Detectable Emissions

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Every industrial process releases some trace of the materials involved. With modern pollution control equipment, releases usually can be kept well below regulatory standards for protection of human health and the environment. Even with the most rigorous controls, however, some gases, liquids, and solid particles escape to the environment. High pressure fluids may seep past pump or valve seals. Every time solid materials are moved, tiny particles are produced that pass through the finest ventilation filters. During moments of carelessness or equipment failure, gross releases may occur.

Processes to produce nuclear materials are no exception. No matter which route is selected to obtain fissile nuclear material, some traces of materials used in the process will be released. Some of these materials are unique to the production of nuclear weapons, while others are indicative of nuclear activities in general. Some are not suspicious by themselves, but would provide a warning signal if detected as part of a pattern of releases or in conjunction with auxiliary data such as from export controls.

This chapter reviews the steps that must be followed by a nation clandestinely producing nuclear material, and identifies the signatures, or potentially detectable indications, that might be detected via environmental monitoring. There are two basic routes to produce fissile material for nuclear weapons: enrichment of uranium to obtain highly enriched uranium (HEU); and irradiation of uranium-238 in a nuclear reactor to convert it to plutonium, which must then be separated from the remaining uranium and by-products in a reprocessing plant. These are diagrammed in figure 2-1. Both approaches are feasible (both were pioneered in the Manhattan Project) and present approximately equal difficulty overall.[1] Iraq considered both routes prior to 1991 but chose enrichment as its primary focus.

URANIUM ENRICHMENT

Virtually all uranium occurring naturally in the world consists of the same isotopes: 99.3 percent

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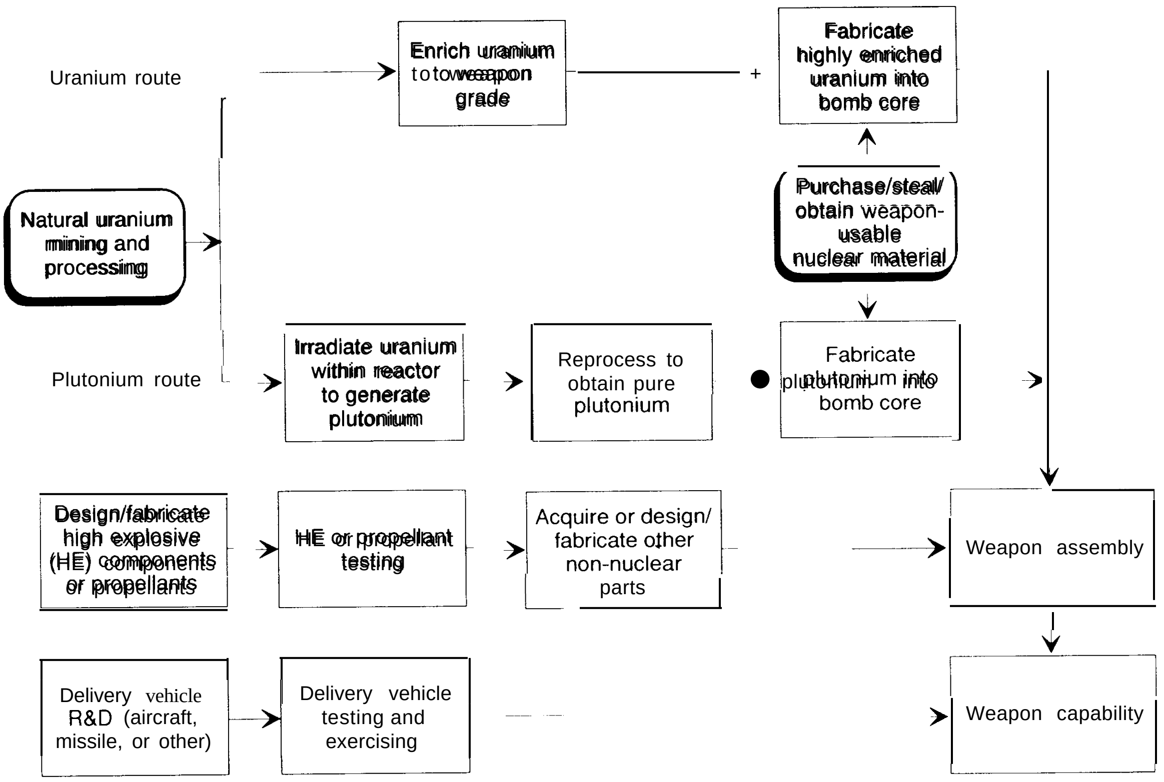
[1] For further information on the two approaches, see U.S. Congress, Office of Technology Assessment, *Technologies Underlying Weapons of Mass Destruction*, OTA-BP-ISC-115 (Washington, DC: U.S. Government Printing Office, December 1993).

FIGURE 2-1: Technical Routes to a Nuclear Weapon Capability



SOURCE Office of Technology Assessment, 1993

U^{238}, 0.7 percent U^{235}, and a trace of U^{234}.[2] To be useful as fuel in a conventional light water reactor (LWR), the level of U^{235} must be raised to about 3 percent, which is known as low enriched uranium (LEU). Weapons require HEU, which is at least 20 percent U^{235} and preferably much higher. Commercial enrichment plants producing LEU currently use either gaseous diffusion or centrifuge technology.[3] Either technology can also be used to produce HEU, but a plant designed to produce LEU would have to be reconfigured, at least in part, to produce HEU.

The process of enrichment is difficult because U^{235} and U^{238} are chemically identical and only slightly different in weight. Basically the process consists of preferentially removing U^{238} so that the end product has a higher fraction of U^{235}. However, current technologies cannot economically achieve a clean separation, so the waste stream (called tails) of depleted uranium still contains a significant amount of U^{235}. Commercial enrichment plants typically produce tails containing about 0.3 percent U^{235}, instead of the original 0.7 percent. Calutrons can achieve greater separation and might produce tails of 0.2 percent or even less.

Uranium for the Hiroshima bomb was enriched using calutrons, a form of electromagnetic isotope separation (EMIS). This is a relatively simple but expensive and inefficient

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[2] A minor exception is the uranium ore found in Gabon, which had undergone a slow chain reaction over a billion years ago, depleting some of the U-235.

[3] Another technology, advanced vortex tube, was used in a South African commercial enrichment plant that was shut down in March 1995.

technique that the United States quickly replaced with gaseous diffusion methods. However, Iraq successfully constructed equipment similar to calutrons and produced a small quantity of HEU. Lasers have also been used for enrichment, but those technologies (atomic vapor laser isotope separation—AVLIS, and molecular laser isotope separation—MLIS) have not yet progressed outside the laboratory. Several other technologies have been considered, including aerodynamic methods such as the Becker nozzle.

An NPT signatory intent on building an HEU nuclear weapon could, if it had one, convert a commercial diffusion or centrifuge plant from LEU to HEU production. It is very unlikely that the entire plant could be converted covertly, so the country would have to abrogate its safeguards agreements. Alternatively, it could try to evade safeguards by converting only part of the plant to HEU, hoping that such actions would escape detection, or it could build an undeclared facility using any of the technologies which it could master.

Both diffusion and centrifuge plants are designed with large numbers of individual units. In a diffusion plant, each unit slightly increases the enrichment of a large process stream. Many diffusion stages are required—about 1000 to produce LEU and maybe 3000 are required for HEU, so the stages are linked in a cascade. In a centrifuge plant, each unit achieves a higher level of enrichment but can handle less material. Many units are connected in parallel to form a stage, but fewer stages are required than in a diffusion plant (fewer than 20 for LEU, about 60 for HEU). For either diffusion or centrifuge, each unit (and the entire plant) has two exit streams: enriched uranium and depleted uranium. The enriched stream proceeds through a series of enrichment levels until the desired level is attained. The depleted stream from each stage (which has slightly less U^{235} than when it entered the stage) drops back one or more stages and is re-enriched, until the desired level of the tails is reached.

A commercial enrichment plant is a highly complex facility that must operate in a carefully prescribed manner. Conventional safeguards are designed to detect if the facility had been reconfigured to produce HEU. However, it is conceivable that a small portion of the cascade could be isolated and dedicated to the production of HEU, particularly if it used LEU as feed material.[4] Safeguards based on materials accountancy would have to be very thorough to detect this. More worrisome, a centrifuge plant could be temporarily reconfigured to produce HEU and then converted back to LEU between inspections.[5] To forestall such a conversion and reconversion, safeguarded centrifuge enrichment plants are subject to a certain number of unannounced IAEA inspections per year. Alternatively, a proliferator might build a new facility close by in order to reduce costs by sharing technical, infrastructure, and administrative support. Materials accountancy would not detect this facility, if it did not feed from or supply any safeguarded facilities.

Natural uranium is ubiquitous, so its detection does not, per se, signify any unusual activity. Any discovery of uranium with isotopes in other than natural proportions (or in chemical form different from natural uranium) is a sure indication of nuclear activity. Emissions from the enrichment process can occur at many places. Natural uranium must be converted to a gaseous form, usually uranium hexaflouride (UF_6). UF_6 is a colorless solid at room temperature, but becomes a gas at temperatures above 134 degrees F at atmospheric pressure. In itself, UF_6 with any isotope of uranium is an indicator, albeit a secondary one, of enrichment because no other processes are known to involve it. Within the enrichment process itself, small quantities of uranium may

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[4] Producing LEU requires over half the separative work (enrichment effort) of producing HEU. Thus starting with LEU instead of natural uranium more than doubles the capacity of the HEU cascade.

[5] This could not be done with a diffusion plant which takes much longer to reach equilibrium, leaving the action open to detection.

escape from anywhere in the cascade, at whatever enrichment level the specific piece of equipment happens to be operating. As the released UF_6 reacts with water vapor in the air, it precipitates out and can migrate from the process area as airborne particles that are deposited outside the plant. UF_6 also reacts chemically with the air to form UO_2F_2. These particles can show the full range of enrichment, from depleted uranium to the maximum enrichment attained. Thus the detection of HEU at an LEU plant is strong evidence that at some time the plant was operated in a HEU mode.

For use as fuel or as weapons material, the UF_6 must be re-converted to metallic uranium following enrichment. This process provides additional opportunities for the release of emissions.

Two other isotopes of uranium are also important—U^{234} and U^{236}. As noted above, U^{234} is a trace constituent of natural uranium, but the fraction is variable, unlike the other natural isotopes. Most uranium contains about 52-54 parts per million (PPM) of U^{234}, but some ores contain several PPM more.[6] U^{234} provides two important pieces of information. It can be used as a tracer to determine the origin of the uranium ore. It also can indicate the type of enrichment used. This is because some enrichment technologies (EMIS and lasers) distinguish between U^{234} and U^{235}, while others pass them through together.

U^{236} appears only in uranium that has been irradiated. It is produced when an atom of U^{235} absorbs a neutron and fails to fission. When spent fuel is removed from a reactor, it may still have more U^{235} than does natural uranium. The fuel can be reprocessed to recover the valuable U^{235}, which must be re-enriched before it can be recycled. The U^{236} will remain with the U^{235} during these processes. Detecting U^{236} at an enrichment plant is proof that the facility has handled reprocessed uranium, and therefore it's likely that plutonium was separated from spent fuel. U^{236} can remain in an enrichment plant for many years after it was introduced, contaminating subsequent loads of natural uranium.

Most large, commercial enrichment plants can be detected through their emissions. If a proliferator wishes to remain covert, emissions can be reduced to the point where they are significantly harder to detect. A small, carefully designed, constructed and maintained plant producing only enough HEU for one or two bombs per year, if equipped with a ventilation system using high-efficiency filters, could be quite difficult to detect.

In addition to isotopically altered uranium, an enrichment plant may emit several other types of signals that could be detected. Gaseous diffusion, aerodynamic, and electromagnetic separation plants are quite inefficient and release a large amount of heat. This might be detected by satellite observation or perhaps measurement of the temperature increase of a river if cooling water is dumped there. Centrifuge plants are much more energy efficient, but they place unusual loads on the electric power system. In particular, the centrifuges operate at high speed and require conversion of the line frequency to much higher frequency. The converters reflect a distinct signal back into the line that can be detected. Finally, under some conditions, the distinct noise generated by centrifuges might be detected and recognized.

PLUTONIUM PRODUCTION AND REPROCESSING

An NPT proliferator has two main choices in producing plutonium for weapons:[7] reprocess spent fuel from its own power reactors, or build a covert production reactor. A country could divert

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[6] U^{238} decays to thorium (Th^{234}), which in turn decays to U^{234}. Intermediary products can have different chemistry than uranium, and therefore may not remain in proportion to the original U^{238}.

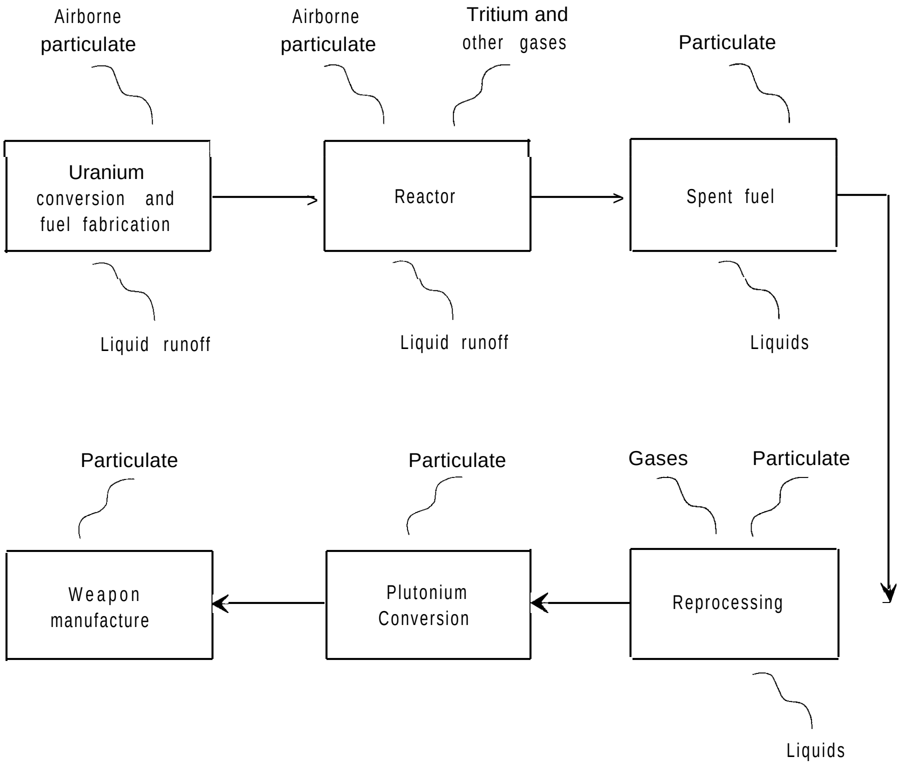
[7] A third choice, buying or stealing plutonium, either from the commercial nuclear power fuel cycle (if and when plutonium becomes a routine part of the cycle) or from a nuclear weapons state, is not considered here because the role of environmental monitoring would be peripheral.

safeguarded spent fuel only fit is willing to conspicuously violate safeguards agreements. The reprocessing could be attempted at a commercial reprocessing plant with the intention to divert the ensuing plutonium. For the foreseeable future, however, no potential proliferator is likely to operate a commercial reprocessing plant because of restrictions by supplier countries and poor economics. If the reprocessing plant were safeguarded, the diversion would have to be covert, risking detection. A variation to the approach would be to construct a small, covert reprocessing plant which could extract plutonium from the spent fuel. However, diversion of spent fuel from a safeguarded reactor runs a high risk of detection by current safeguards procedures.

The second approach probably would involve a research-type reactor, not a power reactor. This fuel would also have to be reprocessed, presumably at a covert reprocessing facility.

However it is done, each step releases emissions that can contribute to detection of the activities. Figure 2-2 shows the major points of emissions where environmental monitoring can play a role. Understanding the signatures from the activities required to produce nuclear weapons is critical to finding and identifying them. The IAEA currently is documenting signatures from all activities.[8] The United States has made considerable effort in this area.

FIGURE 2-2: Major Points of Emissions



SOURCE: Office of Technology Assessment, 1995

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[8] Personal communication with IAEA staff, Mar. 31, 1995.

Reactors

Producing plutonium is technically simpler than enriching uranium,[9] but more steps are involved. Uranium must be fabricated into fuel which is irradiated in a reactor. Plutonium results when an atom of U^{238} absorbs a neutron and, through a decay process, is transmuted to Pu^{239}. All plutonium isotopes are fissile (fission when struck by a neutron) but, instead of splitting, some atoms of Pu^{239} absorb a neutron and become Pu^{240}. This process can continue to produce Pu^{241} and heavier isotopes. The longer the fuel is left in the reactor, the more plutonium is created, and the more is converted to the heavier isotopes.

LWR fuel is in the form of enriched UO_2, pelletized and encased in metal tubes (usually a zirconium alloy, but stainless steel has also been used). LWR fuel technology has been mastered by many countries and some potential proliferators could also be expected to produce adequate fuel. This is a plausible route under some conditions, such as if a country were to abrogate its safeguards agreement and keep the reactor operating with indigenously produced fuel. This approach is not very plausible if the proliferator attempts to remain covert. Furthermore, normal power cycles produce reactor grade plutonium (with a high content of Pu^{240} and heavier isotopes relative to Pu^{239}). Reactor grade plutonium can be used to make an effective nuclear bomb, but it is distinctly inferior to weapon-grade plutonium (which has a low fraction of Pu^{240}).[10] Weapon-grade plutonium can be produced in an LWR, but the reactor must be shut down frequently and the fuel removed and replaced. The lack of power generation during shutdowns is visible and expensive, adding significantly to the cost of the weapon program.

Heavy-water-moderated reactors, such as the CANDU reactor produced in Canada, would be easier to use as a source of plutonium. The CANDU uses unenriched uranium fuel, which would be far easier for most countries to produce. It also is refueled continually instead of being shut down. This provides two advantages to the proliferant: some fuel elements can be exposed only briefly, yielding weapon-grade plutonium; and power is not lost during frequent refueling as in an LWR. However, there are relatively few heavy-water reactors in the world, most of them in Canada. India has several also.

Small, plutonium-production reactors could be built covertly by many countries. Such a reactor could be moderated by graphite or heavy water (if these can be obtained without triggering investigation) and operated with natural uranium, which would eliminate the need for enriched fuel, greatly simplifying the fuel cycle. The fuel itself is also easier to manufacture since it is irradiated under less demanding conditions than in a light water reactor, allowing the use of cladding such as aluminum. A non-power reactor which operates at a thermal output of about 30 MW could produce enough weapon-grade plutonium for 1 or 2 weapons per year.[11] Larger reactors are also possible. The Hanford B Reactor, a very large, graphite-moderated reactor that uses natural uranium, could be a model.[12]

Reactor operation produces a wide range of isotopes. There are three types: fission products; activation products (when an atom of non-nuclear material such as steel in reactor components absorbs a neutron); and actinides (an atom of uranium absorbs a neutron to produce plutonium and higher elements). Some of the isotopes formed in these ways are naturally occurring,

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[9] Designing and building a plutonium bomb is more difficult than producing a uranium “gun-type” weapon. Thus the two routes are comparable in overall difficulty.

[10] Use of reactor-grade plutonium in weapons has a significant probability of substantially reducing the weapon yield. Furthermore, reactor-grade plutonium generates significantly more heat from radioactive decay than does weapon-grade plutonium, complicating weapon design. See U.S. Congress, Office of Technology Assessment, *Technologies Underlying Weapons of Mass Destruction*, OTA-BP-ISC-115 (Washington, DC: U.S. Government Printing Office, December 1993), p. 133.

[11] U.S. Congress, Office of Technology Assessment, *Technologies Underlying Weapons of Mass Destruction*, op.cit., p. 138.

[12] Personal communication with Ned A. Wogman, Pacific Northwest Laboratory, Aug. 17, 1995.

stable atoms which, not being peculiar to nuclear activities, do not provide unique information. Others are highly radioactive and decay so rapidly that they are unlikely to be detected in the environment in sufficient quantity to be a useful signal.

The isotopes that are useful for detection of covert nuclear activities are those that:

a. are produced in reasonable quantity;
b. are not natural;
c. do not decay too rapidly to be detected;
d. have chemical properties favorable for transport and collection;
e. are easily identified, especially through characteristic decay radiation;
f. can be distinguished from those widely distributed by weapons tests or reactor accidents, especially Chernobyl.

Table 2-1 lists the radioactive isotopes that meet these requirements. The exact emissions from any given site would depend on the specific technology chosen and the systems and care applied to minimize them.

Emissions from reactors generally are small. In conventional power reactors, the fuel is sealed inside tubes which in turn are inside the pressure vessel. Leakage of fission products and actinides occurs only if the tubes leak (an increasingly rare occurrence as the technology improves) into the cooling water. From the cooling water, these and other radioactive products must escape past high pressure barriers. In a boiling water reactor, the cooling water directly powers the turbine, providing additional opportunities for emissions. Contamination is routinely removed from the water to maintain its purity. Degasifiers, ion exchange units and other systems are used. These are likely to be the source of most emissions, particularly tritium and the noble gases such as argon and krypton. Solid and liquid matter is generally collected inside sealed systems and should not escape in significant quantities.

Small plutonium production reactors would not need the same barriers. Fuel might be encased in a simple metal jacket not designed to withstand great pressure, and the coolant might be air. Gaseous products are likely to be released, but the level of radionuclides is much lower than in a power reactor.

Reactor operations are more likely to be discovered when something goes wrong. Even a minor upset, such as a thermal excursion that is reversed before any damage occurs, stresses the reactor and may result in short-term emissions.

A different type of signature associated with reactors is the heat they generate, which usually is dissipated to the air or a waterway. Even a small reactor capable of producing only 8 kg of plutonium per year releases about 30 MW of heat. This level can be detected by infrared devices on high-flying aircraft or satellites even if the heat causes a temperature rise of only a few degrees above the ambient. A small reactor could be hidden in an industrial area or near a thermal power plant, which would make the heat emissions less conspicuous.[13] However, the signature would still be useful information.

Whatever kind of reactor is used, the fuel will probably be stored, following exposure, to allow the short-lived fission products to decay. Power reactor fuel is stored in a spent fuel pool because the level of decay heat production requires efficient heat removal. Fuel from a small production reactor could be stored in air. Emissions could occur at this stage also. Liquid emissions could occur from a storage pool because the water must be circulated and cooled. Storage could eliminate gross emissions of short half-life products (e.g., iodine-131, xenon-133) from reprocessing.

Reprocessing

Reprocessing of the irradiated reactor fuel is far more likely to produce telltale emissions than operating a reactor. Typical reprocessing involves chopping up the fuel rods, dissolving

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[13] Anthony Fainberg, “Strengthening IAEA Safeguards: Lessons from Iraq,” Center for International Security and Arms Control, Stanford University, April 1993.

TABLE 2-1: Isotopes Indicative of Reactor Operation

FISSION PRODUCTS

Mass	Element	Half-life	Radiations (energies in Mev)
85	Krypton (Kr)	4.5 hours	0.151 γ (75%), 0.305 γ (14%); 0.8 β
85	Krypton	10.8 years (21%)	0.514 γ (.4%); 0.7 β
88	Krypton	2.8 hours	0.196 γ (26%), 0.835 γ (13%), 0.898 γ (14%), 1.530 γ (11%), 1.836 γ (21%), 2.196 γ (13%), 2.392 γ (35%); 2.8-5.0 β
93	Zirconium (Zr)	1.5 million years	no detectable emissions
95	Zirconium	64 days	0.724 γ (45%), 0.757 γ (55%) Nb cascade: 0.766 γ (100%)
95	Niobium (Nb)	35 days	0.766 γ (100%)
99	Technetium (Tc)	210,000 years	no detectable emissions
103	Ruthenium (Ru)	39 days	0.497 γ (86%)
105	Ruthenium	4.4 hours	0.316 γ (11%), 0.676 γ (16%), 0.724 γ (48%); 1.2 β
106	Ruthenium	374 days	0.512 γ (19%), 0.622 γ (10%), 1.050 γ (.9%); 3.5 β
129	Iodine (I)	16 million years	no detectable emissions
131	Iodine	8.0 days	0.364 γ (81%), 0.637 γ (7%)
132	Tellurium (Te)	3.04 days	0.228 γ (88%), I cascade (below)
132	Iodine (I)	2.3 hours	0.523 (16%), 0.668 γ (99%), 0.773 γ (76%), 0.955 γ (18%); 1-2 β
133	Iodine	20.8 hours	0.530 γ (86%); 1.3 β
133	Xenon (Xe)	5.2 days	0.081 γ (37%)
135	Iodine	6.6 hours	0.527 γ (14%), 0.547 γ (7%), 0.837 γ (7%), 1.132 γ (23%), 1.260 γ (29%), 1.458 γ (9%), 1.678 γ (10%), 1.791 γ (8%); 1.3 β, Xe cascade: 0.250 γ (90%)
135	Xenon	9.1 hours	0.250 γ (90%); 0.9 β
135	Cesium (Cs)	2.3 million years	no detectable emissions
137	Cesium	30.1 years	0.662 γ (85%)
140	Barium (Ba)	12.8 days	0.537 γ (24%); 1.0 β, La cascade (below)
140	Lanthanum (La)	1.7 days	0.329 γ (19%), 0.487 γ (43%), 0.816 γ (22%), 1.596 γ (96%); 1-2 β
144	Cesium	285 days	0.134 γ (11%), 0.696 γ (1.3%), 1.489 γ (.3%), 2.186 γ (.7%); 3.0 β

ACTIVATION PRODUCTS

Mass	Element	Half-life	Radiations
3	Hydrogen (H)	12.3 years	0.019 β
14	Carbon (C)	5730 years	0.15 β
24	Sodium (Na)	15 hours	1.369 γ (100%), 2.754 γ (100%); 1.4 β
56	Manganese (Mn)	2.58 hours	0.847 γ (99%), 1.811 γ (27%), 2.113 γ (14%); 2.8 β
59	Iron (Fe)	44.5 days	1.099 γ (56%), 1.292 γ (43%); 1.5 β
60	Cobalt (Co)	5.3 years	1.173 γ (100%), 1.332 γ (100%)
63	Nickel (Ni)	100 years	0.07 β
64	Copper (Cu)	12.7 hours	0.6 β (40%), 0.6 β+ (20%)

NOTE:

a. Isotopes with half-lives of less than 2 hours were excluded because they are likely to decay before they can be detected. The only isotopes included with half-lives less than 100 days are krypton, ruthenium, iodine, and xenon, which are transported rapidly through the environment, and those which emit strong gamma rays for easy detection.

b. Isotopes with half-lives of more than 100 million years were excluded because they occur naturally.

SOURCE: Office of Technology Assessment, 1995 the fuel in acid, separating and purifying the plutonium using solvent-extraction, precipitation, and ion-exchange, and converting the plutonium to a metallic form. The PUREX process, the most common method, uses well-known chemical processes.

When the fuel is chopped up and dissolved in an acid bath (usually nitric acid), all gaseous products (e.g., krypton, argon) are released. Some of them are hard to trap. Therefore this stage can provide strong evidence of covert activity. The acid bath can also lead to other emissions. The acid itself can fume or leak and be a chemical indicator. Traces of uranium and plutonium as well as other products are likely to be contained. Other chemicals used in the process such as tributyl phosphate (TBP) also may be released.[14] Waste products from the purification process can produce airborne particles or liquid runoff. Cooling water also could carry out various products.

Based on emissions from fuel reprocessing at Sellafield (United Kingdom) in 1991, a small (8 kg of plutonium/year), emission-controlled reprocessing plant is likely to release annually:

12 mg carbon-14 split between air and water;

125 g iodine-129 (for old fuel) to off-site water;

15 g technetium-99 to off-site water;

2 mg strontium-90 split between air and water.[15]

These are small quantities which are then spread over a wide area as the releases disperse. While the concentrations appear to be minute, ultrasensitive equipment such as the accelerator mass spectrometer and processes such as neutron activation analysis (see chapter 3) could detect them in environmental samples.

Final purification and conversion of plutonium to metallic form is likely to produce particles. Machining of the material to produce the weapons component will add more particles to an effluent stream.

Countermeasures

Any country trying to produce plutonium covertly will try to limit key emissions to the greatest extent possible. Ventilation from the process rooms can be filtered with high efficiency filters that remove almost all particles. Even ordinary pollution control equipment can be effective without triggering any export control notice.[16] Liquids can be held within the plant (at least until the volume becomes unmanageable). Gases can be trapped. Even the noble gases can be adsorbed on activated charcoal or removed cryogenically and isolated, although these methods are difficult and not 100 percent effective.[17]

Such measures will reduce emissions, greatly complicating the detection of undeclared facilities and activities. However, they will not eliminate the risk. In addition, on-site storage increases the possibility of major accidental releases, for example if a storage vessel ruptures. Such releases may be easier to detect than continual small emissions.

Under some conditions, a proliferator might even deliberately release contamination to confuse inspectors. This might slow down efforts to locate the key sites, but it also increases the likelihood that a major search will be mounted.

The possibility of countermeasures suggests two things: development of ever more sensitive instruments may be essential; and baseline analyses of suspect sites should be made as soon as possible. The latter, particularly for complicated facilities that already have released some contamination, may make it possible to detect any changes in activity.

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[14] Richard R. Paternoster, *Nuclear Weapon Proliferation Indicators and Observables,* LA-12430-MS, Los Alamos National Laboratory, December 1992.

[15] Briefing notes supplied by Ivan Proctor, Lawrence Livermore National Laboratory, March 1, 1995.

[16] David A. Kay, “Denial and Deception Practices of WMD Proliferators: Iraq and Beyond”, *The Washington Quarterly,* 18:1, Winter 1995.

[17] Anthony Fainberg, op.cit. p. 30.