inventory grew because the only loss process for krypton-85 is its radioactive decay with a half-life of 10.8 years. Over the decades, atmospheric krypton-85 has been dispersed and well mixed in the atmosphere. Since about 2005, new emissions and radioactive decay have been in approximate balance and the total content has stabilized. Source: Adapted from Ahlswede, J., Hebel, S., Ross, O., Schoetter, R., and Kalinowski, M. B., “Update and improvement of the global krypton-85 emission inventory,” Journal of Environmental Radioactivity 115/1 (2013): 34 – 42.](image-url)
The German Federal Office for Radiation Protection has been conducting systematic krypton-85 measurement campaigns since 1973. Weekly air samples are collected in several locations in Europe and at selected locations around the world (see Figure 8). A comprehensive 2010 study on the atmospheric krypton-85 distribution used this data to validate estimated source terms and global transport mechanisms.46

Krypton-85 release data from Savannah River Plant in the U.S. state of South Carolina were used in the 1970s to develop models of release rates and to verify simple meteorological models.47 A regional krypton-85 measurement campaign combined with release data from the Savannah River Plant was conducted in the early 1980s to build a data set for an atmospheric tracer experiment for air pollution model development and evaluation.48

Civilian measurement campaigns were also conducted in Russia during the Cold War and were renewed in 2006 to monitor variations in krypton-85 concentrations. Samples were taken up to twice per week over a period of two years to examine the dependence on the distance and sampling intervals for two nuclear power plants.49 Monitoring campaigns by other countries, especially reprocessing states, are assumed to exist as part of their national technical means, e.g. in the U.S., but no information is publicly available.

The krypton-85 that has been emitted in past decades is by now well mixed into the atmosphere in both hemispheres and forms a global background (see Figure 9). Fed by global emissions over the years, this background has slowly increased. Historically, the background concentration has been somewhat higher in the Northern Hemisphere, because all known past and present reprocessing plants were located there and the exchange of air between the hemispheres is slow. Only since about 2005, when emissions have been reduced due to a decline in reprocessing in France, has equilibrium in the total inventory been reached. As krypton-85 is still released only in the Northern Hemisphere, the Southern Hemisphere continues to have a somewhat lower krypton-85 concentration.

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**Figure 8.** Locations where the German Federal Office for Radiation Protection (BfS) has been taking air samples for krypton-85 measurements. Depending on the location, measurements go back as far as 1973. Source: Adapted from Ole Ross, “Simulation of Atmospheric Krypton-85 Transport to Assess the Detectability of Clandestine Nuclear Reprocessing” (PhD thesis, University of Hamburg, 2010).
Figure 9. Historical development of the global krypton-85 concentration in lower levels of atmospheric air. Due to continued reprocessing activities, the background has increased over the decades. All known reprocessing has occurred in the Northern Hemisphere. The slow exchange of air between the hemispheres led on average to higher concentrations in the Northern Hemisphere. Source: Adapted from Ole Ross, “Simulation of Atmospheric Krypton-85 Transport.”
When a reprocessing plant is emitting krypton-85, the plume adds to the baseline concentration and a concentration above the baseline can be expected downwind. The additional concentration is highest close to the stack and diminishes over distance with the dispersion of the plume. Figure 10 shows the slow increase of the baseline concentration in Freiburg, Germany, over the decades and locally increased concentrations due to recent emissions from European reprocessing activities.

It is important to note that the fluctuations above the baseline shown in Figure 10 are specific for the detector location in Freiburg, Germany, which is downwind from the two largest currently operating reprocessing plants, La Hague in France and Sellafield in the United Kingdom. If measurements were taken closer to reprocessing plants, there would be larger and more frequent peaks above the baseline, while peaks would be less prominent at locations further away. Thus, if one is taking measurements to detect clandestine reprocessing in a region, it is necessary first to establish an understanding of local fluctuations to be able to determine whether a concentration above the baseline is due to a known facility.

Conversely, if a sample does not contain a concentration above the baseline, it does not mean that there is no reprocessing plant nearby. The wind might not be blowing from the plant toward the detector or the peak might be too small to be visible against the measurement uncertainties.

Figure 10. The local development of the atmospheric krypton-85 background over the decades of reprocessing. The baseline built up over time due to the steady feeding of the global atmosphere with krypton-85 faster than it is decaying. An equilibrium was reached in about 2005. The fluctuations above the baseline are due to recent emissions from upwind reprocessing plants. The plotted time series has been detected in Freiburg, Germany; the fluctuations including the highest peaks were caused mainly by reprocessing plants in La Hague in France (since 1967), Sellafield in the United Kingdom (since 1956) and Karlsruhe in Germany (1971–1990). Source: German Federal Office for Radiation Protection (BfS).
Monitoring known reprocessing facilities

The direct, on-site monitoring of reprocessing activities by the IAEA is the best way to safeguard declared reprocessing activities against plutonium diversion. Safeguards technology for such purposes, although costly, is well understood and accurate to about 1 percent. Additional assurance is provided by containment and surveillance at key points where plutonium might be diverted. Alternative forms of monitoring would be desirable, however, at facilities where intrusive on-site safeguards are not possible.

Aquatic monitoring

About 70 percent of the tritium from the reprocessing of spent fuel is released as tritiated water with aqueous or gaseous effluents. Reprocessing plants may have different effluent pathways, depending on the location, environment, and plant design. Waste-water can be released into standing or streaming water – such as rivers, lakes, or oceans – or pumped underground. For small operations, it could even be stored in tanks. The most applicable monitoring scheme would have to be adapted to the geographical location and design of the facility.

Aquatic monitoring for radioactive tracers could be used to monitor a known facility. Water samples from a river, a lake, groundwater, or seawater could be collected every few days and analyzed in a laboratory. As part of the effort during the 1990s to detect undeclared nuclear activities in Iraq, the IAEA conducted radionuclide monitoring of rivers, sediments, and other environmental pathways. Furthermore, the IAEA conducted field trials around well-known nuclear facilities to develop and demonstrate the detectability of various nuclear activities through environmental monitoring. Such techniques are suitable to monitor a specific facility as well as certain rivers or lakes in a region with suspected nuclear activities, including reprocessing.

Due to tritium’s relatively long half-life of about 12 years, there are no practical limits on the time after which it can still be detected. However, the long half-life also means that recently emitted tritium might be difficult to distinguish from accumulated, previously emitted tritium present in a standing body of water and would result in uncertainties about the time of release and even the source, if there is more than one possibility. Detection in a river can be advantageous in this regard, since rivers are self-flushing and have limited dilution.

Atmospheric monitoring

In the absence of on-site safeguards, the continuous monitoring of krypton-85 at nearby locations could be a viable option. The monitoring should be conducted so that it is as close to the source as possible. A stack monitor would be best because it would provide constant coverage of the emissions, independent of wind direction. It does not interact with the reprocessing activities but still reliably indicates average reprocessing rates. However, some facility operators might consider stack access by another organization to be too intrusive.

Alternatively, air samples could be taken outside of the plant’s perimeter or even further away in a downwind direction. Naturally, the minimum detectable reprocessing rate increases with distance. Research has quantified the separation rate of plutonium that would be reliably detectable at various distances (see Table 3). The sampling of
environmental air could be accomplished either by installing fixed monitoring stations with detection systems in the most likely wind directions or by collecting air samples and bringing them to a nearby laboratory where they could be analyzed.

<table>
<thead>
<tr>
<th>Distance</th>
<th>Minimum detectable reprocessing rate</th>
<th>Based on data from reprocessing plant</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.7 km</td>
<td>2 g/week</td>
<td>1 SQ per ~80 years Karlsruhe, Germany</td>
</tr>
<tr>
<td>5 km</td>
<td>40 g/week</td>
<td>1 SQ per ~4 years Karlsruhe, Germany</td>
</tr>
<tr>
<td>40 km</td>
<td>200 g/week</td>
<td>1 SQ per ~1 year Karlsruhe, Germany</td>
</tr>
<tr>
<td>60 km</td>
<td>6,300 g/week</td>
<td>1 SQ per ~10 days Tokai, Japan</td>
</tr>
<tr>
<td>130 km</td>
<td>1,000 g/week</td>
<td>1 SQ per ~2 months Karlsruhe, Germany</td>
</tr>
</tbody>
</table>

**Table 3. Detectable reprocessing rates at various distances under today’s background conditions.**
From the comparison between Karlsruhe and Tokai, it can be seen that the detectability depends not only on the distance but also on local wind patterns between the reprocessing plant and the sampling location.


Due to uncertainty in release patterns and atmospheric dispersion, this method would not be suitable for monitoring reprocessing rates for detecting excess activities above declared reprocessing. However, remote krypton-85 monitoring of a known facility could be used to verify the absence of reprocessing. This could find application in monitoring a known reprocessing plant that has been shut down unilaterally or due to a treaty commitment.

The ranges given in Table 3 also indicate that the absence of significant reprocessing could be verified for a small region within a country or, depending on geography, even outside of its borders.

**Israel.** Satellite imagery of the reprocessing plant in Dimona and suitable locations for remote monitoring are shown in Figure 11. Operational details about reprocessing activities at Dimona are scarce. The results represent lower boundaries for the detectability since they assume a hypothetical constant and continuous release rate, which would offer the highest chance to avoid detection. In reality, spent fuel is reprocessed in campaigns leading to krypton-85 being released in batches. The concentrated releases expected during reprocessing campaigns would lead to higher chances of detection and at longer distances for a given wind direction during these times.

For Dimona, the maximum distances and directions where emissions could still be detected on a regular basis are shown in Figure 11 for an assumed reprocessing rate of 18 kg of plutonium per year (about 350 g/week). The facility is capable of reprocessing at a higher rate, but lower outputs are also possible. A detector in southwestern Jordan would detect reprocessing at Dimona from outside the country on about 10 percent of the days in a year.
The fraction of the time that reprocessing at Dimona would be detectable could be increased by sampling at multiple locations simultaneously – for example, Jordan, Egypt, and the Red Sea. The most reliable verification of a reprocessing stop at Dimona, however, would be with a monitoring station within Israel a few kilometers or tens of kilometers south of the facility.

**Figure 11.** Top: Satellite imagery of the Dimona nuclear complex. It is believed that there are a heavy-water reactor and a reprocessing plant on the site. Krypton-85 is emitted together with other effluents from the stack centered within the circle. Source: Adapted from Google Maps, accessed on 20 June 2018. Bottom: The location of Dimona in the region and possible sampling locations with the fraction of days on which they could detect krypton-85 released from the site for a separation rate of 18 kg of plutonium per year.

**North Korea.** North Korea can produce plutonium in its 5 MWe reactor at a rate of about 20 g/day, amounting to about 1 SQ per year. The core of this reactor contains 50 tHM of fuel, which is exchanged after about 2–2.5 years. The irradiated fuel from one load would then contain about 16–20 kg of plutonium, of which about 96 percent is plutonium-239. The reprocessing plant at Yongbyon, see Figure 12 (top), has the capacity to reprocess about 100–150 tHM/yr, which greatly exceeds the country’s capacity to irradiate uranium in the 5 MWe reactor. It is estimated that a reprocessing campaign for a full reactor core (50 tHM) could produce about 2–2.5 SQ of plutonium in four to six months.
The detectability of krypton-85 emissions during reprocessing campaigns at Yongbyon is shown on the bottom left in Figure 12. Due to local topography and prevailing wind patterns, plumes from Yongbyon are confined to the region around the reprocessing plant and the detection range remains relatively short.

Even with a higher reprocessing rate, the krypton-85 emissions from Yongbyon are unlikely to be detected outside of North Korea (Figure 12, bottom center). Only extremely large releases of krypton-85 – from reprocessing 1 SQ per week or about 1,100 g/day of plutonium – would become detectable outside of North Korea. Even then, there is no dominant wind direction to guide preferred sites for monitoring stations.

However, even for low emissions, elevated krypton-85 concentrations can be detected in Pyongyang, which is about 100 km away. For an assumed reprocessing rate of about 1 SQ every two months (about 135 g/day of plutonium) during campaigns, about 15 percent of emission days from Yongbyon would be observable in Pyongyang.

Figure 12. Top: Satellite imagery of the Yongbyon reprocessing facility and possible points of emissions. Source: Adapted from Google Maps, accessed on 20 June 2018. Bottom left: Detection probabilities around Yongbyon for the estimated current reprocessing rate of 1 SQ of plutonium every two months. Bottom center: At the higher reprocessing rate of 1 SQ/week of plutonium, which could be possible in the future or with a concentrated reprocessing campaign. Due to the location of Yongbyon and the prevailing wind patterns, detections of krypton-85 emissions from Yongbyon are unlikely to be made outside of the country. Detection equipment in Pyongyang would have a chance of detecting krypton-85 emissions from Yongbyon.
Reprocessing at the Pilot Demonstration Center at the Mining and Chemical Combine in Zheleznogorsk, near Krasnoyarsk, in Russia, is starting up. There are plans for a larger reprocessing plant at the site. These plants may lead to increased fluctuations of the krypton-85 concentration over North Korea, impeding the monitoring of activities at Yongbyon.

If it begins full-scale operation, the much delayed Rokkasho reprocessing plant in Japan with its capacity to reprocess 800 tHM/y will not directly increase the krypton-85 fluctuations over North Korea and affect the detectability of North Korean reprocessing. The general wind direction from west to east in these latitudes will push krypton-85 emissions from Rokkasho over the Pacific Ocean. However, reprocessing at Rokkasho plant could contribute significantly to the global krypton-85 output and increase the background baseline concentration. This would make the detection of clandestine reprocessing plants more difficult on a global scale, and offset the gains from the planned shutdown of the Sellafield reprocessing plant.
Detecting clandestine reprocessing facilities

Undeclared nuclear programs and facilities remain among the most difficult challenges in nuclear nonproliferation. A recent example of a clandestine nuclear activity in the Middle East was a Syrian reactor at the al-Kibar site that was destroyed by an Israeli airstrike in 2007. Israel alleged that the facility had been built for the production of plutonium. That it was a reactor was confirmed in May 2011 by the IAEA. The reprocessing plant at which plutonium would have been separated out from the uranium irradiated by the reactor has not been identified. Such a facility could be difficult to detect before becoming operational.

Typical reprocessing plants tend to have a long, narrow building, called a canyon, with thick radiation-shielding walls, which can be a telltale sign during construction. Otherwise, reprocessing plants do not have a unique feature for remote satellite detection, and WAES is the most promising method for finding clandestine facilities. Aquatic monitoring of radionuclides is not reliable for this purpose. However, atmospheric monitoring of krypton-85 concentrations offers ways to detect such activities. Since improvised and newly built reprocessing plants alike can remain undetected via satellite imagery and therefore would not trigger focused monitoring efforts, any detection method must cover large areas.

Stationary monitoring networks

A worldwide fixed-site network of krypton-85 monitors to detect clandestine reprocessing would pose a number of challenges similar to those involved in the detection of undeclared nuclear explosions using radionuclide releases as part of the verification of the Comprehensive Test Ban Treaty (CTBT). An unannounced nuclear explosion could, in theory, happen anywhere on the globe at any time. The CTBT’s worldwide verification system therefore includes fixed monitoring stations that constantly take and analyze atmospheric air samples for the characteristic radioactive tracers that would be released by a nuclear explosion. Noble gas releases from an underground nuclear test can be prompt and short in duration.

For CTBT verification, 20 radionuclides of interest have been identified, based on their fission yield, transport losses, background levels from other sources, and their ability to be detected. For the radionuclides of choice, it has been calculated that 80 monitoring stations will be sufficient to cover the globe so that there will be 90 percent certainty of detecting an explosion that has a yield equivalent to 1 kiloton of TNT. Four radioactive xenon isotopes are monitored as the most likely to be leaked from underground explosions. Due to their low background levels, detection of these isotopes is possible at distances of thousands of kilometers. The stations have therefore been spaced at distances in the range of 1,000–2,000 km. A total of only 40 monitoring stations have been deemed sufficient to detect unusual radioactive xenon releases anywhere. With a capital cost of $1–2 million per station and an annual operational cost of $100,000–200,000, the establishment and maintenance of the CTBT monitoring network is a multinational effort supported with annual contributions.
Because of the short half-lives of the radioactive xenons, the background that impacts CTBT verification is only found downwind from medical radioisotope production facilities and clusters of nuclear power reactors. In contrast, the krypton-85 background has accumulated in the atmosphere all around the world for decades and a release of krypton-85 from a small clandestine reprocessing facility would become undetectable against the background within a few hundred kilometers. A fixed krypton-85 monitoring network therefore would require a much higher station density than the CTBT radioactive xenon network. Figure 13 shows a hypothetical network with a station separation of 300 km aimed at detecting clandestine reprocessing activities in Europe. Such a network with hundreds of stations may be seen as too costly for treaty monitoring.

Random sampling

The potential cost of a high-density, fixed monitoring network could be avoided by taking air samples. This would avoid the capital and maintenance costs of fixed monitoring stations but shift costs to sampling logistics and laboratory analysis. Air samples also could be taken at random locations to prevent proliferators from timing their emissions to avoid detection. After collection, samples would be brought to the nearest laboratory for analysis.

An operating reprocessing plant emits krypton-85 into the atmosphere. To successfully monitor a large region for ongoing reprocessing by taking air samples at random locations, a major factor is the spatial extent of the plume where this krypton-85 release is still detectable. This plume area can be taken to be the krypton-85 footprint of the
Remote detection of undeclared reprocessing facility. The daily emissions from any given day can overlap with the emissions from previous days, however. Over time, the plume continues to form, shift, and disperse while adding to the local krypton-85 background concentration. Table 4 shows the sizes of typical footprints for different reprocessing rates and different emission patterns. The footprint, where the plume concentration is still detectable, depends on the background baseline and fluctuations.

<table>
<thead>
<tr>
<th>Plutonium separation rate</th>
<th>Days for 1 SQ</th>
<th>Emission pattern</th>
<th>Average krypton-85 footprint [km²]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Today's background</td>
</tr>
<tr>
<td>22 g per day</td>
<td>365</td>
<td>Daily</td>
<td>850</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Weekly</td>
<td>1,800</td>
</tr>
<tr>
<td>270 g per day</td>
<td>30</td>
<td>Daily</td>
<td>23,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Weekly</td>
<td>25,000</td>
</tr>
<tr>
<td>1,100 g per day</td>
<td>7</td>
<td>Daily</td>
<td>120,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Weekly</td>
<td>125,000</td>
</tr>
</tbody>
</table>

Table 4. Detectable footprint sizes of krypton-85 emission from various reprocessing rates and emission patterns under today’s background conditions and after 30 years of background decay after emissions have stopped. Daily emissions are released continuously; weekly emissions are released over one day per week. The footprint is averaged over the time needed to separate 1 SQ. If the background could decay for thirty years due to an emission stop, the footprint would increase about tenfold. Source: Based on calculations presented in Schoepfner, and Glaser, “Present and future potential of krypton-85.”

If an air sample is being taken within a larger area that contains an operating reprocessing plant, the chance that the sample contains an elevated krypton-85 concentration is determined by the size of the plant’s footprint. The probability of detecting the reprocessing activities with one sample is simply the ratio of the footprint to the total area that is being considered. By repeatedly taking samples at random locations, one can increase the probability of taking a sample from within the krypton-85 footprint of a clandestine reprocessing plant. Comparing the size of the footprint with the total size of the area to be monitored, one can derive how many random samples must be taken in order to reach a desired detection probability. For example, for verification purposes, a detection probability of 90 percent may be desirable. Then, depending on the verification goals for a given reprocessing rate, one can calculate the number of samples that are required. The search area can be reduced by excluding sea, desert and mountain regions without infrastructure.

**Timeliness of detection**

A fixed monitoring network makes it possible to detect releases whenever they occur and as soon as possible. This ability is crucial for detection of the type of unannounced, short-puff releases that can occur from nuclear explosions. However, reprocessing plants release krypton-85 over longer periods of time. If the detection goal is to detect a signal within the time required to separate 1 SQ of plutonium, the number of samples can be reduced drastically.
A clandestine reprocessing facility that separates large amounts of plutonium in a short time generates a plume of high krypton-85 concentrations that has a large footprint and can be detected over long distances. In such a scenario, quick detection would be required to react to the clandestine activities before enough plutonium for an explosive device could be produced. The large footprint of such a process would facilitate the timely detection. On the other hand, a reprocessing facility that slowly accumulates plutonium creates a much lower krypton-85 signature that is more difficult to detect, but it also allows for a longer time for detection before a significant quantity of plutonium is separated. This trade-off is shown in Table 4 and Figure 14.

**Figure 14.** Qualitative krypton-85 emissions from 1 SQ of plutonium for different separation rates. The krypton-85 is released during the reprocessing campaign; emission levels and duration depend on the separation rate of plutonium. A high plutonium production rate leaves a larger footprint and is therefore easier to detect but allows less time for detection before 1 SQ has been separated. Similarly, a low plutonium production rate has lower emissions and therefore is more difficult to detect, but it allows more time for detection. 
*Source: Schoeppner, “Detecting Clandestine Reprocessing Activities in the Middle East.”*

**Impact from the krypton-85 background**

The number of samples needed to detect clandestine reprocessing plants over large areas depends strongly on the background conditions. The baseline and the fluctuations downwind from large reprocessing plants can hinder the successful detection of emissions from clandestine facilities.
A high background level with strong fluctuations effectively decreases the detectable footprint of a reprocessing plant and therefore increases the number of random samples required for detection. This effect is strongest for low reprocessing rates where low krypton-85 emissions are unlikely to rise above the background and become more easily hidden in the background. Strong emissions and their footprints are less affected by background levels. Therefore, even given the longer time available to detect a small reprocessing plant, with today’s background conditions, more random samples are required to detect a small clandestine than a large reprocessing plant. If there were no accumulated background, the number of samples needed per day would be roughly the same for all reprocessing rates to be detected before 1 SQ of plutonium can be separated.

The future of the krypton-85 background

The future of large-scale reprocessing will determine the detectability of clandestine facilities. Historically, the global emission rate of krypton-85 reached its peak in the late 1990s and since then on average has declined due to a lowered rate of reprocessing. This led to a slight decline of the global background levels toward a new equilibrium. Future reductions of emissions would result in the background decaying to a new equilibrium or even zero. Such reductions in reprocessing are possible because civilian reprocessing has proven to be uneconomical. For example, the United Kingdom is in the process of shutting down its two reprocessing plants in Sellafield.

Even after a hypothetical stop of all emissions, however, the krypton-85 background would only decay with a half-life of 10.8 years. Thus, a significant reduction would take a few decades. If a reprocessing plant stops its emissions from one day to the next, the fluctuations downwind from that plant would cease within a few days. As the fluctuations from fresh plumes are reduced, the detector uncertainty becomes more important to also identify small variations in the krypton-85 concentration.

In the long term, the lowered global emission rate would also lead to a new equilibrium for the baseline. Four scenarios on the future development of krypton-85 emissions and the resulting background are presented in Figure 15.

A reduced background would facilitate the detection of clandestine reprocessing plants. Over time, with a reduced background, a lower number of samples would be necessary to maintain the same detection probability. Alternatively, a constant number of samples would increase the speed of detection.

A reduction of krypton-85 emissions through krypton-85 capture before release would yield the same effect as lowered reprocessing rates. Also, if reprocessing operators made available their emission data, it would make it possible to predict the background with atmospheric transport modeling and subtract it to better detect clandestine reprocessing. However, operators of reprocessing plants have no strong incentive to publish time-resolved data on radioactive stack emissions.
Figure 15a. Hypothetical future developments of krypton-85 emissions and atmospheric background. Increased emissions would add to the background. Source: Based on and extended from Ahlswede et al., “Update and improvement of the global krypton-85 emission inventory.”

Figure 15b. Hypothetical future developments of krypton-85 emissions and atmospheric background. Constant emissions would lead to an equilibrium. Source: Based on and extended from Ahlswede et al., “Update and improvement of the global krypton-85 emission inventory.”
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Figure 15c. Hypothetical future developments of krypton-85 emissions and atmospheric background. Decreased emissions would let the background slowly decrease. Source: Based on and extended from Ahlswede et al., “Update and improvement of the global krypton-85 emission inventory.”

Figure 15d. Hypothetical future developments of krypton-85 emissions and atmospheric background. Zero emissions would let the background decay with the krypton-85 half-life of 10.8 years. Source: Based on and extended from Ahlswede et al., “Update and improvement of the global krypton-85 emission inventory.”
Simulations have shown that compared to today's background conditions the number of samples required for a given detection probability would be reduced on average by 20 percent if krypton-85 emissions stopped. The reduction would be large in regions with active reprocessing and negligible in regions without fresh plumes, such as the Southern Hemisphere. By stopping krypton-85 emissions and allowing the background baseline to decay for 10 years, the number of samples could be reduced by 60 percent. After 30 years, the number of samples could be reduced by about 90 percent — that is, only 10 percent of the number of samples needed today would be needed to maintain the same detection capabilities.

Under the current krypton-85 background conditions, verifying the absence of undeclared reprocessing activities is a technical, logistical, and financial challenge. If emissions from declared reprocessing were stopped or drastically reduced, over time, verification with random sampling would become a small and feasible operation.

**Number of samples**

The main factor that determines the feasibility of random sampling for krypton-85 monitoring is the number of samples that need to be collected and analyzed per day to ensure the desired detection capability. The number of random samples required to detect clandestine reprocessing with 90 percent probability can be calculated based on the footprint in relation to the total area to be monitored as shown in Table 4. Under today's background conditions, about 50 samples per day are required to monitor a region of 10 million km² in the Northern Hemisphere for the absence of reprocessing activities of more than 1 SQ per month (more than 270 g of plutonium per day).

Due to the high background level, lower reprocessing rates would have significantly lower probabilities. For example, there would be only a 50 percent probability of detecting a reprocessing program that had a rate of 1 SQ per year (22 g Pu per day). For such a reprocessing rate, a detection probability of 90 percent would require more samples — about 150 per day.

With regard to the territory covered, 10 million km² is roughly the area of China or the United States or the collective area of the Middle Eastern countries with research reactors or plans for a nuclear power program (Algeria, Egypt, Iran, Iraq, Israel, Jordan, Libya, Saudi Arabia, Syria, and the United Arab Emirates). The area of Europe also is about 10 million km², but krypton-85 monitoring efforts to detect clandestine reprocessing are made virtually impossible there due to the large reprocessing activities in France and the United Kingdom that create high krypton-85 concentrations above the background baseline. These fluctuations would mask the signal from an undeclared reprocessing plant.

Table 5 lists some measures to reduce the number of samples and the estimated benefit. Each region is subject to specific meteorological patterns. Individual case studies would be needed to provide more-specific results.
The number of samples scales linearly with the total size of the search region. Most regions contain areas with a lack of infrastructure such as roads, power supply, and housing that would be needed to start and run a reprocessing plant and possibly a nuclear reactor. Excluding such areas from random sampling could drastically reduce the number of samples. Sampling then would be focused on areas with at least basic infrastructure. Halving the total search region by exclusion of unlikely areas would, for example, also halve the number of required samples.

<table>
<thead>
<tr>
<th>Samples per day per 10 million km²</th>
<th>Reduction to</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reprocessing of &gt;1 SQ per month (270 g Pu per day), under today’s background conditions and a target detection probability of 90% in area with medium background fluctuations</td>
<td>50</td>
</tr>
<tr>
<td>Reprocessing rate of 1 SQ per year (22 g Pu per day)</td>
<td>150</td>
</tr>
<tr>
<td>Making krypton-85 release data available</td>
<td></td>
</tr>
<tr>
<td>Reducing the search region, e.g. by excluding half</td>
<td></td>
</tr>
<tr>
<td>Reducing target detection probability from 90% to 50%</td>
<td></td>
</tr>
<tr>
<td>Stopping emissions from declared reprocessing (automatically applies to Southern Hemisphere)</td>
<td></td>
</tr>
<tr>
<td>10 years later</td>
<td></td>
</tr>
<tr>
<td>30 years later</td>
<td></td>
</tr>
</tbody>
</table>

Table 5. Overview over the numbers of samples to be taken at random locations to detect undeclared reprocessing and measures to reduce them. Medium background fluctuations of the krypton-85 concentration in the atmosphere are considered standard deviations between 0.05 and 0.5 Bq/m³. Sources: Based on Schoepfner and Glaser, “Present and future potential of krypton-85,” and M. Schoepfner, “Detecting Clandestine Reprocessing Activities in the Middle East,” Science & Global Security, 26(1), 2018.

**Sampling-site selection**

Most parts of a large area to be monitored would be covered by taking air samples at random locations, while known and shut-down reprocessing plants could be covered by fixed monitoring stations.

**Sampling at random locations.** The detection of a previously unknown reprocessing plant can be accomplished with random sampling. Taking a number of samples at random locations over a large area is mainly a logistical effort. With a daily goal of samples that have to be taken, the location where each sample should be taken should be determined every morning or the day before. That way, the information would not be available to the operators of the clandestine facility, who could respond by suspending operations when a sample is planned to be taken within detection range. This is an advantage over a fixed network of monitoring stations, where the operator of a clandestine reprocessing plant could use meteorological forecast data to release waste gas on days when the plume would not hit a monitoring station.
The detection probability could be further optimized by ensuring certain quality checks for the distribution of random locations. Such quality checks could include a minimum distance between two sampling locations on a given day, a minimum number of samples per sub-region, and sophisticated search patterns to avoid creating the same blind spots on successive days. Search strategies and flight patterns were discussed in the IAEA’s STR-321.65

Another way to optimize the effectiveness of detection would be for certain findings to trigger particular follow-up actions. For example, a suspicious concentration could trigger another sample at the same location on the following day. Satellite imagery or news reports also could trigger further investigation after logistics or construction of a new industrial complex in a remote location was discovered.

**Monitoring stations.** Known, shut-down reprocessing sites that have to be monitored for their inactivity could be covered by two or three fixed monitoring stations. Depending on the typical meteorological patterns, these krypton-85 monitoring stations can give virtually constant coverage throughout the year. Such monitoring stations also could support the random-sampling efforts, as no additional samples would have to be taken at such monitoring stations.

Existing national stations are sparse but could provide additional data points to support the random-sampling mission. Additional samples could be taken during routine IAEA safeguard inspections; some of these inspections are conducted without notice. With this information in hand, analysts could exclude the possibility that clandestine reprocessing activities had been taking place within detection range of these sites.

**Sampling and analysis.** Once the locations for the taking of samples on a particular day are determined, several options exist for collecting them. To detect recent emissions from a clandestine reprocessing plant, the air samples should be collected below the planetary boundary layer. The height of the layer is variable and depends on local and seasonal meteorological conditions; it typically ranges between 1 and 2 km, depending on wind and temperature conditions. Above that layer, any emission becomes much more dispersed and diluted and is likely to be undetectable against the existing background. Sampling at higher altitudes was useful to estimate total plutonium stockpiles during the Cold War when the background was low, but it would not be suitable for detecting recent activities today.

Airborne collection offers the best way to cover large distances and collect multiple samples in one go. Aircraft and drones with various ranges offer a variety of options. Air samples for current-generation detectors need a volume of more than 200 liters. Next-generation detectors will require much smaller sample volumes – on the order of a few liters. For such small samples, drones equipped with a few sample bottles could be used for low-cost collection.

Assuming that a large number of air samples have to be collected over large areas, however, medium-sized airplanes and drones with ranges of several thousand kilometers...
would be more appropriate. As the samples have to be collected at lower altitudes, the aircraft or drone would frequently have to dive down below the planetary boundary layer to collect each sample and return to cruising altitude.

Once the samples have been collected, they have to be delivered to one or more laboratories. In its safeguards missions, the IAEA splits environmental samples to send them to more than one laboratory for increased confidence. The determination of the krypton-85 concentration of an air sample with conventional beta-decay measurement systems takes about four to 12 hours, depending on the sample volume. Each measurement system can therefore handle about two to six samples per day. The next-generation ATTA detector takes about three to four hours and could therefore handle about six to eight samples per day.

Samples indicating significantly higher than expected krypton-85 concentrations could trigger a closer investigation including satellite imagery, helicopter overflights, or ground-based inspections of buildings.

**Middle East**

If the Middle East were to be monitored for absence of reprocessing, the task could be accomplished by a combination of measures. Ten Middle Eastern countries have or had nuclear programs or have announced plans for nuclear research and/or power reactors: Algeria, Egypt, Iran, Iraq, Israel, Jordan, Libya, Saudi Arabia, Syria, and the United Arab Emirates, covering a total of about 9.8 million km². As part of the effort to reduce the monitoring area and to maximize the detection efficiency, the sampling should be focused on those countries.

Figure 16 illustrates how a region of 10 million km² can be divided into sub-regions around airport hubs. For a 90 percent probability of detection of medium-sized reprocessing plants, about 50 samples per day would be required.

In this example, where the region is divided into three sub-regions, each airport would need to collect about 17 air samples on average. The reduction of the search area by excluding contiguous desert without infrastructure and a further decline of declared reprocessing activities around the world and especially in Europe would reduce the number of samples (see Table 5).
North Korea

The principle of random airborne sampling can also be applied in North Korea. North Korea has a total area of 120,000 km². Monitoring the whole country for clandestine reprocessing activities would require two to three samples per day at random locations. Due to the relatively small size of the country and the limited number of samples, one aircraft at an airport in South Korea, China, or Russia could sustain such an operation.

Reprocessing in neighboring countries could affect krypton-85 monitoring efforts in North Korea. The general wind direction in these latitudes from west to east tends to bring air masses from China and Russia, but krypton-85 releases from these countries are currently not strong enough to significantly impede possible monitoring efforts in North Korea. Only massively increased reprocessing activities or new reprocessing plants closer to North Korea would potentially significantly influence krypton-85 concentrations over North Korea.

Due to the general wind direction, reprocessing in Japan is predicted to have almost no effect on the local detectability of krypton-85 in North Korea. But the startup of Japan’s Rokkasho reprocessing plant would contribute to the global atmospheric inventory of krypton-85 and, over time, hinder monitoring efforts in North Korea.
Conclusion

A clandestine reprocessing program can be taken as a reliable sign that a state is producing plutonium as part of pursuing the acquisition of nuclear weapons. An international capability that reliably detects clandestine reprocessing activities would be a valuable tool for the verification of the NPT, a future FMCT, and possibly also regional nuclear-weapon-free zones.

The best chance to learn about the operation of a clandestine reprocessing plant is to detect the gaseous fission product krypton-85 released when the cladding of spent uranium fuel from a reactor is opened and the fuel dissolved to recover the plutonium from the spent fuel. It also offers ways to detect undeclared activities in a known facility. Decades of reprocessing have created a global krypton-85 background in the atmosphere that reduces the range at which a downwind plume can be detected, however. Krypton-85 monitoring could still be readily applied to confirm shutdown of known facilities by taking air samples at a small number of fixed locations in the most frequent wind directions.

Due to the high krypton-85 background, the required high-density network of fixed monitoring stations to detect clandestine facilities in a large region would be prohibitively costly. Taking air samples every day at random locations would be a potentially less costly alternative. The number of samples needed each day would be dictated by the fact that a reprocessing plant does not have to be detected the moment it starts reprocessing. It may be sufficient to detect its operation within the time the plant needs to separate 8 kg of plutonium, the amount the IAEA defines as the significant quantity required to build a nuclear weapon.

To impede clandestine reprocessing before it can even be started, spent fuel should be subject to international safeguards wherever possible. In a scenario in which such safeguards are effective, a country seeking to make nuclear weapons would need to build and run a clandestine reactor and a clandestine reprocessing plant.
Detection of clandestine reprocessing through krypton-85 emissions could be made more difficult by installing effective systems at the plant to capture and store the krypton. Such systems are technically feasible but are sufficiently costly and difficult to operate that no current reprocessing plant is known to have installed one for this purpose.

Verification of the absence of clandestine reprocessing through random air sampling on a global scale would currently require a large logistical effort. It is more feasible on a local or regional level. The single biggest hindrance of such a focused approach is the high krypton-85 background that has accumulated over decades of reprocessing. If emissions from civilian reprocessing facilities were declared to the IAEA or another international agency assigned responsibility for krypton-85 surveillance, it would be possible to make more accurate predictions of the plumes downwind from known facilities and better identify smaller krypton releases from undeclared sites.

In a world without permitted reprocessing, it would be easier to detect undeclared reprocessing activities. The separation of plutonium on a large scale for nuclear-weapon purposes ended with the end of the Cold War, and reprocessing for civilian purposes could end also. It has proven to be uneconomic and unnecessary for nuclear energy programs. There is no sensible argument for states to continue their reprocessing programs. An end to civilian reprocessing would mean krypton-85 background would be much reduced in the future as the existing krypton-85 decays.

Ending all production of plutonium for any purpose as part of an FMCT would make it easier to detect clandestine reprocessing than if the FMCT ended only plutonium production and separation for nuclear weapons or other explosive devices. Ending reprocessing for any purpose should therefore be seen as a step towards improving global nonproliferation, arms control and disarmament verification.
Endnotes

1. North Korea was a party to the Nuclear Non-Proliferation Treaty but withdrew in 2003.

2. Pakistan’s first nuclear warheads were produced with highly enriched uranium (HEU). Most or all of Israel’s nuclear weapons use domestically produced plutonium, but the first fissile material available to the country may have been HEU stolen from a U.S. naval fuel production facility. See also Grant F. Smith, *Divert!: Namoc, Zalman Shapiro and the Diversion of US Weapons Grade Uranium Into the Israeli Nuclear Weapons Program* (Washington, DC: Institute for Research: Middle Eastern Policy Inc., 2012).

3. The nuclear weapon tested on 16 July 1945 was plutonium-based. The uranium-based bomb type dropped on Hiroshima has not been tested prior to the attack on 6 August 1945.

4. U.S. and South Korean national laboratories developed pyrochemical reprocessing, or pyroprocessing. It is based on electrorefining to separate fission products. Spent fuel is converted to metal through oxide reduction. Electric current dissolves the spent fuel and causes actinides to accumulate on the cathode. Due to the higher temperatures of this process, the spent fuel requires less cooling before it can be reprocessed. Pyroprocessing has a lower separation efficiency compared to the PUREX process and therefore is currently only used for special operations in the United States, South Korea, and Russia.


9. Japan, as a NPT non-nuclear-weapon state, is subject to IAEA safeguards. Civilian nuclear activities in France and the United Kingdom are subject to Euratom safeguards. The United Kingdom plans to withdraw from Euratom as part of Brexit, but it also plans to end reprocessing within the same time frame.


15. Ferguson, Simple, Quick Reprocessing Plant.


19. Goodman, Spying on the Nuclear Bear.


24. IRSN, “Radionuclide sheet.”

31. IRSN, “Radionuclide sheet.”


33. Radioactive xenon isotopes are a suitable tracer for nuclear explosions but are also emitted by nuclear power plants and medical-isotope production facilities.

34. This figure is based on Monte Carlo N-Particle (MCNP) calculations; see also Michael Schoepner and Alexander Glaser, “Present and future potential of krypton-85 for the detection of clandestine reprocessing plants for treaty verification,” *Journal of Environmental Radioactivity* 162–163 (2016): 300–309, http://dx.doi.org/10.1016/j.jenvrad.2016.06.001.


36. After removing NO 2 and other impurities, the gas consists mostly of air, reduced amounts of NO x, small amounts of hydrogen, and rare gases. The gas mixture is processed through a rhodium catalyst bed at 540–650°C to reduce the NO x to nitrogen and let the hydrogen form water. Then the dried gas can be cooled with heat exchangers and pumped through a cryogenic column where O 2 , krypton, xenon, and argon are absorbed into a countercurrent stream of liquid nitrogen.


38. This method employs dichlorodifluoromethane (freon-12) for selective absorption and was developed at Oak Ridge National Laboratory and the Karlsruhe Research Center.

39. This method employs CO 2 as a solvent and requires an off-gas streams with high CO 2 concentrations. Therefore, it could be suitable for off-gas streams from reprocessing of graphite fuels.

40. Soelberg et al., “Radioactive iodine and krypton control for nuclear fuel reprocessing facilities.”
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Jacob, *Introduction to Atmospheric Chemistry*, Chapter 4.


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 fluorescence.


From personal experience of the author at CTBTO events.

Ross, “Simulation of Atmospheric Krypton-85 Transport.”

The footprints are based on atmospheric dispersion forward simulations for hypothetical reprocessing plants. The footprints are averaged for locations scattered on continents of northern and southern hemispheres. Depending on the location of a specific reprocessing plant the footprint can be smaller or larger depending on prevailing wind patterns and closeness to declared reprocessing plants.

Schoeppner and Glaser, “Present and future potential of krypton-85.”

*Plutonium Separation in Nuclear Power Programs.*

Schoeppner and Glaser, “Present and future potential of krypton-85.”

This analysis is based on methods presented in Schoeppner and Glaser, “Present and future potential of krypton-85.”

Fifty samples per day to monitor 10 million km² is taken to be an approximate and representative number for various scenarios in the Northern Hemisphere. Depending on the monitored region’s wind patterns and background fluctuations from recent emissions, this number can vary. Special case studies would have to be conducted for specific regions to determine more-concrete numbers.

IAEA *Use of Wide Area Environmental Sampling.*

Zappala et al., “Setting a limit on anthropogenic sources of atmospheric "Kr."

This analysis is based on results presented in Schoeppner, “Detecting Clandestine Reprocessing Activities in the Middle East.”
About the author

Michael Schoeppner is a researcher working on the technical verification of nuclear arms control agreements. His work focuses on computer modeling related to environmental radioactivity, nuclear-weapon tests, detectability of nuclear activities, and the consequences of nuclear accidents. He received his PhD in physics from Roma Tre University in Italy and was a postdoctoral researcher at the Program on Science and Global Security at Princeton University. He is based in Vienna, Austria.

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