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Environmental Detection of Clandestine Nuclear Weapon Programs

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Abstract Environmental sensing of nuclear activities has the potential to detect nuclear weapon programs at early stages, deter nuclear proliferation, and help verify nuclear accords. However, no robust system of detection has been deployed to date. This can be variously attributed to high costs, technical limitations in detector technology, simple countermeasures, and uncertainty about the magnitude or behavior of potential signals. In this article, current capabilities and promising opportunities are reviewed. Systematic research in a variety of areas could improve prospects for detecting covert nuclear programs, although the potential for countermeasures suggests long-term verification of nuclear agreements will need to rely on methods other than environmental sensing.

1. INTRODUCTION

The first environmental detection of a secret nuclear weapons program happened by accident. In September 1945, batches of fresh X-ray film produced by Eastman Kodak were corrupted with speckles and faint lines. Scientists at Kodak traced the problem to beta-emitting isotopes in the cardboard used to package the films. The cardboard had been produced at paper mills in Indiana and Iowa that drew water from rivers that had recently flooded and, in the process, had washed in radioactive fallout from the Trinity Test of July 1945, which occurred nearly 1,000 miles away and was still a national secret (Webb 1949). As a result of this revelation, the US Army Air Force began a program to search for nuclear detonations in other countries by sampling air for radioactive aerosols. This program successfully detected the first Soviet nuclear test in September 1949. The episode paved the way for environmental monitoring of many nuclear activities and accords under the heading of "national technical means" (Richelson 2006, Ziegler & Jacobson 1995).

The environmental detection of nuclear explosions is now a well-established science (Bowers & Selby 2009, Richards & Zavales 1990). However, for many security objectives, detecting a nuclear detonation comes too late. More useful would be an ability to detect nuclear weapons programs well before the achievement of a nuclear device. To do this, International Atomic Energy Agency (IAEA) Safeguards were created to detect and deter the misuse of civil nuclear technology (IAEA 2002, Taylor 1975). However, Safeguards are not able to detect proliferation activities at clandestine locations, as have been operated at various times by Australia, Brazil, Canada, China, Egypt, France, India, Iran, Iraq, Israel, Libya, North Korea, Pakistan, Romania, Russia, Serbia, South Africa,

South Korea, the Soviet Union, Sweden, Switzerland, Syria, Taiwan, the United Kingdom, and the United States.

In the several cases where clandestine programs were detected at an early stage, the large majority were stopped through political engagement (Campbell et al. 2004). There is, as such, a strong motivation for improving the probability of detection by environmental sensing (CISAC 2005, IAEA 1999, US Congr. OTA 1995, Wogman 2013). The usual focus of detection is on the production of fissile materials, the nuclear explosive ingredient in a nuclear weapon. Fissile materials require considerable technology and energy to make, and their production is usually the rate-limiting step in a proliferation program. Furthermore, production involves exotic facilities, materials, and processes, which can generate particular environmental signatures. However, despite decades of work, researchers have yet to discover a reliable and cost-effective detection mechanism. This review summarizes the technical parameters of the problem, known source terms for detectable signals, the capabilities of existing techniques, and promising avenues for future research.

2. FISSILE MATERIALS

A fissile material is any material capable of sustaining a fast-fission chain reaction. Such materials contain high concentrations of one or more fissile isotopes, which are the isotopes that have nuclei with $90 \le Z \le 100$ protons and that satisfy the relation $2Z - N = 43 \pm 2$, where N is the number of neutrons. Of the 33 identified by this formula, only the uranium isotopes ²³⁵U and ²³³U and the long-lived plutonium isotopes ²³⁹Pu and ²⁴⁰Pu are considered under most monitoring schemes; the others have half-lives that are too short, or critical masses that are too large, to be practicably used in nuclear weapons. A handful of exotics, namely ²³⁶Np, ²³⁷Np, and ²⁴⁵Cm, are practical for nuclear weapons but are not practical to produce as the methods used can simultaneously produce much larger quantities of plutonium or ²³³U.

Of the isotopes meriting concern, ²³⁵U is the only one found in nature. It makes up 0.711 wt% of natural uranium, which in turn makes up about 2 ppm of Earth's crust, although the richest ores can exceed 2% uranium by mass. The ²³⁵U isotope can be concentrated to weapon-usable levels by a variety of isotope separation techniques. Dozens of techniques have been demonstrated, but only a few have been found to be viable at scale, and only the gas centrifuge is in commercial use today. Laser isotope separation (LIS) is a promising method that might become competitive in the near future and is being actively studied by several countries. Additionally, aerodynamic methods are now being revisited (Ronander et al. 2012).

By contrast, plutonium, ²³³U, and all other fissile isotopes are synthesized by neutron transmutation of naturally occurring fertile isotopes, such as ²³⁸U and ²³²Th. Because transmutation requires about four moles of neutrons per kilogram of fissile material, this process is most commonly done with nuclear reactors, the least expensive way to produce large numbers of neutrons. It is also technically feasible, however, to produce these materials by using neutrons from particle accelerators or even fusion reactors (Glaser & Goldston 2012, Kemp 2005). All reasonably feasible paths must be considered when envisioning a detection system or else the system risks highlighting one proliferation path that is immune to detection.

Table 1 outlines the nuclear fuel cycle in three major categories: precursors, separation methods, and transmutation methods. A tight system of detection requires that all the technologies in the precursors category be detectable, or that all technologies in one of the two functional steps in the separation methods category and one of the two functional steps in the transmutation methods category be detectable.

Table 1

Precursors	Separation Methods		Transmutation Methods	
Mining	Conversion	Isotope Separation	Transmutation	Reprocessing
Mining U or Th open pit underground in-situ leach seawater† byproduct extraction	Fluorination of U to UF ₄ by HF to UF ₆ by F ₂	Uranium Enrichment gas centrifuge gaseous diffusion molecular LIS atomic LIS*† exotics†	thermal reactors breeding blankets accelerators fusion reactors†	organic-aqueous precipitate electrorefining†
Concentration solvent extraction ion-exchange resin	Reduction of oxides, are not significant sources. * Atomic LIS does conversion.	Plutonium Isotope separation of spent fuel. not depend on		

[†] Not covered in this review owing to technical immaturity.

3. DETECTION GOALS

The strategically meaningful minimum detectable signal emanating from any of the above-listed processes depends on the rate of nuclear weapons production considered to be a significant perturbation of international security. The most stringent detection goals are defined for states that do not already possess nuclear weapons, where the acquisition of even a single weapon would result in a significant strategic shift. IAEA Safeguards were established to "detect and deter through the risk of detection" the acquisition of a nuclear weapon by this category of states. By international consensus, the IAEA's detection goal was set at what is effectively one nuclear weapon per year. This objective is operationalized using significant quantities (SQ), the amount of material at various stages of readiness notionally needed to make a single weapon, and by IAEA established timeliness requirements for detection at each stage in the process (IAEA 2002). The equivalent facility throughput rates based on these values are given in Table 2. Although IAEA standards have been criticized for being too permissive (Cochran & Paine 2005), the values used are both widely accepted and consistent to within a factor of about two with most historical covert programs (Kemp 2010a).

Table 2: IAEA Implied Scale for Proliferation Facilities

Process	1 SQ	Process Time	Throughput [SQs/yr]	Plant Size
Mining & milling	10 t U	3–12 mo	1-4	10–40 t/yr
Conversion	10 t U	3–12 mo	1-4	10–40 t/yr
Enrichment from Nat-U	$25 \text{ kg}^{235} \text{U}$	3–12 mo	1-4	4800–19,000 kg-SWU/yr ^a 1200–5000 kg-SWU/yr ^b
Enrichment from LEU	$25 \text{ kg}^{235} \text{U}$	3–12 mo	1-4	1200–5000 kg-SWU/yr ^b
Reactor irradiation	8 kg Pu	3–12 mo	1-4	15–120 MWt ^c
Reprocessing	8 kg Pu	1–3 mo	4–12	4.5–18 MtHM/yr feed d

Notes

Environmental sensing can also support arms-control and disarmament verification efforts. In these cases, the detection program is directed at states that already possess many hundreds or thousands of nuclear weapons. For these states, new weapons production at rates even ten times higher than the IAEA standard definition may still be below the threshold of strategic significance, and the detection goals can be relaxed. In this review, the IAEA's 1 SQ/yr detection goals will be used as a reference case, but readers should bear in mind that methods with an order of magnitude poorer sensitivity may still be useful in an arms-control context.

The distance at which a signal must be detected depends primarily on the cost of building and operating the detector—although the acceptable cost has not been rationally quantified. Very roughly, viable candidate signals must typically be sensible at many hundreds of kilometers to render detection economically feasible at a national or global scale. There is considerable flexibility in this estimate, however, as technology advances may proceed either (a) along the dimension of sensitivity gains, making possible a system with a few very long-range sensors (e.g., satellite reconnaissance), or (b) along the dimension of cost reduction, with many thousands of inexpensive sensors (e.g., weather stations). In addition to the cost-sensitivity trade-off, the target signal must also be resistant to countermeasures, such as attenuation, dilution, or camouflage. In practice, if countermeasures are inexpensive and extra sensitivity is expensive, the number of viable techniques may be rapidly constrained.

4. SIGNALS, SOURCE TERMS, AND DETECTION

4.1. Uranium Mining and Concentration

Although there are dedicated uranium mines, uranium is so abundant that it is often coextracted alongside other minerals, as occurs commercially at the Olympic Dam copper mine in Australia, and as was done to support a covert nuclear-weapons program at the Al Quaim phosphate mine in Iraq. Uranium is about 25 times more abundant than mercury and twice as abundant as tin; as a consequence nearly every country possesses some extractable resource. The periphery of sandstone aquifers, for example, can have a uranium lens of 400–3,000 ppm, and these are thought to contain roughly 20% of the

^a Assuming ideal cascades with 90% U-235 output and 0.3% tails. A kg-SWU, or kilogram-separative work unit, is a measure of separation work; see (Villani, 1979).

^b Assuming ideal cascades with 90% U-235 output, 4% feed and 0.7% tails

^c Assuming 15–30 MWt/SQ/yr at 300 irradiation days/year; 30 MWt is a central estimate

^d Assuming 2 MWd/kg burnup, corresponding to about 90% fissile Pu isotopes

planet's uranium, making them one of the world's largest and widely distributed sources (<u>IAEA 2009</u>). A typical gold mine has 60–300 ppm uranium, a typical phosphate mine has 100–400 ppm uranium, and granite contains 20–50 ppm uranium. This abundance of uranium makes detection of undeclared uranium mining especially challenging (<u>OECD NEA & IAEA 2012</u>, Kemp 2006).

IAEA targets from Table 2 imply a detection goal in the range of 10–40 metric tonnes of uranium (metal mass) per year. Neither the uranium itself nor its radioactive decay products such as ²²²Rn are sufficiently concentrated in ore to be detectable at significant distances against the natural environmental background. Secondary signatures generated by mining operations or the processing of ores appear more promising.

4.1.1. Hyperspectral images found them to be of some value in detecting status changes at declared uranium mines but of little use in detecting new mines (DigitalGlobe 2013, Leslie et al. 2002, Truong et al. 2003). Hyperspectral imaging, which relies on absorption spectra to determine surface chemistry, is of interest for its potential to discover new mining operations in host geologies that might normally be rich in uranium, which could then trigger an on-site inspection. The only open-source hyperspectral satellite in operation at present is Hyperion, with 220 spectral bands and poor 30-m resolution. Both parameters are limited by the signal-to-noise ratio of the optical system. Initial attempts to use Hyperion for rock classification were frustrated by variable insolation, atmospheric absorption, and scattering (Borstad et al. 2005). New hyperspectral sensors, geology classification algorithms, and the use of pseudoinvariant calibration targets promise improved performance.

Not all relevant host geologies have strong spectral signatures. For example, phosphates are a spectrally bland and widespread source of uranium (Borstad 2014, Lim et al. 2006). Hyperspectral imaging would also be ineffective for in situ leach mining, a technique where a uranium-rich solution is removed through wells and the geology is not exposed at the surface. It is notable that leach mining has become the most economical method for extracting uranium from low-quality deposits.

4.1.2. CHEMICAL DETECTION. After mining, uranium is leached from the ore, and the resulting liquor must be concentrated by removing other solved minerals, a process that is sometimes carried out with chemicals that could serve as a detectable signature. The vast majority of leach liquors are concentrated using solvent extraction or ion-exchange resins. In general, solvent extraction is more selective, continuous, and cost effective than ion exchange and is the preferred method for most mining operations. However, ion exchange can be preferable when there is the potential for the cost of liquid chemistry to become prohibitive, as can be the case when leach liquors contain high fractions of suspended solids that result in chemistry lost to emulsification, or when extracting uranium from extremely low-yielding leach liquor. Of the two, only the solvent extraction process is obviously amenable to detection by chemical effluents.

For solvent extraction, usually one of two types of chelating solvents is used. For anionic liquors—for example, when uranium is complexed with sulfate after sulfuric acid leach—secondary or higher amines are employed, most commonly tertiary amines such as trioctylamine (Crouse et al. 1955). The heavier the amine, the more efficient the extraction process, although long-chain versions have very low vapor pressure (10⁻⁵ to

10⁻⁶ torr at 25°C). If released to the environment, they tend to be stable against chemical and photo-oxidation (Chemicals Inspection and Testing Institute 1992).

For a cationic liquor, such as occurs for uranium extracted from marine-sediment phosphorite prevalent in the Middle East and North Africa (Wymer et al. 2013), dialkylphosphoric acids are used in combination with trialkylphosphates, dialkyl alkylphosphonates, trialkylphosphine oxides, or other neutral organophosphorus compounds (IAEA 1987, Walters et al. 2008). A particularly common extractant is di(2-ethylhexyl)phosphoric acid (DEHPA). These solvents are widely used in nonuranium mining as well; therefore, detection would necessarily require a secondary on-site inspection and, by extension, regular monitoring of known user facilities to ensure no new coextraction of uranium had begun. In general, organophosphorus compounds have similarly poor volatility to long-chained amines but have lower persistence as they are subject to photo- and bacterial decomposition. A table of these compounds, decomposition products, and calculated vapor pressures was published by Probasco et al. (2002).

The magnitude of the source term cannot be easily estimated because it depends strongly on the manner in which particles are entrained in off-gas, filtered, and released. Although the vapor pressures for all common extractants are low, mechanical contacting equipment such as bubblers results in fine aerosols. The physics of aerosol production associated with these chemicals has been extensively studied, but particular plant equipment and off-gas management systems ultimately determine the magnitude of the source term (Long 1978, p. 361ff.). Data from studies of typical off-gas rates have been released under the Freedom of Information Act as a result of this study, but not in time to be incorporated herein (Birnbaum et al. 2004).

There are a number of mechanisms for detecting chemical extractants in the atmosphere. Gas chromatography mass spectroscopy can detect long-chain amines at 40 pg/m³ in air (Huang et al. 2014). Infrared differential absorption lidar can detect higher alkenes at 40 μ g/m³ (Nabiev & Palkina 2014). There are some data to suggest high sensitivities might be obtained using laser photoacoustic spectroscopy (Van Neste et al. 2008). Newly released source-term data may provide better direction for developing new sensors.

A further issue is the potential for countermeasures. Chemistry can be removed from off-gas using scrubbers and filters. Techniques specific to the chemical families used have long been studied for safety in nuclear-fuel reprocessing (Long 1978). It is also possible to use ion-exchange resins instead of solvent-extraction chemistry, and because the ion-exchange process uses less specific chemistry, prospects for detection are lower. Finally, uranium liquors can be concentrated by selective precipitation. Although slower and more expensive, precipitation methods are standard when using alkaline leaching from carbonate host geologies (Taylor et al. 2004). Most precipitation processes use inorganic chemistries that are not specific to uranium processing. These chemistries, when combined with mining operations that lack both hyperspectral and chemical signatures, can enable a proliferator to escape detection.

4.2. Conversion

Conversion refers to the chemical processing of uranium concentrate into purified uranium hexafluoride (UF₆). This step is nearly always a precursor to enrichment, the

critical step in producing highly enriched uranium. Although there are ways to enrich uranium without conversion to UF₆, most commonly electromagnetic and atomic-vapor laser isotope separation, these processes are expensive and sufficiently difficult that they are unlikely to be successfully used.

Because UF₆ is produced almost exclusively as a precursor to enrichment, it is a highly specific signature. Its production does not involve especially exotic chemistry, consisting primarily of fluorination of uranium oxides to UF₄ by HF followed by conversion to UF₆, by either by reaction with O₂ or fluorination by F₂ at temperatures of 500°C, at which only the latter process is commercially used. The process employs higher-thanatmospheric pressures and high temperatures, causing HF, F₂, and UF₆ to leak to the environment. The first two have poor persistence and poor specificity for nuclear processing. In particular, HF forms structurally nonrigid donor acceptance complexes of $(HF)_m...(H_2O)_n$, with absorption frequencies that make it difficult to detect by optical methods. More specific is UF₆, which hydrolyzes on contact with water vapor to form uranyl fluoride (UO₂F₂), an environmentally stable aerosol (Kemp 2008b). Experimental studies have shown that UO₂F₂ particles exist in agglomerates of wide-ranging hydration. The U–F bond is a detectable indicator signifying manmade origin. The rate of fluorine disassociation is determined by the relative humidity of the environment, but measurements with nano-secondary ion mass spectrometry (nanoSIMS) confirm that some fluorine remains bound to uranium after months of environmental exposure (Kips & Kristo 2009, Kips et al. 2012).

Aerosol size depends strongly on release conditions, but it is most often in the range of 0.5–1.5 μm in diameter for routine-release type kinetics. Spill-type release kinetics tends to result in larger aerosols. Deposition velocities depend on size, terrain surface roughness, and turbidity. Absent experimental studies of long-range transport, a deposition velocity of 1 mm/s appears reasonable (Kemp 2008b). A survey of operating plants found median release rates ranging from 0.1 to 16 grams UF₆ released to the atmosphere per metric tonne UF₆ produced, corresponding to 1 to 160 g/SQ, with the variation being attributable to plant design (Kemp 2010b). These parameters suggest, very roughly, that parts-per-trillion sensitivity would be needed to detect a clandestine plant producing 1 SQ/yr at 300 km, with orders of magnitude variation around this number associated with variability in plant process design, atmospheric dispersion, deposition uncertainties, and analytical losses. At a 300-km detection range, thousands of inexpensive air-sampling systems would be needed for global coverage.

The primary challenge in establishing the feasibility of UO₂F₂ detection is the sensitivity of the analytical technique used to detect U–F bonds. Because aerosols transported great distances will necessarily be diluted by other solids, including other uranium- and fluorine-bearing compounds, it is necessary to carry out a particle-level search. Two basic approaches are available: (*a*) aerosol collection followed by laboratory analysis and (*b*) real-time detection by optical methods.

In laboratory analysis, aerosol cakes can be collected on filter paper and then disaggregated using ultrasonic vibration (Wang et al. 2013). Identification of uranium-bearing particles from background is efficiently carried out using neutron-induced fission-track analysis. Each of these must be subsequently analyzed for fluorine content, traditionally done with thermal ionization mass spectrometry (TIMS) or SIMS. The process requires many manual operations and is time consuming in its present form,

suggesting that research into an automated method may be worthwhile. Although not yet demonstrated in an analytical context, laser-induced fluorescence has been used to detect uranyl (UO₂²⁺) ions on surfaces at 100 ng/m² and in solutions at 100 ng/L (Namestnikov 2007). The technique might reduce the analytical burdens and might also be useful in surveying vegetation or soil contamination using unmanned aerial vehicles.

Real-time probing of the atmosphere for trace species has long been studied using a variety of techniques, namely differential absorption (DIAL) or coherent anti-Stokes Raman scattering (CARS) (Nabiev & Palkina 2014). These methods show some promise but currently lack the required sensitivity. Laser photoacoustic spectroscopy currently has sensitivities in the parts-per-billion to parts-per-trillion range, and researchers claim room for substantial improvement (Nabiev & Palkina 2014, Yönak & Dowling 2003). Cavity ring-down spectroscopy may also offer significant sensitivity. One of the difficulties in using optical methods is that the hydration state of UO₂F₂ changes its spectral response. Additionally, spectroscopy data are scarce and sometimes contradictory. High-resolution, systematic studies of the spectroscopic responses of oxyfluoride particles against other atmospheric chemical species would substantially improve assessments of UO₂F₂ as a possible indicator.

With current technology, routine monitoring for UO_2F_2 aerosols would require a considerable effort, yet countermeasures are easily implemented. For example, high-efficiency particulate air (HEPA) filters would remove 99.97% of aerosols at the most difficult-to-filter diameter of 0.3 μ m (US DOE 2005). Properly used, such filtration would reduce the signal by four to five orders of magnitude. Countries that have legitimate conversion plants could also hide a small, covert plant near a legitimate plant such that the signal from the covert plant was lost in the effluent noise of the legitimate plant. These simple countermeasures, and the asymmetry of effort required for detection versus hiding, raise doubts as to the wisdom of relying on the detection of UO_2F_2 absent some new, sensitive, and very inexpensive detector.

4.3. Isotope Separation

The separation of fissile ²³⁵U from natural uranium can be effected by a number of methods. Gaseous diffusion was the method of choice from 1944 until the mid-1970s, but the process required large facilities and considerable electrical power, about 1.5 MW/SQ/yr (Whitaker 2005). The high rate of energy consumption made it possible to detect and monitor diffusion plants by their thermal pollution. The United States successfully monitored Soviet plants using this method throughout the Cold War (CIA 1970, Kemp 2014).

4.3.1. CENTRIFUGE. Starting in the mid-1970s, gaseous diffusion was replaced by the gas centrifuge, a more efficient process that uses 0.11–0.17 MW/SQ/yr for first-generation centrifuges; advanced centrifuges use about half as much. Because these plants are also more compact, at 1,000–5,000 m²/SQ/yr for first- and second-generation centrifuges, a small plant could have a footprint similar to that of an average office building but consume less energy per square meter, such that it is nondisticnt when analyzed by thermal methods (US EIA 2006).

Centrifuge plants produce very little chemical effluent. The only chemical routinely used is UF₆, and nearly all process lines operate below atmospheric pressure to prevent UF₆ from solidifying. Monitoring at British centrifuge plants showed a typical UF₆

release rate of about 50 mg/SQ ($\underline{\text{Kemp 2010b}}$). When released, the UF₆ hydrolyzes into UO₂F₂, as it does for conversion plants; however, the specific release rate is a thousand times smaller than that of a conversion plant. Because conversion is a necessary precursor to enrichment, chemical detection efforts are better focused on conversation plants.

Centrifuges also produce electromagnetic emanations, both free-space radiation and artifacts induced on powerlines by power supplies. The free-space signal from the centrifuge motors can be estimated by treating the motors as dipole radiators emitting at distinct frequencies in the range of 350–1,500 Hz, overlapping with wideband ultra-low-frequency emissions from geoelectric, magnetospheric, and radio-atmospheric phenomena (Habib 2007). The wavelengths of these signals imply a near-field treatment with intensity decaying as distance cubed for distances up to a few tens of kilometers. Beyond that, the Earth-ionosphere waveguide intercedes, effectively channeling any residual signal. However, simple calculations of the magnetic field with highly optimistic assumptions indicate that emanations are likely to reach undetectable levels between a few meters and few kilometers. Furthermore, the self-cancelling nature of three-phase power, which is used for most centrifuge motors, and the skin-depth attenuation by typical aluminum motor housing, would probably attenuate these signals to levels that are undetectable at distance for even the world's most sensitive magnetometers (Savukov et al. 2005).

Power supply noise induced on electrical lines is a well-known phenomenon that factories such as textile plants must address to maintain line-power quality. The artifacts are generated at twice the frequency of various AC stages in the power supply as the alternating voltage momentarily drops below the bias threshold of semiconductor components. This creates step-function artifacts in the current draw, called line notches, which result in wideband back-EMF noise on the line (Jarc & Schieman 1985). Propagation of this noise can be modeled using the telegrapher's equations (Krejci et al. 2014). A calculation shows that characteristic frequencies in the Fourier decomposition may travel along typical power lines for many hundreds of miles if not filtered by a power substation. However, monitoring power lines presents several difficulties. First, the signal is specific not to centrifuge plants but rather to switching power supplies that are commonly used in manufacturing industries. Second, monitoring requires direct access to power lines by the monitoring agency, and a state-run clandestine program may be aware of the risk and take steps to prevent monitoring. Third, the energy consumption of a proliferation-scale centrifuge plant is small enough that it can be easily operated with a diesel or natural-gas backup generator, removing power lines from consideration altogether.

Some very exotic methods of detection have also been explored. For example, the rotating mass of the centrifuge rotors generates a Lense-Thirring gravitational disturbance (Ciufolini & Wheeler 1995). The author calculates, for a typical first-generation centrifuge plant, the induced precession around the vector normal to Earth's surface in a gyroscope located 100 m from the plant would be $\sim 10^{-28}$ rad/s, with the effect falling off as the distance cubed. Superfluid helium gyroscopes can plausibly measure to the level of $\sim 10^{-18}$ rad/s in daylong measurements (Packard & Vitale 1992). Although these gyroscopes are sensitive enough to detect the Lense-Thirring effect of Earth at $\sim 10^{-13}$ rad/s, they would not be able to detect a centrifuge plant. The effect could also be eliminated by spinning half the centrifuges in the opposite direction.

In sum, there is as yet no obvious signal for long-range detection of a covert centrifuge plant. The prevalence of centrifuges in proliferation activities suggests their properties merit further study. For example, high-frequency free-space electromagnetic signals generated by electronics rather than motors and ideas such as infrasound beat frequencies have not been systematically explored.

4.3.2. LASER ENRICHMENT. Although not yet commercially deployed, uranium isotopes can be separated by selectively exciting one isotope by laser irradiation. Methods rely either on atomic-metal vapor [atomic vapor laser isotope separation (AVLIS or SILVA)] or on vibrational transitions of molecular UF₆ [collectively termed molecular laser isotope separation (MLIS)]. The MLIS category is subdivided into methods that decompose UF₆ into UF₅ (called MOLIS) and those that do not (SILEX and CRISLA).

As no laser enrichment facilities exist, estimation of source terms is necessarily speculative. Proponents of laser enrichment claim lower energy consumption and a smaller footprint than centrifuges (Jensen et al. 1982). As centrifuges already consume electricity at a rate that is too low to be detected, this implies that thermal detection of laser isotope separation would also be infeasible. The chemical effluent is more difficult to estimate. Batch processes, such as AVLIS or MOLIS, require process chemistry between enrichment stages, potentially leading to source terms that are larger than those for centrifuge plants but that probably have an upper bound similar to that of conversion plants, with similar chemistry considerations and caveats. SILEX and CRISLA do not require process chemistry, such that their chemical effluent is likely to be lower, perhaps approaching that of a centrifuge plant. Electromagnetic emanations are also difficult to predict. Some processes rely on high-powered pulsed lasers (at least at present). These may produce electromagnetic signals from capacitive discharge, but the characteristics have not been studied. To date, statements about the detectability of laser enrichment facilities (Grossman 2010) appear to be based on the potential for intelligence agencies to monitor the purchase of exotic optical components used in some 16-µm-based MLIS systems. No environmentally sensible source terms are known.

4.3.3. CHEMICAL ISOTOPE SEPARATION. Chemical isotope separation is a process similar to uranium concentration, utilizing either solvent extraction (as in the French CHEMEX process) or a chromatographic process based on ion-exchange resins (as in the Japanese Asahi process). Historically, these methods were not economically competitive. They were also unattractive for proliferators because the equilibrium times needed to reach weapon-usable enrichments were measured in months to years and special provisions had to be made to avoid criticality problems at high enrichments (Fuji et al. 1989). However, advances in ion-exchange resins have led to thousand-fold improvements in speed, and under certain organizational circumstances, chemical exchange may be appealing.

The French CHEMEX process consumed energy at 0.17 MW/SQ/yr (Villani 1984), a rate similar to that of first-generation centrifuges but with lower energy density owing to the larger plant size. Substantial reductions came about after research in Japan led to high-efficiency ion-exchange resins, resulting in smaller plants and energy consumption in the range of 0.06–1.0 MW/SQ/yr, similar to that of modern centrifuge plants (Maomi et al. 1981). With further improvements aided by computational chemistry, chemical-separation methods may become even more efficient in the future.

Because no chemical enrichment plants exist, there are no firm data for emissions. However, there is a sound argument for assuming they might release less process chemistry than a uranium concentration plants: For solvent extraction methods, the economic value of highly tuned process chemistry would likely be higher, suggesting a cleaner, more controlled process with lower effluent rates. In contrast, the resin-based ion-exchange methods that have showed the greatest improvement in recent years could have mundane chemistries that are poor signatures of the process. Apart from chemistry and energy consumption, there are no other features of the technique that readily suggest a strong signature. Although chemical exchange is proliferation resistant in that it could take many months or years to begin production of highly enriched uranium, the system could be resistant to detection during this time.

4.4. Transmutation

The alternative to separating fissile material from natural uranium is to create fissile isotopes by transmutation. The isotopes of interest are ²³⁹Pu and ²³³U, both produced when a fertile isotope captures a neutron and undergoes beta decay. About 33 mol of either ²³⁹Pu or ²³³U is required per SQ, requiring an equivalent number of neutrons to be generated, a process that is most readily done in a nuclear reactor but can also be carried out with particle accelerators. A detection effort could target physical processes that occur during transmutation—such as heat generation, cogeneration of exotic isotopic gases, and antineutrino emissions—or could focus on the chemical processes used to purify the final fissile material.

4.4.1. REACTORS. Nuclear reactors produce neutrons by controlled fission chains. Between two and three neutrons are produced per fission, one of which must be consumed to sustain the chain reaction. The remaining neutrons are statistically distributed between leakage, nonproductive absorption in various materials, and the creation of fissile material. Typical light-water reactors produce 0.6–0.8 fissile atoms per fission; specially designed breeder reactors have achieved conversion ratios of 1.2 fissile atoms per fission, and simulations suggest values up to 1.8 may be possible.

About 200 MeV of energy is released per fission event, which for realistic reactors corresponds to 15–35 MWt/SQ/yr (where MWt stands for megawatts thermal). Heat generation at this rate is rare and requires special cooling arrangements, which makes nuclear reactors especially detectable by photoreconnaissance monitoring of snowmelt or infrared imaging of heat plumes. However, it is possible to hide the heat that comes from a reactor. For example, Sweden had plans to use cold, deep lake water to cool its reactors, with the output temperature being the same as surface water. Alternatively, one could camouflage the heat signature against the background of other high-heat processes, such as an aluminum smelting plant.

Reactors generate detectable radioactive noble gases: ⁴¹Ar (half-life 109 min) is produced by neutron reactions in the reactor core, and ⁸⁵Kr (10.8 yr), ^{131m}Xe (11.9 days), ¹³³Xe (5.2 days), ^{133m}Xe (2.2 days), and ¹³⁵Xe (9 h) are fission products. Of these, the largest releases during routine operation are of ¹³³Xe, although measuring multiple isotopes could be useful in screening out other sources of radioxenon (Kalinowski & Pistner 2006). Detection of these gases, along with atmospheric transport backprojection, could be used to locate a clandestine reactor or reprocessing facility (Collon et al. 2004). Sensitivity is limited by atmospheric backgrounds, which are created by the accumulated xenon from other nuclear reactors, medical treatment facilities, and, especially, medical-isotope production facilities (Igarashi et al. 2000). For regions in the

Northern Hemisphere not directly downwind of a primary source, the average background concentration of ¹³³Xe is approximately 1 mBq/m³. The Comprehensive Test Ban Treaty's International Monitoring System (IMS)—designed to detect nuclear-weapon detonations by radioxenon—will have 40 monitoring stations capable of radioactive noble gas detection, with the option to extend this number to 80. These stations have been designed to be sensitive at 1 mBq/m³ with a 24-h sampling period (Kalinowski & Tuma 2009).

A survey of 3,670 emission records from 111 reactor sites between 1995 and 2005 showed the estimated release per nuclear reactor is highly variable. The median ¹³³Xe continuous release rate was 1.6 ($\sigma = 3.4$) MBg/day/MWt, assuming 85% capacity factor and 33% thermal-to-electric efficiency (Kalinowski & Tuma 2009). At 15–35 MWt/SQ/yr, these rates suggest emissions for a 1-SQ/yr reactor would reach background levels in tens of kilometers under average (Pasquill class D) atmospheric conditions. Order-of-magnitude sensitivity improvements over IMS levels and careful characterization of background fluctuations would be necessary to detect small operations at distances of hundreds of kilometers. The investment needed would be similar to that for UO₂F₂ detection of conversion facilities, putting the economics of the concept into question. Although countermeasures for noble gases are more difficult, they are nonetheless possible. A cryogenic scrubbing system was demonstrated at the Idaho Reactor Testing Station in 1968 (Bendixsen & Offutt 1969), a proposal for trapping noble gases in zeolites was tested in Japan (Fukumatsu et al. 1998), and efficient trapping of noble gases by organic cage molecules has recently been demonstrated at the lab scale (Chen et al. 2014).

A third alternative would be to monitor radiation directly, of which the only significantly penetrating kind is the antineutrino. Antineutrinos are detectable by a range of reactions, the most significant of which is inverse beta decay (IBD) of the electron flavor, $\bar{v}_e + p \rightarrow n + e^+$, which has an energy threshold of 1.8 MeV. Of the ~6 antineutrinos produced per fission, about 1.5, or $5 \times 10^{16} \, \bar{v} / \text{s} / \text{MWt}$, are above the 1.8-MeV IBD detection threshold (Bemporad et al. 2002), although below 2.6 MeV there is a significant background from uranium and thorium decay in Earth. In January 2003, the KamLAND experiment reported the detection of antineutrinos from all reactors in Japan with a mean distance of 180 km, demonstrating the feasibility of long-range detection (Eguchi et al. 2003). Based on this data, the detection rate for 1.8-MeV antineutrinos can be approximated as:

$$0.832 \,\overline{v}_{e} / \operatorname{day} \times (P/1 \,\mathrm{MWt}) \times (1,000 \,\mathrm{km}/D)^{2} \times (M/1 \,\mathrm{Gt}) \times F(E,D/E),$$

where P is the reactor thermal power (15–35 MWt/SQ/yr), D is the detector distance from the reactor, M is the detector mass if the detection medium is water, and F is a term that accounts for neutrino oscillations with a value between 0.5 and unity and that depends on energy and distance (Learned 2004). The reactor neutrino spectrum evolves as the ratio of plutonium-to-uranium fission rate increases through the fuel cycle (Kopeikin 2012), which also makes it possible to discern diversion of fuel from legitimate reactors if one can achieve adequate count rates.

As long as the direction of the antineutrino is not measured, the signal from any given reactor must be set against an otherwise irreducible and dominant background generated by all other reactors. The geolocation of a new, clandestine reactor would depend on equipotential surfaces from numerous detectors. For the current global installed reactor capacity of ~880 GWt (gigawatts thermal), this background varies with position on Earth from about 2,500 to 300,000 counts per megaton detector per year (counting only >2.6 MeV in order to rule out terrestrial background, and assuming a 0.4-km depth). The highest counts are in western Europe, the eastern seaboard of the United States, China, and Japan—regions where nuclear power is heavily used (Lasserre et al. 2010). To detect a 1-SQ/yr clandestine reactor (30 MWt) with 1σ error, located 1,000 km from the detector and in the Southern Hemisphere, where the antineutrino background is low, would take a 300-Mt detector 1 yr, assuming a constant 500-count/yr/Mt background. The same detection scenario but for a reactor located in the Middle East would require 3 Gt detectors, given the region's higher, 10,000-count/yr/Mt background.

Conversely, if the background from other reactors can be suppressed via directionality, detector sizes could be reduced to megaton scales for the same standoff and integration time. However, only crude and stochastic antineutrino directionality for IBD events has been achieved, requiring tens or hundreds of detected events to reconstruct an average direction. Due to the low counting statistics in the clandestine-reactor search, suppression of reactor antineutrino backgrounds would require event-by-event directional reconstruction, which has not been demonstrated with IBD detectors.

Large detector volume pose major engineering challenges. Detectors 50 kt in size have been built, and megaton detectors have been proposed, but at present the technology to build 100 Mt and larger detectors on land platforms does not exist and appears to be prohibitively expensive (Bahcall et al. 2001). Light collection also remains an economic challenge. At current prices, photomultiplier tubes alone would cost on the order 10–100 billion US dollars for a 1-Gt spherical detector; an inexpensive light collection technology would provide an important enabling development (Learned 2004). Silicon photomultipliers built from arrays of avalanche photodiodes might offer one such possibility.

The appeal of antineutrino detection is the inability to prevent the signal from escaping the reactor, but the method is not immune to countermeasures. The fission rate in a commercial power reactor is continuously shimmed to compensate for declining fuel reactivity and variability caused by the ingrowth and burnout of neutron-absorbing fission products, but the process cannot be controlled to better than about ±0.5%, or ±15 MWt for a full-size ~3-GWt reactor, and neutrino flux varies by more than this amount during operation because of isotopic shifts. This implies uncertainties on the order of 15 MWt per commercial reactor, which is approximately the same size of a clandestine proliferation reactor. Although these uncertainties should average out over the yearlong integration, they open up a cheating strategy in which a power reactor's output power is systematically underreported, or its fission-spectrum modified, to eclipse a small, nearby clandestine reactor. Another cheating strategy would be to produce fissile material in a commercial reactor directly. Fissile-material production rates can be enhanced above normal levels without increasing the fission rate or antineutrino flux of the reactor, for example, by cladding some fuel elements with a cadmium foil. Elaborate IAEA

Safeguards could search for these particular countermeasures to reduce the chances of successful cheating, but at such effort that the appeal of antineutrino monitoring declines.

4.4.2. ACCELERATORS. The same set of signatures available for the long-range detection of reactors applies to transmutation driven by accelerators, but with shifts in the strengths of various signals. Accelerators depend on reactions other than fission chains to produce the neutrons needed for transmutation. With low-energy accelerators (below several tens of millielectronvolts), the most efficient reaction is $^7\text{Li}(p,n)$, but yields are typically <0.1 n/p (neutrons per proton) and concurrently produce considerable amounts of tritium. More practical is to use protons with energies in the range of 150 MeV to 1 GeV for spallation of thick depleted or natural uranium (^{238}U) targets. These yield $0.0367 \times (E_p - 120 \text{ MeV})/(1 \text{ MeV})$ n/p, where E_p is the incident proton energy. The neutrons produced can be efficiently captured by fertile material by slowly thermalizing them in lead or a liquid-salt target (Kemp 2005).

Thermal signatures from accelerator-driven transmutation will generally be higher than those from reactors unless proton-beam energies exceed 400 MeV. An ambitious setup that is still plausible given current technology would be a high-field superconducting isochronous cyclotron yielding 3 mA of protons at 1 GeV (Antaya 2009). Lower-energy versions of this same accelerator concept have demonstrated 33% plug-to-beam efficiency. The 1-GeV version would be capable of producing 1 SQ/yr at about 10 MW thermal power output, about one-third that of a reactor. However, accelerators with such high energies and beam currents will require considerable engineering to bring into existence, especially with regard to radiation damage to structural materials. A more realistic scenario based on an existing design would be the 250-MeV MIT Megatron (Hartwig et al. 2011), but at 250 MeV, the system requires 16 times more power to achieve the same neutron yield, rendering it considerably more detectable than the equivalent nuclear reactor.

High-energy spallation produces a variety of fission fragments, including radioactive noble gases that could serve as signatures of the process. The rate at which these are produced depends on the incident beam energy and cross-section, and data are generally scarce. Fission product yields are, however, available for 1-GeV proton beams on ²³⁸U targets, and the long-lived radioactive noble gas yields are reported in Table 3 (Bernas et al. 2003, Ricciardi et al. 2006).

Table 3: Spallation noble gas production from 1GeV protons on a ²³⁸U target

Isotope	half life	nuclei/proton [1GeV on ²³⁸ U]	nuclei/neutron [1GeV on ²³⁸ U]	production rate (MBq/d)/(SQ/yr)
³⁹ Ar	369 yr	0.00038	1.2×10 ⁻⁵	41
⁴² Ar	32.9 yr	0.00017	5.3×10 ⁻⁶	200
⁸⁵ Kr	10.8 yr	0.00690	0.00022	25,000
¹³³ Xe	5.25 d	0.00370	0.00012	9.9×10^{6}
¹³⁵ Xe	9.14 h	0.00320	0.00010	1.2×10^{8}

For spallation sources yielding >2 n/p, the rate of radioactive noble gas production (internal to the target) per SQ is only 0.1-2% of that produced in a thermal fission reactor. However, despite lower production, the amount released by an accelerator to the

environment could be greater because the containment and structural overburden of a fission reactor is very effective in preventing the release of gases to the environment. One study of a liquid salt (FLiBe) spallation target found that 1% of noble gases produced escaped from the target into the expansion cavity after \sim 15 h of irradiation and 15 h of cooling (Thiolliere et al. 2011). If a liquid target saturates over many months of irradiation, the escape fraction could be higher; conversely, if the target is a solid that swells from gas production, the escape fraction might be lower. At \sim 0.01% escape, the ultimate release rate to the environment would be a thousand times larger than for the proliferation equivalent pressurized water reactor. Although release rates will ultimately depend strongly on spallation target design, they might serve as a viable detection signature, although for a rather unlikely route. That said, as with reactors, the potential for scrubbing noble gasses from the off-gas by cryodistillation or zeolitic adsorption remains.

4.4.3. REPROCESSING. Reprocessing is the chemical separation of fissile material from irradiated nuclear fuel or fertile targets and is the final step in the recovery of fissile materials produced by transmutation. Although commercial reprocessing involves large, expensive facilities, some of which are identifiable in structure, a small, makeshift operation using standard industrial supplies is feasible (Ferguson & Culler 1977, US GAO 1978). Such a plant could be constructed to have no visual signatures that would reveal its location by overhead imaging, be built in several months, and once operational produce weapon quantities of fissile material in several days.

Isolation of fissile material is done by dissolving fuel or targets followed by precipitation or solvent extraction of the fissile isotopes (Long 1978). The oldest and simplest process involves coprecipitation of Pu(IV) with BiPO₄ while U(VI) remains in solution with SO_4^2 . Precipitated Pu is then oxidized to Pu(VI) by BiO₃, or other oxidizers. The process involves no organic chemistry to contribute to external signatures. The bismuth phosphate process was replaced by the REDOX solvent-extraction process, in which Pu(VI) and U(VI) are extracted into hexone. The addition of Fe(II) reduces Pu. Hexone is a widely used organic solvent for paints and varnish, with 300,000 t produced annually as of 1996 (OECD 2011). The REDOX process was replaced with the plutonium uranium redox extraction (PUREX) process, which is the most widely used process today and has very forgiving chemistry. PUREX is based on tri-n-butyl phosphate (TBP) diluted in an aliphatic hydrocarbon such as dodecane. TBP is less common than hexone, with annual production at 3,000–5,000 Mt/yr, and purchases of TBP are reportedly monitored for signatures of proliferation activity. The primary use of TBP is as a hydraulic fluid for aircraft and as an extractant for rare-earth mining, but it is also used as a defoamer for cement, coatings, and floor finishes; as a carrier for fluorescent dyes and inks; and as a solvent for gums and glues. TBP is chemically sticky and easily adsorbs onto surfaces, making it easy to filter. It degrades rapidly in the environment on exposure to sunlight (OECD 2004a). Finally, it is possible to do reprocessing using some of the same chemistry as is used in uranium mining. Both dialkyl and trialkyl phosphoric acids have been used, the former in the DIAMEX-SANEX process and the latter in Chinese and German pilot plants (Berthon & Charbonnel 2009).

Radiolysis of organic extractants and dilutants creates a vast slew of chemicals of varying degrees of volatility, stability, and uniqueness. These may include isomers

unique to radiation environments and radiolytic products that are fairly exotic by themselves (Berthon & Charbonnel 2009). An example is dibutyl phosphate, produced commercially at only 250 t/yr (1996 comparison year) (OECD 2004b), but far more exotic species are also produced. Even if effluents are individually nonspecific, such as acidic gases, certain mixtures of effluents may be specific for reprocessing. A catalog of chemical variants has been published (Probasco et al. 2002), and a study of source terms is now publicly available (Birnbaum et al. 2004). Considerable research will be needed to discern which of the many hundreds of species are reliable and detectable signatures.

A final detection strategy is to search for volatile radioisotope fission products that are released to the environment when fuel is dissolved at the start of reprocessing. Of these, the most interesting is ⁸⁵Kr. As a noble gas, it evolves rapidly from solutions and typically in concentrations much larger than those found leaking from nuclear power plants. Its 10.77-yr half-life means that substantial quantities may be present even decades after the fuel was irradiated. Detection of ⁸⁵Kr is complicated by a substantial, persistent, and varying background produced by commercial reprocessing plants (Igarashi et al. 2000, Ross 2010). A study based on 348 weekly samples of releases from the Tokai reprocessing plant in Japan, and a detector located 55 km away, found that the probability of detection at 55 km fell rapidly to zero for release rates below 2 SQ/yr (Kemp 2008a). More careful characterization of the background, multiple sensors, and continuous atmospheric transport modeling have the potential to improve detectability, although a systematic study of this potential remains to be carried out. As with reactor noble gas emissions, filtering of noble gases from reprocessing plants remains a viable countermeasure.

5. CONCLUSIONS

The foregoing assessments indicate that, although detection modes may exist for many pathways, countermeasures can usually be found to hide small programs. For this reason, a tight detection web able to discover any pursuit of a nuclear weapon appears to be infeasible for the time being. Moreover, the asymmetry between detection and countermeasure costs suggests that a large investment in a global network of detectors might not be justified. Existing detection technology may nonetheless be attractive in a limited intelligence-gathering capacity targeted at specific states or regions of concern.

With further innovation, the cost-benefit calculation may shift in favor of technical detection. Of note is the potential for improvement in spectroscopic methods for in situ monitoring of trace gases. One could imagine many thousands of very inexpensive, autonomous optical sensors with sensitivity in the parts-per-billion to parts-per-trillion range able to detect either chemical or isotopic signatures at medium to long ranges. These sensors would be especially applicable to detecting reprocessing, but possibly also to detecting clandestine reactors, accelerators, uranium conversion plants, and even uranium mining. Detailed spectroscopic data and systematic studies of release effluents are needed to identify the most promising research directions. By contrast, more exotic physics-based methods focused on electromagnetic, gravitomagnetic, or antineutrino signals appear to be impractical despite their innate appeal. Although several such methods—such as high-frequency electromagnetic signals and infrasound—remain unexplored, there is little reason to believe they hold much promise.

Although there are prospects for improvement, the limits of environmental sensing must be considered in a political context. All the technical detection methods described here suffer from a flaw in that if a method of detecting a signal can be devised, a way to reduce, eclipse, or confuse that signal is probably also possible, and with less effort than it takes to detect the signal in the first place. If that imbalance holds, then in the race between hiding and finding nuclear programs, the hiders will have the advantage. For nonproliferation, this suggests that with the passage of time technical detection will tend to be of less and less use. For arms control, the prospects for using technical verification without cooperation to stabilize a world free of nuclear weapons becomes unlikely, and long-term confidence in the stability of arms-control agreements would therefore need to rely on traditional intelligence or a reduction in the perceived value of nuclear weapons. These futures, however, are several decades away, and environmental sensing might usefully buttress existing nonproliferation frameworks and provide confidence-building measures for interim agreements.

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