Environmental Monitoring for Nuclear Safeguards

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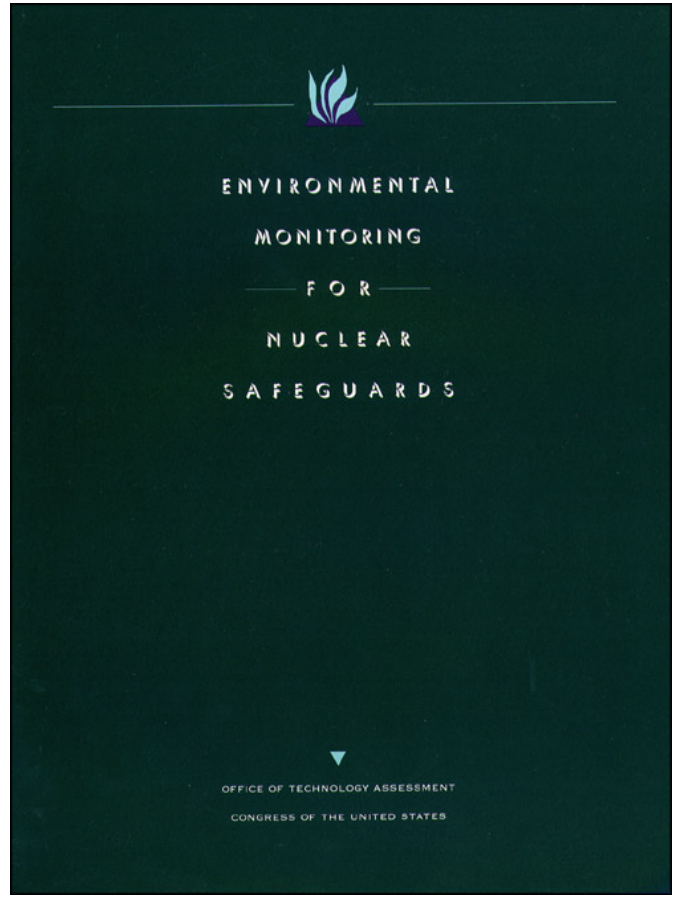

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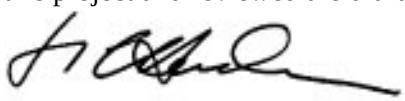
Foreword

In June 1995, the Office of Technology Assessment published the report *Nuclear Safeguards and the International Atomic Energy Agency*, the sixth in OTA's series of publications on the proliferation of weapons of mass destruction. That report found that the International Atomic Energy Agency's traditional mission of detecting the misuse of known nuclear materials and facilities addressed only part—and probably not the most important part—of the proliferation problem. To assure that states are not violating their Non-Proliferation Treaty commitments, the International Atomic Energy Agency (IAEA) must also verify that states do not possess *covert* nuclear facilities—a mission that prior to the 1991 Gulf War, it had neither the political backing nor the resources to conduct. In the June report, OTA concluded that providing the IAEA with the resources, the information, and the political support it needs to look for such sites may turn out to be the most important aspect of a reinvigorated safeguards regime.

The IAEA recognizes the importance of this new mission and is in the process of assuming it. One of the tools it is exploring to provide some indication of the presence of secret, or undeclared, nuclear activities and facilities is *environmental monitoring*. Modern sampling and analysis technologies provide powerful tools to detect the presence of characteristic substances that are likely to be emitted by such illicit activities. This background paper examines the prospects for such technologies to improve nuclear safeguards. It concludes that environmental monitoring can greatly increase the ability to detect undeclared activity at declared, or known, sites, and it can significantly increase the chances of detecting and locating undeclared sites.

Completed in the last month of the Office of Technology Assessment's existence, this paper will be the last OTA publication related to the proliferation of weapons of mass destruction. OTA appreciates the invaluable advice and assistance of the people who contributed to this project and reviewed the draft material.



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Introduction and Summary 1

Environmental monitoring is a potentially powerful supplement to current safeguards techniques intended to prevent the spread of nuclear weapons. Prior to the 1991 Persian Gulf War, the International Atomic Energy Agency (IAEA) administered safeguards primarily on the nuclear materials associated with known commercial or research facilities. Accounting for this material would provide notice were a proliferator to divert any to obtain the necessary nuclear fissile material for nuclear weapons.[1] Events in Iraq revealed after that war have demonstrated that such a safeguards approach addresses only part of the problem. Probably more important to halting proliferation is ensuring that countries do not violate their non-proliferation agreements by constructing covert facilities for nuclear material production. *Environmental monitoring,* which tests for the presence of materials that are likely to be emitted by such activities, can help inspectors detect undeclared activities at safeguarded sites and may be able to detect covert facilities at undeclared sites.

Environmental monitoring was first used by the IAEA in Iraq following the Gulf War. The agreement ending the war included the right for the United Nations to inspect all Iraqi nuclear facilities (declared and suspected) to determine if any nuclear-weapon related activities had occurred. The IAEA has a mandate from the United Nations Security Council to perform these inspections. In the month between the end of the war and the start of the inspections, Iraq removed much of the most incriminating equipment, such as the calutrons used for enrichment, and concocted stories to explain the remainder. Inspectors took samples of materials within and near facilities, and swipes of dust that had collected on the surfaces of equipment. These were analyzed at various laboratories, including in the United States. These analyses played a key part in demolishing Iraq's cover stories and exposing its nuclear weapon program, which included uranium enrichment and plutonium experimentation. The program had not been detected in earlier inspections despite the co-location of pro-

[1] For a review of the history of nuclear safeguards and the International Atomic Energy Agency that administers them, see: U.S. Congress, Office of Technology Assessment, *Nuclear Safeguards and the International Atomic Energy Agency*, OTA-ISS-615 (Washington, DC: U.S. Government Printing Office, June 1995).

totype facilities for the production of weapons materials with civilian, safeguarded facilities.

Subsequently, the IAEA used similar techniques in South Africa to provide additional assurance that all nuclear materials produced for its voluntarily terminated weapon program were fully accounted for. These techniques were also used to check the North Korean declaration of facilities and activities under the Non-Proliferation Treaty (NPT). The results from the application of these techniques, together with other information accumulated by inspection teams, led the IAEA to conclude that there were inconsistencies between the plutonium identified in North Korea's initial report and the reprocessing activities actually carried out.

The IAEA is now completing a series of field trials in cooperating member states to determine the efficacy of the techniques in a broad range of applications, and the Agency proposes to make environmental monitoring an integral part of the inspection process for safeguarding peaceful nuclear installations. The intent is to provide additional assurance that a country is not engaged in undeclared nuclear activity. Through this and other proposals, the IAEA is assuming a much more activist role in searching out any efforts to produce weapons.

This report analyzes how environmental monitoring works and what can be expected of it as part of safeguards. Chapter 2 reviews the emissions that can be expected from nuclear facilities that are supporting a weapons program. Chapter 3 looks at the techniques used for detecting those emissions, including sampling in the field and laboratory analysis. Chapter 4 reviews IAEA activities to implement environmental monitoring. Finally, chapter 5 looks at technologies now in the laboratory that could improve the effectiveness of environmental monitoring in the future. The remainder of this chapter provides a summary of the report and additional background.

MAJOR FINDINGS

- Use of environmental monitoring can significantly increase the ability of safeguards to detect undeclared nuclear activities at declared sites.
- Environmental monitoring is not a panacea and must be used in conjunction with other non-proliferation tools. However, some relaxation of conventional safeguards may be warranted as the new techniques are implemented in a broader, more integrated scheme.
- Technologies under development can significantly increase the chances of detecting and locating undeclared sites.
- Costs to the IAEA will be modest as long as efforts are focused on sampling in and around declared sites. Wide-area monitoring of the atmosphere to detect undeclared facilities would be very expensive. Wide-area monitoring of waterways appears more practical, but its application must be further investigated.
- The support of the United States and other member nations is essential to make the new techniques work. In addition to financial support to get the program moving, laboratory analysis, training of inspectors and IAEA laboratory staff, and research and development are needed.

NUCLEAR MATERIALS AND SAFEGUARDS

Many nations have the capability to develop nuclear weapons, though some would require considerable investment in facilities and manpower. The most difficult part of producing nuclear weapons is obtaining the fissile material (unless it can be stolen).[2]

[2] For a discussion of the technical requirements for making nuclear weapons, see U.S. Congress, Office of Technology Assessment, *Technologies Underlying Weapons of Mass Destruction*, OTA-BP-ISS-115 (Washington, DC: U.S. Government Printing Office, December 1993), chapter 4 ("Technical Aspects of Nuclear Proliferation") and especially appendix 4-A ("Components, Effects, and Design of Nuclear Weapons").

Nuclear explosives can be based on uranium or plutonium. Natural uranium can be found everywhere, but in order to be used in a weapon, it must be enriched. Natural uranium consists of three isotopes, uranium-238 (99.3 percent), uranium-235 (0.7 percent), and a trace amount of uranium-234. Only the uranium-235 can support a chain reaction, meaning not only that its nuclei can release energy by *fissioning*, or splitting, when struck by a neutron, but also that each fission releases sufficient neutrons to continue the process. Enrichment, an expensive and technologically difficult task, increases the concentration of uranium-235.[3] Fuel for light water power reactors must be enriched to about 3 to 5 percent uranium-235. A nuclear explosion cannot be achieved with less than 20 percent enrichment. For an effective weapon, the uranium-235 content must be much higher.

Alternatively, a proliferant can use plutonium, which is produced by irradiating uranium-238 in a nuclear reactor. Spent fuel from a conventional power reactor contains plutonium, but using it presents several difficulties (especially for the covert proliferator). Every country known to have produced a plutonium explosive has chosen to build a reactor whose primary task is the production of plutonium that is optimized for use in weapons.[4] In addition to a reactor, the proliferator needs reprocessing capability to extract plutonium from the irradiated uranium-238.

The NPT came into force in 1970 to provide a mechanism for nations to gain access to peaceful nuclear technology without giving rise to suspicions that they were using their facilities to produce weapon-related materials. Safeguards were instituted to check on their compliance as part of the treaty. Signatories to the NPT that had not tested a nuclear weapon before January 1, 1967 (i.e., all except the United States, the U.S.S.R. [now Russia], Britain, France, and China), pledged not to acquire nuclear weapons or to build nuclear facilities with unsafeguarded nuclear material. Only Iraq is known to have violated its commitments on a significant scale, but North Korea is not in compliance with its safeguards agreement with the IAEA because it refuses to accept certain activities identified by the Agency as necessary to assess the completeness of its declaration.

Most nations have signed the NPT. Exceptions include Israel, Pakistan, and India. Nonmembers are not legally bound to refrain from developing nuclear weapons or to accept safeguards on their facilities. However, essentially all nuclear exporters require all nuclear materials and critical nuclear-related equipment purchased by other countries to be placed under safeguards. Thus only indigenously developed facilities (and a few pre-NPT exports) are not safeguarded. Safeguards by themselves cannot stop proliferation. They only provide warning that a nation is not complying with its agreements. It is up to the United Nations and the international community to take action in response.

When a nation becomes party to the NPT, it is required to conclude a safeguards agreement with the IAEA, to declare all its nuclear materials, and to establish a system of controls for them. When the safeguards are implemented, each of the country's nuclear facilities must be specified (declared) in an attachment to the agreement. However, prior to the Gulf War, the IAEA did not verify the completeness of this declaration. Nor could it inspect undeclared facilities of non-members. It was never the IAEA's expectation that its efforts would deter all weapon programs, but it assumed that national intelligence programs would uncover covert efforts.[5] Iraq, North Korea, and South

[3] Uranium enrichment technologies are discussed in ibid., appendix 4-B ("Enrichment Technologies").

[4] For discussion of the use of "reactor-grade plutonium" and "weapon-grade plutonium" for nuclear weapons, see ibid., pp. 131–134.

[5] Bruno Pellaud, "Safeguards in transition: Status, challenges, and opportunities," IAEA Bulletin, vol. 36, No.3, 1994, Vienna, Austria, pp. 2–7.

Africa[6], three very different cases, showed that these assumptions could not be depended on in all situations.

There are several aspects to safeguards. The most prominent is materials accountancy, in which the total inventory of nuclear material in a country is monitored to ensure that none is diverted to weapon purposes. Besides measuring material inventories and material flows, inspectors check facility operating records to see if everything is consistent. In addition, there may be perimeter monitoring devices and cameras around critical areas to detect any undeclared removal of nuclear material.[7]

Consider the situation of an NPT signatory (or a country that has agreed to equivalent safeguards in order to import equipment) that decides to obtain nuclear weapons. It may have power reactors or research reactors, all of which would be under safeguards. Either might be used to produce fissile material, but the risk of detection under current safeguards is high. The country could abrogate its safeguards agreements and prevent IAEA inspectors from discovering that these facilities were being used for weapon purposes, but that would almost certainly end that nation's ability to import nuclear fuel and equipment, and would probably precipitate international sanctions. A large commercial reprocessing plant (or enrichment plant) is far harder to safeguard effectively than a reactor used for research or for generating power, but no nation suspected of entertaining thoughts about proliferating is operating one under full-time safeguards.[8]

Therefore a potential proliferator is likely to favor a small reactor/reprocessing plant or an enrichment plant dedicated to the production of weapon materials even if it has a power reactor. IAEA inspectors do not typically have access to facilities not on their list, even to buildings right next door to ones they inspect regularly.[9] In fact, from the perspective of a proliferant state, co-locating legitimate and illegitimate facilities has several advantages. The peaceful facilities can provide some camouflage for the illicit activities, some personnel may work on both, and they can share utilities, security arrangements, and other functions.

To strengthen its safeguards system, including the ability to detect undeclared nuclear activities, the IAEA initiated Programme 93+2 in 1993. Environmental monitoring is a centerpiece of this effort, including field trials that are now being completed. The IAEA's Board of Governors endorsed the general direction of Programme 93+2 in its meeting March 27-30, 1995, and requested the Secretariat to prepare specific proposals for implementation.[10]

THE POTENTIAL OF ENVIRONMENTAL MONITORING

Modern analytical technology has made environmental monitoring an effective tool. A series of instruments has been developed that can identify extremely small traces of materials. Uranium and plutonium can be detected and identified by iso-

[6] The Iraqi program is discussed in chapter 3. North Korea apparently built its facilities prior to signing the NPT but has refused to declare and accept safeguards on all of them even though the IAEA discovered the others. South Africa dismantled its weapon program prior to signing the NPT. Since the NPT applies only to its members, neither it nor the IAEA can be faulted for the behavior of non-members. It is important to remember that the NPT, even though the primary international non-proliferation mechanism, is not capable of addressing the entire problem.

[7] The safeguards process is discussed in greater detail in U.S. Congress, Office of Technology Assessment, *Nuclear Safeguards and the International Atomic Energy Agency*, op. cit., footnote 1.

[8] India, not an NPT party, operates reprocessing facilities that are under safeguards only when reprocessing safeguarded spent fuel.

[9] The IAEA does have the authority to request so-called special inspections of any site if such inspections are necessary for the agency to fulfill its safeguards responsibilities. However, requesting a special inspection must be firmly based on information that the agency is unable to meet its safeguards obligations without access to specific named locations. This would be an extraordinary act that would not typically be done in the course of a routine safeguards inspection.

[10] IAEA Press Release, "IAEA Board of Governors Holds Spring Meeting, Considers Nuclear Safety Issues and Strengthening of Safeguards Verification System," Mar. 31, 1995.

tope in less than nanogram quantities (a nanogram is one billionth of a gram; there are about 28 grams to the ounce). Particles as small as a micron (one millionth of a meter or about 0.000039 inches; the diameter of the period at the end of this sentence is about 350 microns) can be analyzed. Many instruments are sufficiently sensitive to detect the fallout of plutonium from above-ground nuclear bomb tests, almost all of which occurred more than thirty years ago.[11]

No industrial process can prevent minute traces of materials from escaping. Even the most sophisticated filtration systems can only reduce, not eliminate, releases. In particular, enrichment plants release traces of enriched and depleted uranium, including highly enriched uranium (HEU) for weapons if it is being produced. It is easy to distinguish isotopically altered uranium from natural uranium, and its presence is an indicator of enrichment activity (but not necessarily near where it is found). A detection of HEU where only natural or low enriched uranium should be is a clear warning signal that activities that could contribute to a weapons program are underway. Reprocessing plants release many fission products and other products as well as uranium and plutonium. Plutonium is entirely manmade, so its discovery in any significant quantity (i.e., at levels above those expected to be found from known atmospheric nuclear tests or other contamination) or with an isotopic composition inconsistent with a State's declaration is also a warning signal. Emissions are discussed in chapter 2.

These releases can be readily detected at levels that are far below those that pose hazards to human health. Tiny particles may settle out within process buildings or float out and be carried by the wind, sometimes for very long distances. Wherever they settle—on plants, in the soil, in waterways—they may be detected. Gaseous releases can be carried even further away, but dilution and varying wind patterns can make them harder to detect.

The first step in environmental monitoring is sampling: wiping surfaces in the facility with a cloth or tissue, or collecting leaves or other parts of plants, digging up soil, scooping up water, and other means of collecting material that has settled outside the buildings. Sampling is not particularly difficult in most cases, but the inspectors need training as to where to take the samples and how to avoid cross-contamination (from one sample to another, e.g., a trowel used to dig soil samples must be cleaned between each use, or particles from the first could give a false reading in the next sample).

The samples are then sent to a laboratory for analysis. Two general types of analysis are used—bulk and particle. Bulk analysis looks at the entire sample or a significant part of it. Analysis involves the application of many instruments such as mass spectrometers, which can separate isotopes of an element by their masses, allowing measurement of the relative abundance of the isotopes. Other instruments measure the emissions of radioactive decay to determine the radioisotope. Particle analysis selects individual particles, usually from the surface wipes, by examination under a microscope. Once isolated, the particle can be individually analyzed, using many of the same techniques. Particle analysis is more sensitive than bulk analysis because individual particle analysis can yield information on the precise formation of the particle, while bulk analysis averages the particles together. However, particle analysis also is significantly more expensive.

The IAEA's field trials involved sample collection at known nuclear facilities in various countries to explore the best ways to take samples and to determine the kind of information that could be gleaned from the analyzed samples. Facilities examined included enrichment plants,

[11] The United States, the Soviet Union, and the United Kingdom ceased testing nuclear weapons above ground when they signed the Limited Test-Ban Treaty in 1963. France continued atmospheric testing, at a level far below the pre-test-ban U.S. and Soviet rates, until 1975. China detonated the world's last above-ground nuclear test in 1980.

reactors, reprocessing plants, and research complexes. These tests have been completed, but public release of the results is contingent on the approval of the host country. Preliminary indications are that the field trials were generally successful. In most cases, the sampling was able to verify the declared activities at the facilities tested.[12] These activities sometimes could be detected several kilometers from the plant.

The IAEA believes that if environmental monitoring had been part of routine safeguards inspections in the 1980's, it would easily have revealed Iraq's weapons activities.[13] Of course, such monitoring would not necessarily have deterred these activities, because Iraq may not have co-located them with safeguarded activities that were subject to environmental monitoring had it known that this form of surveillance would take place. However, as noted above, separating weapon activities from legitimate nuclear activities would have significantly raised the costs and difficulty of the weapon program, and would perhaps have made it easier to detect covert facilities through other forms of surveillance.

The United States supported the IAEA field trials in several ways. Altogether, the U.S. financial contribution has been $6-8 million over 3 fiscal years.[14] Inspectors were trained; DOE's national labs did many of the bulk analyses; and the Air Force Technical Applications Center handled most of the particle analysis. The K-25 enrichment plant in Oak Ridge, Tennessee was one of the field trial sites. In addition, the United States is helping the IAEA design and construct a new clean lab for contamination-free handling and analysis of samples. Other IAEA members also supported the environmental monitoring program, and labs in many countries were used. Few if any other countries, however, can match the analytical precision of the U.S. labs, particularly for particle analysis.

In most cases, environmental monitoring will not produce a "smoking gun." Rather, it will supply information that must be combined with other sources to determine what activities have taken place. Thus it is a supplement to conventional safeguards, not a replacement. It could be an extremely important supplement, particularly if the IAEA is successful in obtaining access to sites and facilities related to the fuel cycle that are currently not available for investigation.

Implementing environmental monitoring may be controversial. The IAEA has determined that it has the authority to institute such monitoring at declared sites under agreements that have previously been negotiated with states with comprehensive safeguards agreements, but any change from current practices may alarm some countries even if they have no undeclared facilities. One problem is in the detection of plutonium and other radioisotopes. If sensitive environmental monitoring detects emissions from facilities claimed not to be emitting anything, public fears may be raised—even though the emissions may be far below levels that could threaten public health and safety. Furthermore, licensing difficulties may be encountered if any contamination is found in the area of a facility. Also, under some conditions, competitive information may be divulged, such as the operating conditions of an enrichment or fuel fabrication plant. Thus the nuclear industry in some countries may be concerned. On the other hand, the IAEA is used to protecting any proprietary information it has obtained under safeguards practices, and it would reasonably withhold any such information collected through environmental monitoring, assuming that no safeguards violations were found. Implementation must be done carefully and sensitively. One compensating factor is that the information may be helpful to the host state in meeting its own health and safety goals.

[12] Personal communication with International Atomic Energy Agency staff member, Mar. 31, 1995.
[13] Personal communication with International Atomic Energy Agency staff member, Apr. 4, 1995.
[14] Personal communication with Ira Goldman, U.S. Department of Energy, Jan. 11, 1995.

The field trials have demonstrated that costs to the IAEA of implementing environmental monitiring should be modest. Sampling can be done in conjunction with regular inspections. Environmental sampling requires personnel with only several days of training and relatively simple equipment. The lab analyses are more expensive than the sampling, especially for particle analysis. The data indicate that bulk analysis is probably adequate to detect undeclared activity at most facilities, and that with appropriate selection of samples to be tested, the costs will not be prohibitive.

In some cases, the IAEA may be able to reduce inspections when environmental monitoring is introduced. For example, if environmental monitoring confirms that a country is not operating a reprocessing plant, then inspection of spent fuel need not take place as often in order to provide timely warning of diversion.[15]

Wide area monitoring to detect undeclared facilities is much more problematic. Some materials can be carried long distances, either in the air or in waterways. Monitoring rivers is not difficult, and positive findings can be traced upstream. Furthermore, sediments often collect at various places, establishing a record of what has come downstream. The IAEA already is monitoring water in Iraq. However, it is relatively easy for a small, covert facility to minimize liquid runoff, and in dry areas there may not be sufficient rain to wash away and concentrate material that settles out from the atmosphere. Therefore, clear signals may not emerge. Effective air monitoring requires a great many stations, because a plume can follow an erratic pattern. These stations must be monitored frequently over an extended period if they are to catch a sporadic, short-duration plume, such as might result from the opening of a reactor or the reprocessing of a batch of fuel. Furthermore, all samples collected by these stations would need to undergo laboratory analysis. At a minimum, samples would have to be screened with a relatively low cost technique to determine if any require more precise analysis. Since the number of samples would be high, costs would be also. Hence air monitoring can be quite expensive.

Technology now under development should improve the capability to conduct environmental monitoring in several ways. Some will allow real-time, remote sensing. The Department of Energy's CALIOPE (Chemical Analysis by Laser Interrogation of Proliferation Effluents) Program, a collaborative effort at 5 national labs, is intended to produce instruments that can, from outside a site's perimeter, measure the constituents of a plume of emissions in the air. Real-time xenon and air particulate measurements are being developed by the Department of Energy. Other developments would increase the sensitivity of laboratory instruments, permitting the analysis of samples even more dilute than those that can be studied today. Portability of instruments is another goal so that inspectors can get an immediate indication of suspect isotopes or chemicals and monitor more intensively. Successful development of these projects should significantly improve the effectiveness of environmental monitoring. However, some of these projects may involve technology that cannot be given to the IAEA because of U.S. national security concerns.

If successfully implemented, environmental monitoring will be an important part of international non-proliferation efforts. In addition, it may prove to have a role in verifying the Comprehensive Test Ban Treaty and nuclear material production cutoff agreements.

[15] Environmental monitoring will not detect a complete but unused reprocessing plant, nor any plans to send the spent fuel to another country for reprocessing. However, both these approaches probably would entail considerable delay and uncertainty in the procurement of plutonium relative to having a proven capability already.

Detectable Emissions 2

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.

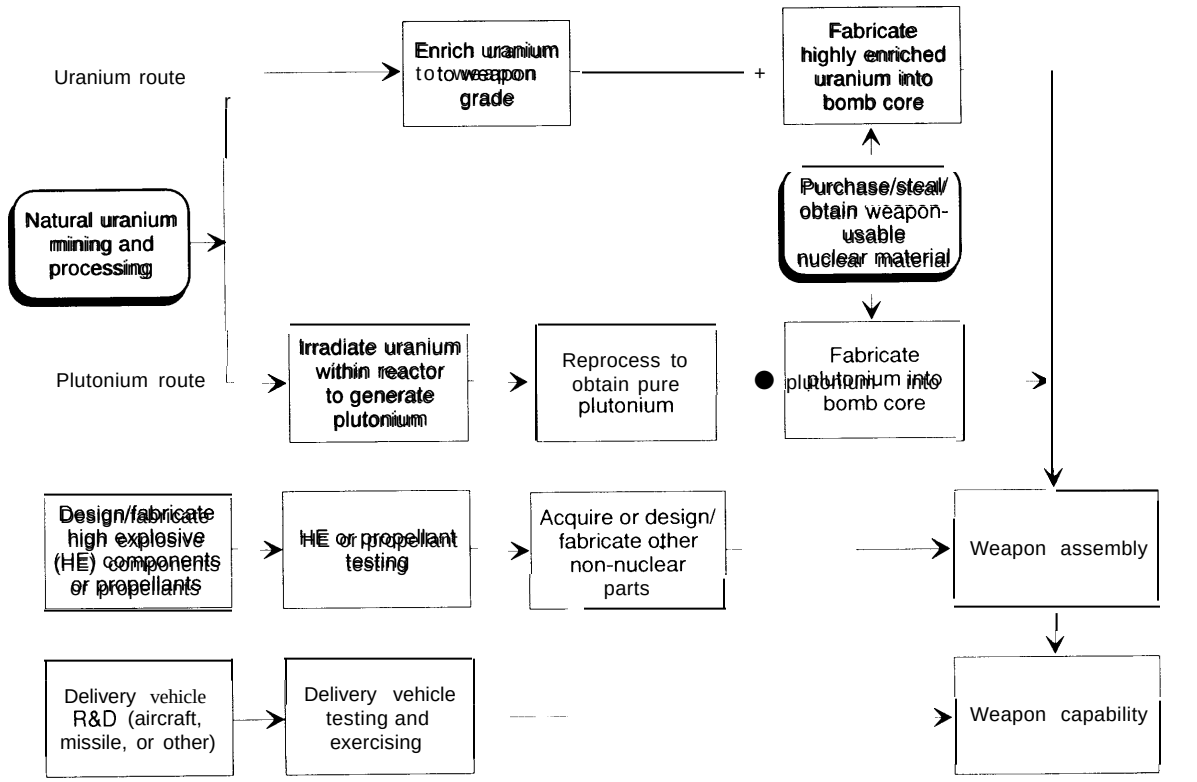
This chapter reviews the steps that must be followed by a nation clandestinely producing nuclear material, and identifies the signatures, or potentially detectable indications, that might be detected via environmental monitoring. There are two basic routes to produce fissile material for nuclear weapons: 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

[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

[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 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

[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

[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}.

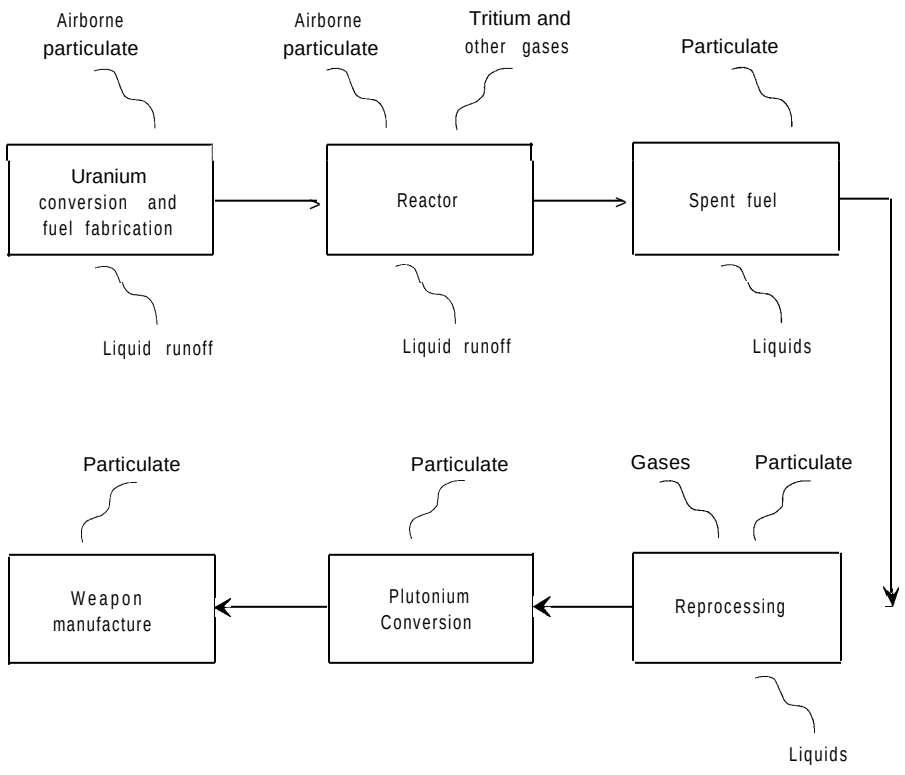
[7] A third choice, buying or stealing plutonium, either from the commercial nuclear power fuel cycle (if and when plutonium becomes a routine part of the cycle) or from a nuclear weapons state, is not considered here because the role of environmental monitoring would be peripheral.

safeguarded spent fuel only if it 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

[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,

[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

[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.

Technologies for Detection of Emissions 3

Detecting the emissions discussed in the last chapter requires a variety of techniques. The samples must be collected from carefully selected locations using procedures to prevent cross-contamination. They must then be transferred to the analytical laboratory. Many different lab instruments are available. Selecting the instruments to be used depends on the type of sample, the materials that might be found in it, the precision needed for the answers, and other factors. Then the results must be interpreted.

This chapter starts with a description of environmental monitoring as used in Iraq, the first public demonstration of its value. Then it describes the various steps that constitute environmental monitoring. Progress in making environmental monitoring a routine part of IAEA safeguards is discussed in the next chapter.

THE IRAQI EXPERIENCE

The agreement ending the Gulf War included Iraq's acceptance of United Nations inspection of all facilities suspected of contributing to a nuclear weapon capability. Within six weeks, inspectors from the International Atomic Energy Agency arrived to begin an exhaustive, and at times dramatic, survey of Iraq's nuclear weapon facilities.

Much of the equipment and materials had been hastily removed and hidden during the war and the month following, so the inspectors used means other than conventional materials accountancy and equipment examination to detect activities. Hundreds of samples were collected and sent to various labs for analysis. Many of these samples were quite nontraditional. Inspectors took smears from the surfaces of equipment and from the buildings themselves. They also collected samples of soil, vegetation and water outside the buildings. The first eight inspections in 1991 produced 464 samples of non-nuclear materials.[1] The first set of analyses of samples provided information to guide subsequent sampling. It was important to get the results of the analyses rapidly so that subsequent inspections could be designed to build on that information.

[1] Including samples of construction materials such as concrete and steel, which are not considered part of environmental monitoring. D.L. Donohue and R. Zeisler, "Behind the Scenes: Scientific Analysis of Samples from Nuclear Inspections in Iraq," *IAEA Bulletin*, January, 1992.

The analyses were performed by the IAEA itself and at labs in several member states, including the United States. The IAEA has two labs at Siebersdorf, Austria: the Safeguards Analytical Laboratory (SAL) and the Physics, Chemistry and Instrumentation (PCI) Laboratory. SAL, the prime safeguards laboratory, analyzes nuclear materials for uranium and plutonium content and isotopic composition. PCI uses a different set of techniques to analyze for radionuclides and other elements in the environment; much of its work has been on the International Chernobyl Project.[2] Specific techniques are discussed in the following sections. The analyses for the Iraqi samples are listed in table 3-1.

The most surprising result of the analyses was the discovery of isotopically altered uranium which did not match any known (declared) materials. Iraq was not known to have any enrichment capability at all, making any evidence of enrichment a surprise. That surprise was greatly compounded with the discovery of extremely depleted uranium (i.e., with a very low fraction of U^{235}) that could only have been produced with electromagnetic or laser separation techniques—techniques not known to be in commercial use

TABLE 3-1: Samples from Iraq Processed at IAEA Laboratories in 1991

Inspection	Non-nuclear materials	Nuclear materials
1st	48	31
2nd	35	0
3rd	139	51
4th	41	0
5th	49	61
6th	7	0
7th	139	141
8th	6	105
Total	464	389

Measurements requested

Sample category	Sample types	Analyses requested
Non-nuclear materials	Smears	Presence of U, Pu, or radionuclides
(Environmental)	Vegetation	Amount of U, Pu
	Soil	Presence of F, Cl
	Debris	U, Pu isotopics
	Rocks, Ores	Presence of high explosives
	Water	
(Materials of construction)	Graphite	Purity, type or identity
	Steels	
	Beryllium	
	Unknown metals	
Nuclear materials	Uranium metal	Amount of U, Pu
	Uranium compounds	U, Pu isotopics
	Plutonium compounds	Amount of polonium
	Polonium	Compounds of U, Pu
	U, Pu waste and scrap	Trace elements in U compounds

SOURCE: D.L. Donohue and R. Zeisler, "Behind the Scenes: Scientific Analysis of Samples from Nuclear Inspections in Iraq," *IAEA Bulletin*, January 1992.

[2] D.L. Donohue and Rolf Zeisler, "Analytical Chemistry in the Aftermath of the Gulf War," *Analytical Chemistry*, 1993, 65,359A-368A, American Chemical Society.

anywhere in the world. As discussed in chapter 2, the only commercial-scale enrichment technologies—gaseous diffusion, centrifuge, and advanced vortex tube—discharge depleted uranium with tails of about 0.3 percent U^{235}. No one believed that Iraq was ahead of western developers of laser enrichment, but it still took a while for experts to accept that anyone would use such an old, energy-inefficient technology as electromagnetic separation.[3] After three inspections the evidence from sample analysis and other means was compelling, and Iraqi officials were forced to reveal the EMIS program. Enrichment equipment had been operated at Tarmiya and Tuwaitha, and additional facilities were under construction at Ash Sharkat. When calutron components themselves were recovered and materials analyzed, uranium in one sample was found to have only 0.06 percent U^{235}.[4]

Other analyses also disclosed undeclared irradiation of uranium to produce very small quantities of plutonium. However, this activity, detected by analyses of conventional nuclear material samples, was a minor part of the Iraqi program. In addition, uranium from three different ore bodies was discovered, including indigenous production as a byproduct from an Iraqi phosphate plant.[5]

Environmental monitoring was used to verify the accuracy of the final Iraqi declarations. It is still used to verify that no covert nuclear material production is underway. In particular, wide-area monitoring of waterways is performed, yielding a high probability of detecting any significant production of plutonium.[6] The IAEA takes water, sediment, and biota (plants and animals) samples at 50 river stations in Iraq every two years. The IAEA does not maintain air monitoring stations because of the expense. It is, however, considering air particulate samplers mounted on its vehicles. These would direct air through a filter as the vehicle moves, collecting dust and other materials. This is a promising approach in Iraq because the agency has many vehicles and access everywhere. Car engine air filters have also been used as collectors. Other forms of environmental monitoring, such as the collection of soil and plant samples, are still being performed to verify compliance with agreements. On the average, one IAEA inspector is in the field every day in Iraq.

Early problems in the Iraqi environmental monitoring program have largely been overcome. Contamination ruined some samples. The trace quantities characteristic of environmental samples can be contaminated by exposure to almost any source of radionuclides. For example, one scientist from a U.S. national lab apparently left tiny particles of nuclear materials from his home lab at several sites, giving erroneous readings when they were unknowingly collected with the environmental samples.[7] Greater attention to cleanliness and double bagging of samples has been important. In addition, contamination from Chernobyl, especially cesium-134 and -137, was measured in Iraqi samples and had to be taken into account. Even natural uranium created problems. One building was made of concrete that had 25 parts per million of uranium, a level close to minable ore quality.[8]

SAMPLING

The collection of samples is conceptually simple, but there are some complicated aspects. The object is to collect gaseous, particulate or liquid emissions from a covert nuclear operation. The intent could be to determine if undeclared activi-

[3] Anthony Fainberg, *Strengthening IAEA Safeguards: Lessons from Iraq*, Center for International Security and Arms Control, Stanford University, April 1993, pp. 11–15.

[4] Since this sample was found on the collector of a calutron, it was not the result of environmental monitoring. See D.L. Donohue and R. Zeisler, "Behind the Scenes: Scientific Analysis of Samples from Nuclear Inspections in Iraq," *IAEA Bulletin*, January 1992.

[5] Personal communication with IAEA staff, Apr. 3, 1995.

[6] Personal communication with IAEA staff, Apr. 4, 1995.

[7] Ibid.

[8] Ibid.

ties are taking place at a declared facility, or to search for an undeclared facility. As practiced so far by the IAEA, sampling has mainly been conducted at declared facilities. The most important technique has been taking swipes from equipment or other surfaces in the targeted facility. Swipes (or smears) are simply pieces of cloth or paper (kept scrupulously clean to avoid contamination) which are wiped on the surface and stored in a plastic bag. Wiping picks up particles that have settled on the surface. Even thorough washing of surfaces is unlikely to remove all the particles, and inspectors learn where to find the optimum places to take swipes. The swipes need not be taken directly in the process area to detect what activities took place there; common areas and adjacent rooms collect enough telltale particles also, carried in by personnel, equipment, or by air currents.[9]

Taking swipes inside a facility is the most effective technique of environmental monitoring. It is a powerful tool that is generally unavailable to national intelligence efforts, which normally do not have such facility access.

Outside the plant, sampling can still involve swipes, for example on window ledges, signs, or shiny leaves such as aloe. However, it is often easier to simply pick the leaves or other parts of the plants. Pine needles are good collectors because they have a sticky residue that is particularly likely to hold particles. Not only do plants (biota) capture particles that have settled on their surfaces, but they also collect soluble material carried to their roots in groundwater. Thus biota act as integrators of emissions over periods of weeks to years. Grazing animals can also collect particles, which are concentrated in their excrement.

Similarly, soil near the surface can be collected. Particles settle from the air and can remain on the surface for many years.

Air samples, either gaseous or particulate, can be taken either at stationary sampling stations or from vehicles traveling in suspected areas. If the plume from the facility is caught, very accurate and timely information can be gleaned. Gaseous plumes can, under some conditions, be detected for hundreds of miles. There are several problems with gas and particulate sampling, however. First, some emissions from nuclear activities occur in puffs rather than on a continuous basis. For example, when a reactor is opened for refueling, or a batch of spent fuel is chopped to extract the plutonium, gaseous emissions occur briefly. Second, the movement of the plume can be very erratic, depending on local winds. Thus the network must be fairly dense to have a high probability of capturing the signatures. Not only is such a network expensive to construct, but the stations need to be visited frequently to collect samples or otherwise service the equipment. The dense network required with different sampling techniques for gases and particulates, their maintenance, and particularly the laboratory analysis of the large number of samples required add up to a very large operating expense. Furthermore, once a plume is detected, it must be attributed to a source. This requires a meteorological database as well as models to analyze the transport and dispersion of effluents.

Emissions can get into water from liquid runoff from the facility, by settling out onto the ground and getting washed away by rain, or by settling directly into the water. Holding ponds are particularly desirable as sources because they catch the runoff directly. There are two techniques for sampling water. One is to just fill a bottle. The other is to pump water through a filter to collect suspended material or an absorber to collect soluble or colloidal species. Lakes, small streams, rivers, and seas/oceans are all possible sources, though oceans tend to dilute the signal rapidly due to mixing.

A related collection medium is sediment in a river or the ocean, which can be dug up and bagged. Sediment can accumulate over a long period of time, providing a record of what came down river over time. It is tricky to select a rep-

[9] Personal communication with IAEA staff, Apr. 3, 1995.

resentative deposit; depending on the river flow pattern, a specific piece of sediment could be a single recent deposit or an old one with no recent material. In either case, the sediment could be deposited when no nuclear material was present even though at other times it was present. Collecting water plants is another option, as with terrestrial plants. One advantage of water as a source is that it is much easier to trace a detected substance up-river than up-wind. Whereas winds change, the watershed remains fixed.

Construction materials, while not literally a source for environmental sampling, are also considered because the same analytical techniques can be used on them. Moreover, they are likely to be collected along with environmental samples as part of the IAEA's upgrading of safeguards. Steel and other materials from a reactor can carry considerable information on the operation of the reactor. Even analysis of unirradiated material can be useful as with, for example, steel pieces in Iraq that had been thought to be parts of centrifuges for enrichment, but which were shown not to be maraging steel, a material critical for centrifuge rotors.

A careful plan is needed to know where and how to sample to have the greatest chance of finding telltale emissions with a minimum of effort. Inspectors cannot simply arrive at a facility and start collecting samples. For example, if prevailing winds blow consistently in one direction, there is little point in sampling intensively upwind. If an HEU plant is suspected, tritium or noble gases are unlikely to be emitted, so gas sampling will serve little purpose. The plan must take into account the type of suspected activity, the terrain, local vegetation, weather patterns, funds available for analysis, and other factors.

Once collected, the samples are double-bagged to prevent contamination, and sent to the IAEA's laboratories which protect the samples from particle contamination. A clean room is being built there with U.S. assistance. The clean room, which will have carefully controlled and filtered ventilation, will contain the shipping area, storage areas for sample archives, and analytical instruments. The requirements for the clean room far surpass anything the IAEA has had to construct in the past, because environmental samples contain so much less critical material than do the traditional safeguards samples of process materials. It is critical to avoid cross contamination.

ANALYTICAL INSTRUMENTS

A broad range of analytical techniques is needed for light and heavy radionuclides with different modes and rates of decay (see box 3-1), inorganic chemicals, and organics. The specific analytical technique chosen for a sample depends on the type of sample, the signature suspected, accuracy required, the rapidity with which results are needed, and costs. Samples may be divided and tested by several means. As environmental monitoring becomes routine, the IAEA will archive samples, or parts of them, to establish a baseline that can be compared with anomalies discovered in the future, perhaps with more sensitive instruments.

The major categories of analytical lab instruments are:

- Radiometers (counters) which measure the type and intensity of radioactive decays (alpha particle, beta particle, gamma ray).
- Spectrometers to characterize the radiation emitted by decaying or excited atoms; may be combined with counters.
- Mass spectrometers, that separate isotopes according to their different masses and measure their relative abundance (i.e., they derive a spectrum of masses). Several technologies may be combined with the mass spectrometer to improve sensitivity.
- Microscopes and electron microscopes, for examination of particles.
- Traditional chemical instruments such as titration or chromatography apparati, which determine which chemicals are present and their concentration.
- Specialized instruments such as lasers and reactor irradiation for activation analysis.

BOX 3-1: Radioactive Decay

Radioactive nuclei spontaneously decay (break up), emitting observable radiation and transmuting themselves into other nuclei. This process continues until a stable form is reached. Transmutation can be accomplished by the emission of alpha rays (helium nuclei), or beta rays (electrons or positrons), or by the absorption (capture) of an orbiting electron. Gamma rays and x-rays, which are part of the electromagnetic spectrum that includes visible light, may also be emitted, but this changes only the energy level, without itself causing transmutation.

Only the heaviest nuclei emit alpha particles. These are mainly actinides, elements of atomic number 89 (actinium) and higher. Uranium (92) and plutonium (94) are actinides. Alphas are emitted with characteristic energies that can be measured accurately to identify the isotope of origin. Alpha spectrometry is used to characterize nuclear weapon material.

Isotopes lighter than actinides that indicate nuclear reactor operation are fission products (from the fissioning of nuclear fuel) or activation products (non-nuclear materials such as fuel cladding or reactor components that have absorbed neutrons). These usually have excess neutrons, and so decay by emitting electrons (positron emission or electron capture occur in nuclei that have too few neutrons). The beta particles emitted by a given isotope range in energy from zero to a characteristic maximum. Measuring the energy of a given beta particle gives very little information about its source, although a spectrum or collection of beta particle energies can be used to identify a particular beta emitter. Thus, substances such as tritium (hydrogen-3, which has an unusually low-energy beta) can be measured by a combination of chemistry and beta spectrometry.

Each decay carries with it a characteristic pattern of gamma rays and x-rays. In many cases, gamma-ray spectrometry can identify an isotope by measuring the energies and relative abundance of the gammas it emits. However, many isotopes do not emit an identifiable spectrum; the spectral lines can be too weak or so myriad that a pattern cannot be identified. Also, the "fingerprints" of different isotopes overlap, so that an abundant constituent can mask a rare one. Experience is needed to understand the cases where gamma rays yield useful information.

SOURCE: Office of Technology Assessment, 1995.

Some of the most important instruments that can be used to analyze environmental monitoring samples are described here. Microscopes and conventional chemical techniques, such as are already used for safeguards materials analysis, are not included.

Gamma-ray spectrometry—measures energy of gamma rays from radioactive decay. Each decay has a specific energy level(s) which may be identified easily. This method is used for screening samples as it is quick and requires no elaborate preparation. It also can identify a broad range of isotopes, including fission products (e.g., cesium-134 and -137, ruthenium-106), activation products (e.g., cobalt-60), and actinides (e.g., uranium-235, plutonium-239, and americium-241). It does not require isotope-specific preparation, and can be used for any of the environmental samples discussed above. Sensitivity can be high; short half-life isotopes can be identified from samples containing only thousands of atoms.

Alpha-particle spectrometry—measures the energy spectrum of alpha particles emitted by plutonium and other actinides so that the isotopes present can be identified by their characteristic energy levels (similar to the gamma-ray spectrometer). This technology can measure the ratio of uranium isotopes in a 1 microgram sample, providing useful information as discussed below. Plutonium can also be detected in amounts less than one nanogram.

Beta spectrometry—a counter for beta decay. The emitted beta particles (electrons) are

directed through a fluid. In a liquid scintillation counter, fluid molecules struck by particles are excited and emit flashes of light. The liquid scintillation counter is used to measure strontium-90, an important fission product that can be released during reprocessing. Tritium is measured by gas proportional counters, which produce current pulses whose magnitude is proportional to the energy of the incoming beta particle.

Neutron activation analysis—the sample is irradiated with neutrons from a reactor or other source to generate radioactive isotopes. Then the gamma ray spectrum lines are measured to identify the radioisotopes; the intensities of the gamma emission lines indicate the concentration. The process can be automated and rapid. It is especially good for iodine-129, technetium-99 and elemental fluorine.

Delayed neutron counting—the sample is irradiated with neutrons. Fissioning isotopes (U^{235} or Pu^{239}) emit more neutrons, but not all at the moment of fissioning. Some are delayed for a matter of seconds. Counting the delayed neutrons gives a measure of the fissile isotopes present. Nanogram quantities of uranium or plutonium in the sample can be measured.

X-ray fluorescence (XRF)—the sample is stimulated by X-rays, gamma rays, or energetic particles to produce fluorescent emissions. Emissions from elements ranging from sodium to the heaviest elements of the periodic table can be measured and identified by comparison to standards for the various elements. This technique is used for rapid screening of samples. It identifies chemical elements—and is good at detecting metals, including uranium and plutonium—but it does not differentiate among isotopes of the same element.

Laser fluorimetry—the sample is ashed (burnt or oxidized under controlled conditions to remove combustibles and preserve the noncombustibles) and dissolved in hot nitric acid. It is then illuminated with an ultraviolet (UV) laser and any fluorescence measured. Many uranium compounds fluoresce, and this instrument can measure them with high sensitivity and accuracy.

Inductively Coupled Plasma Mass Spectrometry (ICPMS)—the sample is atomized in a high temperature plasma and directed into a mass spectrometer. Once inside, the atoms pass through a magnetic field which forces them to move in a curve, with the lighter atoms curving more sharply than the heavier ones. This separation allows them to be collected separately and measured, permitting their identification. ICPMS can measure nanogram-quantities of uranium or plutonium with good accuracy. For example, the uranium-235/238 ratio of a sample with several tens of nanograms of uranium can be measured with 2 percent accuracy.[10] The procedure is rapid and requires no elaborate preparation. It is particularly good for water samples because the suspect material must be in solution anyway. Over 70 elements can be surveyed.

Thermal Ionization Mass Spectrometry (TIMS)—the sample is ashed and the residue (or just a particle) is attached to a filament. The filament is heated by an electric current to a high temperature, which vaporizes the sample. The vapor is ionized and the ions directed into a mass spectrometer, as with the ICPMS. The TIMS technique can detect small deviations from natural isotope ratios. It is the most accurate instrument in common use to measure uranium and plutonium composition and is especially valuable for low concentration samples (e.g., nanograms per liter). TIMS is more accurate than ICPMS, allowing the detection of more dilute samples. The ratio of uranium 235 to 238 can be measured to at least 0.5 percent accuracy.[11] However, the cost can be several thousand dollars per sample, about 2 to 5 times higher than that of ICPMS.

Accelerator Mass Spectrometer (AMS)—uses a particle accelerator as input to the mass spectrometer to achieve greater separation. This

[10] Personal communication with Philip Miller, Lawrence Livermore National Laboratory, Mar. 1, 1995.
[11] Ibid.

is the most sensitive instrument currently available, and relatively few labs have one. It can be used to measure small changes in the concentration of naturally occurring but rare radioisotopes with long half-lives (e.g., carbon-14 and iodine-129, which are created by cosmic rays as well as by nuclear activities). AMS can detect isotopic concentrations of 1 atom in 10^{15}, making it excellent for trace samples collected far from the emitting site.[12]

BULK AND PARTICULATE ANALYSIS

Once a sample is collected, it can be examined in two major ways. First, the entire sample (or just a portion) can be tested using one or more of the instruments discussed above. This is called bulk analysis. Not only the radioactive or chemical material sought, but the medium (e.g., soil, vegetation) and sometimes the collecting agent (swipes, filters) are tested. Bulk analysis reveals information about average properties of the sample and indicates the presence of anomalous components.

Alternatively, suspect particles themselves can be isolated and analyzed. A swipe of a dirty piece of equipment can collect many thousands of particles, so identification of the relatively few interesting ones can be difficult. The method used by the Air Force Technical Applications Center (AFTAC) laboratories is to extract the particles from the sampling media (swipe), attach them to a clear plastic (Lexan) and irradiate them in a nuclear reactor. Uranium and plutonium atoms fission during irradiation and leave tracks in the Lexan. When viewed under a microscope, the particles that produced the tracks can readily be identified and isolated.[13]

Individual particles can be examined optically and with an electron microscope, and then tested by TIMS or other mass spectrometers as discussed above. Much can be learned visually by a trained observer. In fact, Russian particle analysis is largely based on intensive visual inspection. This is a very different approach from U.S. analytical techniques and can discover different things about a particle.[14] The morphology, or structure of a uranium or plutonium particle bears information as to how it was formed. For example, a particle of uranium might contain UO_2, U_3O_8, and U_4O_9 in various structures, depending on the temperature at which it was formed. That indicates the process that was used. A scanning electron microscope (SEM) is used to provide a preliminary analysis, such as whether the particle is crystalline or amorphous, and whether other materials are embedded in the particle.

Particle analysis is considerably more labor intensive and costly than bulk analysis. Thus it is used only where extreme sensitivity and precision is needed. In the IAEA field trials, bulk and particle analysis yielded approximately the same results when samples rich in particles were directly compared. Thus the two approaches complement each other, serving somewhat different purposes.

DATA INTERPRETATION

The information produced by the techniques described above cannot be expected to unambiguously define whatever activities have taken place. It may be conclusive, as when HEU is found in a variety of samples at a LEU plant. Most activities, however, can be concealed sufficiently that only some samples will have any indications, and these may be inconclusive. A few particles of plutonium do not guarantee that reprocessing is taking place, as they may be fallout from the Chernobyl accident or weapons testing, or contamination from a legitimate research center. Analysts must be experienced with the operations that could have produced them, and

[12] Ibid.

[13] Personal communication with Peter Aldred, Advanced Nuclear Applications (AFTAC analytical lab), Vallecitos Nuclear Center, Mar. 2, 1995.

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

the tools, such as burn-up computer codes, that can be used to understand the possibilities. In addition, they must know how much data is required to reach conclusions, and how reliable the data must be. Further sampling and analysis may have to follow the first indication to determine if it was an isolated anomaly or truly indicative of illicit operations.

Reaching conclusions on the existence of undeclared activities is likely to require a multidimensional analysis, of which environmental monitoring will be only one part. Environmental monitoring itself will make a strong case if several different signals (as discussed in chapter 2) can be verified, especially for reactor operation and reprocessing. Uranium and plutonium isotopes, perhaps in conjunction with other actinides, are a good indication of reprocessing. Finding other radionuclides such as fission and activation products, and chemicals such as tributyl phosphate, would greatly strengthen the indication. Finding tritium in waterways or the air strongly suggests reactor operations.

As has been noted above, even minute traces of radioisotopes can be detected. With the more sensitive instruments, a few million atoms of uranium can be sufficient for measurement and a few billion can determine the full isotopic fingerprint, as shown in table 3-2. The ratio of certain isotopes can yield valuable information. For example, the ratio of uranium-234 to -235 can indicate the type of enrichment that was used. The bigger the sample, the more information that it yields. With a sample containing 0.1 to 1.0 grams of plutonium (implausibly large for an environmental monitoring sample, but easily available from conventional safeguards or a smuggling case), an analyst may be able to tell the date of separation from spent fuel; the method of casting; irradiation time; the original enrichment; and the reprocessing technique.[15]

The lab information must be analyzed by an experienced analyst to extract the appropriate conclusions. What are the possible sources of the isotopes or chemicals found? How consistent with these sources are the forms of the discovered substances? If the suspect activity is taking place, what else should be found? How long, and at what scale, has it been taking place? Is additional sampling necessary to confirm these findings, and what sort of sampling should it be?

TABLE 3-2: Measurement Sensitivities (TIMS)

Element	Detection	Isotopic analysis
U	5×10^6 atoms	5×10^9 atoms
Pu	5×10^5	5×10^8
Am	2×10^5	2×10^8
Np	2×10^5	2×10^8

SOURCE: Don Rokop, Los Alamos National Laboratory, 1995.

This information also must then be analyzed on a country- and site-specific basis before any accusations can be made. What other activities are known to be taking place? Is the country capable of mounting this kind of operation? What other indications (e.g., imports of equipment, movement of technical personnel) support or contradict the conclusions? Are the topography and local weather patterns of the suspected site consistent with finding samples where they were found?

Analyzing large numbers of environmental samples will generate huge amounts of data. Keeping track of all the data will be difficult. Adequate provisions must be made for computers and software to handle, process, and store the information.

CONCLUSIONS

The analytical techniques described in this chapter can detect and characterize routine emissions from nuclear facilities. They are sufficiently sensitive to have a high probability of detecting covert activities to produce nuclear weapons materials if the sampling is close to the facility. Long-distance monitoring, especially of the air, is more problematical. The more dilute the emissions become, the less likely that critical

[15] Personal communication with Sid Niemeyer, Lawrence Livermore National Lab, Mar. 1, 1995.

materials can be distinguished from background, or that they can be traced back to the source. Sampling and analysis will improve over the next few years as the IAEA becomes more experienced and uses already available, more sensitive equipment. Technologies under development may improve analytical capabilities, as discussed in chapter 5.

Status and Plans for Implementation 4

The International Atomic Energy Agency (IAEA) is moving to take advantage of environmental monitoring to strengthen its safeguards. The United States and other members have strongly supported this initiative with funding, expertise, and assistance with lab analysis. Implementation will call for careful planning to minimize costs and maximize effectiveness.

FIELD TRIALS

The IAEA has conducted a series of field trials in 11 cooperating member nations to determine how best to conduct environmental monitoring and the results that can be expected. A variety of installations were tested in order to gain experience with reactor operation, enrichment, reprocessing, and other functions that are likely to be employed in a weapon program.

Results from several countries have been released including Sweden, South Africa, Australia, Argentina.[1]

Sweden—tests were conducted in the vicinity of five separate nuclear facilities. Three were nuclear power stations with a total of 10 reactors. The others were a nuclear research facility and a fuel fabrication plant. All were on or near the coast (Baltic Sea or Kattegat, across from Denmark), and samples were taken of coastal water, sediment, and biota up to 30 km away. Analyses proved capable of detecting activation products from reactor operation (e.g., Co-60) up to 20 km from the site. In addition, a small amount of high-burnup plutonium, clearly distinguished from fallout, was found near the research center.

South Africa—an enrichment plant at the Pelindaba site produced the highly enriched uranium used in South Africa's seven nuclear weapons.[2] A second plant on the site produced LEU for South Africa's power reactors. These plants are now closed. Soil, vegetation, and water samples were collected in and near the facility grounds. Swipes were taken in and near the process buildings. The vegetation samples, including those taken well away from the facility,

[1] These results are drawn largely from: Bruno Pellaud and Richard Hooper, "IAEA Safeguards in the 1990s: Building on Experience," *IAEA Bulletin*, vol. 37, No. 1, March 1995.

[2] For further information, see Adolf von Baeckmann, Garry Dillon, and Demetrius Perricos, "Nuclear Verification in South Africa," *IAEA Bulletin*, vol. 37, No. 1, March 1995.

showed traces of enrichment activities. Particle analysis of swipes was consistent with bulk analysis and showed in detail various levels of uranium enrichment, including depleted and natural uranium, and LEU and HEU. The swipes gave comparable results whether taken in the process area, auxiliary rooms, or outside the buildings.

Australia—the Lucas Heights Research Laboratories has conducted a variety of activities. Swipes inside a building housing a small centrifuge enrichment development program that was closed and dismantled 14 years ago still showed evidence of enrichment, including LEU and depleted uranium. One surprising result was the discovery of uranium depleted below declared levels, as low as 0.3 percent. Investigation revealed that this was from initial tests, when the centrifuge was fed with depleted uranium.[3] Starting off with depleted uranium, which has a uranium-235 fraction lower than that of natural uranium, produces tails that are more strongly depleted than are produced from natural uranium. Isotope production included molybdenum-99 (for medical use) from irradiating targets in a reactor. Swipes showed both target material and the irradiated products.

Argentina—the Pilcaniyeu gaseous diffusion enrichment plant produced LEU until 1991. The output was used to improve performance in a heavy water reactor, so the enrichment level was low, only 1.2 percent. This level is harder to distinguish from natural uranium than the 3 percent enriched LEU used for light water reactors. Swipes were taken inside the process and other buildings. Other samples included vegetation and soil around the site, and river water, sediment and biota both up- and downstream of the facility. Analysis showed depleted and natural uranium and LEU consistent with declared activities at the site.

Other countries participating in the field trials included the United States (the K-25 enrichment plant at Oak Ridge, Tennessee), Hungary, Japan, the United Kingdom, Indonesia, Netherlands, and South Korea. A total of 12 trials (two in Japan) were conducted.

Inspectors visited the sites, collected the samples, and sent them to IAEA headquarters. From there they were distributed to various laboratories, including the IAEA's own lab at Seibersdorf, as noted in chapter 3. Labs in the United States, United Kingdom, Russia, Hungary, Finland, Canada, and Australia also participated in the analysis. In the United States, bulk samples were sent to the Oak Ridge National Laboratory, which distributed them among the analytical labs at Oak Ridge, Savannah River Technology Center, Los Alamos and Lawrence Livermore National Laboratories, and Pacific Northwest Laboratory. For example, Livermore analyzed more than 200 samples for the field trials, using ICPMS and TIMS for uranium isotopics, and AMS for iodine-129 from reprocessing or reactor operation. Samples intended for particle analysis were sent to the Air Force Technical Applications Center for distribution to its network of labs, such as at the Vallecitos Nuclear Center and McClellan Air Force Base in California.

The field trials demonstrated the practicality of environmental monitoring under a broad range of conditions. Most declared activities were verified, although in some cases, particularly in the ocean sampling off Japan, the signatures had been so diluted that the results were limited. As would be expected, streams are better sources for samples than oceans. The process can be improved as more experience is gained. Inspectors will be trained to avoid contamination and to pick the best locations for sampling. Distribution and analysis should become more efficient. Data interpretation, in particular, is a skill that takes time to learn. For example, correlations among the isotopes detected in a sample often are more informative of the process under investigation than the quantity of any isotope alone.[4]

[3] Personal communication with IAEA staff, Apr. 4, 1995.
[4] Briefing by IAEA staff, IAEA, Apr. 3, 1995.

In addition, not all samples will have to be analyzed. Some can be archived until anomalous results suggest a more intensive analysis. Further work will also identify the key signatures that environmental monitoring can expect to identify, allowing improved focus and fewer analytical deadends. Both sampling and analysis will be better in a few years, allowing improved results at lower cost.

If the field trials had only verified activities that were known to have taken place at known sites, then little could be concluded on the feasibility of discovering clandestine activities. However, several anomalies were also discovered that turned out to be due to activities that were unknown to the inspectors. One was the detection of depleted uranium at the Australian site noted above. Another was the detection of plutonium at the Oak Ridge enrichment plant. The source turned out to have been reprocessed uranium with traces of plutonium from the Hanford, Washington facility that had been re-enriched at Oak Ridge many years earlier. Clearly, environmental monitoring can uncover previously unknown activities.

Other anomalies have yet to be fully explained. Cesium found in Hungary could have been from leaky fuel at the reactor, or it could have been from Chernobyl. More was found in downstream sediments than upstream, indicating a reactor source, but the difference could also have been a result of variable fallout or poor sampling.[5] If from the reactor, one would have expected to find cobalt-60 (an activation product of steel) also, as was the case in Sweden, but none was associated with the Hungarian cesium. At another site, a particle of HEU (30 percent enriched) was found that had no relationship to any activities at the site. It may have come from a previous visitor. Contamination, whether from an inspector or introduced in staging areas, is the most probable explanation for most anomalies.

As indicated in chapters 2 and 3, enrichment plants and reprocessing plants are easier to detect than reactors. In particular, reprocessing plants produce a variety of emissions that can be found in several different types of samples. Iodine from Dounreay and Sellafield in the United Kingdom has been detected in samples taken at the Chalk River facility in Canada by using a very sensitive accelerator mass spectrometer. Vegetation samples 30 km from Dounreay showed clear radionuclide evidence of reprocessing.[6] Of course, Dounreay is a very large source; a small, clandestine reprocessing plant probably could not be "seen" from as far away. Enrichment plant signatures are hard to detect using water sampling techniques, even those that concentrate radionuclides from high volumes of water. Lichens and moss are better media for detecting signatures from enrichment plants. Reactors were detected mainly from on-site water samples. A small, clandestine reactor would be quite hard to detect, especially if the background included a significant amount of contamination from fallout from nuclear weapon tests, Chernobyl, or the production and use of radioisotopes for medical or research purposes. R&D facilities were best characterized by swipe sampling inside the buildings; these samples produced unambiguous signatures, especially for isotopes such as uranium-235. Outside such facilities, vegetation and water samples showed evidence of nuclear activities as far as several km away.

The measurement of radionuclides from on-site sampling proved particularly effective. For isotopes of uranium and plutonium, particle analysis gave more precise results than bulk analysis. The combination of on-site samples and particle analysis was so sensitive that samples taken in common areas in enrichment plants showed comparable results to process room samples. However, the field trials also demonstrated that other sampling and analytical techniques work well too; nuclear activities can be detected at

[5] Personal communication with IAEA staff, Apr. 4, 1995.
[6] Ibid.

least several kilometers away from the emission point, and bulk analysis gave the same general results as particle analysis, although not as clearly. However, bulk techniques could analyze a larger portion of the sample, increasing the chances of getting a "hit." The different techniques complement each other and any of them may be most appropriate for particular conditions.

TRAINING

Taking samples for environmental monitoring is somewhat different than conducting conventional safeguards inspections, and inspectors must be adequately trained. Most inspectors are technically competent (they routinely sample process lines and operate sophisticated instruments), and environmental sampling tasks are relatively straightforward. However, the sampling strategy must be carefully planned, and contamination standards are much more stringent than for conventional safeguards. Furthermore, the quality assurance procedures are demanding. For example, inspectors have to record exactly where a sample is taken. The mechanics of the sampling can be taught in a day or two, but proper procedures must be learned over a longer period. In addition, enhanced observational skills (e.g., the ability to notice suspicious or anomalous equipment) must be taught so that inspectors can comply with the new activism in seeking out evidence of proliferation, as suggested by the IAEA Programme 93+2.

Only a few inspectors have had training in environmental sampling techniques. Oak Ridge provided training for most of those involved in the field trials (about 10). An initial training program has been conducted at Seibersdorf, with the first group in June 1995.

TECHNICAL CAPABILITIES AND THE NEW CLEAN ROOM

The IAEA did relatively little of the lab analysis for the field trials and expects to continue to rely on member states for much of the analytical work as environmental monitoring becomes a routine part of safeguards inspections. In fact, it may have to expand its network of laboratories as the work load grows.

The IAEA is building one essential facility for the new safeguards program—a clean lab to receive samples from the field, perform some analyses, and hold other samples until they can be transferred to its network of outside labs. Samples cannot be sent directly to an outside analytical lab because it is necessary to maintain confidentiality for the inspected facility. The samples are kept anonymous, which may involve repackaging and splitting them. As has been noted, it is essential to avoid contaminating these samples, and a clean room—where the air flow is carefully designed and filtered—is necessary to properly handle them. The clean lab will be at Siebersdorf, Austria, where the IAEA already has a large laboratory, but it will be kept separate from the other labs to minimize the risk of cross contamination. It will contain some of the basic instruments such as an electron microscope and a TIMS. The IAEA must be able to independently confirm results (especially positive results) to maintain its credibility with inspected states, but for analyses requiring expensive techniques such as AMS and particle analysis it must continue to rely on member states.

The clean lab is expected to be in operation by the end of 1995. The total cost for the lab itself (not including instruments) will be $3 million, of which $1.5 million is being covered by the United States. The equipment to be housed in the facility will cost approximately another $2 million, which will come from the IAEA's regular budget.[7] Additional funding ($160,000) could be used for a low-level gamma spectroscopy system to screen samples. The clean lab operations will require two professionals and two technicians. U.S. help is technical as well as financial. An American expert has been loaned to the IAEA

[7] Briefing by IAEA staff, Apr. 3, 1995.

for two years to help design and construct the lab, and other expertise also is being transferred.

Many improvements are expected in the future. Instruments and techniques under development in the United States are reviewed in chapter 5. Other work that may be relevant includes environmental restoration of weapons facilities. Studies of soil, radionuclide absorption by vegetation, and groundwater or ocean current movement provide information and models that the IAEA could use.

QUALITY ASSURANCE

The trace levels of materials sought in most samples require very stringent quality control in order to avoid contamination and inaccurate results. For example, inspectors may need new throwaway suits and booties every day. The IAEA is establishing proper procedures for taking and handling samples. Sampling kits have been designed and provided to inspectors with U.S. lab help. Generally two inspectors are needed—one to collect the sample and the other to hold the bag it goes into. The two cannot change roles during the day because of the risk of cross contamination.

Analytical labs also must practice strict quality control, both in the handling of samples and in the analysis. Some of the network labs may have been unaccustomed to the need for such quality control, because contamination occurred several times.

One of the clean lab's important functions will be to ensure adequate quality assurance at all stages.

IMPLEMENTING ENVIRONMENTAL MONITORING

The IAEA has been shifting its fundamental approach to safeguards. In prior years, particularly before the 1991 Persian Gulf War, its objective was primarily to verify that a state was doing what it said it was doing. States were required to declare all their nuclear material and establish a system of controls for it. The IAEA measured the material and verified that none had been misappropriated. Verification that no other activities were taking place was not seen as part of the IAEA's job, even though countries commit under the Non-Proliferation Treaty (NPT) to refrain from developing nuclear weapons in any way. This approach was adequate only if all members could be trusted, in which case such verification arguably would not be needed at all. It was shown to be fundamentally flawed when countries such as Iraq and North Korea proved themselves willing to ignore their commitments.

The new approach embodied in the IAEA Programme 93+2 is much more activist. If fully implemented, as the Board of Governors indicated it would be at the March 1995 meeting, the IAEA will search for undeclared activities. The State's declaration will still be the starting point for verification, but the IAEA will look for signs that contradict it. For example, the IAEA could cooperate with supplier states to determine if any countries are importing equipment inconsistent with peaceful, declared uses of nuclear power. It could also conduct literature searches and demand more information from inspected states.[8] The IAEA might take a country-wide approach—where might an undeclared facility be, considering factors such as national abilities, expertise, and topography, and how does one find it?

The main safeguards focus will still be on nuclear material, but the scope of verification activities will be expanded considerably. Environmental monitoring will be a key part of this approach. Environmental monitoring may serve as a warning flag that other inspections are needed. By itself, it is more likely to indicate subtle inconsistencies, rather than gross discrepancies, if in fact undeclared activities are taking place. It will also raise confidence where they are not taking place. Thus it will be a sorting tool used in combination with other conventional

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

safeguards, not a replacement. For example, if a country has reactors but no reprocessing facilities, the IAEA could use environmental monitoring to verify that no reprocessing is taking place. With that assurance, the frequency of inspections to verify that spent fuel has not been diverted (now every three months) could be reduced, since environmental monitoring can give a timely warning.[9]

Such a reduction in inspections entails some risk because a country might send its spent fuel to another country for reprocessing without notifying the IAEA. Alternatively, it could construct a clandestine reprocessing plant and not operate it until ready to quickly construct nuclear weapons. Environmental monitoring would not detect either of these avenues. However, both introduce considerable uncertainty and the potential for delay. Shipping spent fuel could be slow and might itself be detected. The reprocessing country might not apply the same level of secrecy, increasing the potential for detection, or might not even return the plutonium. Unused reprocessing plants, even small ones, are likely to require a significant shakedown period and are likely to release detectable emissions before purifying substantial amounts of plutonium. Therefore, environmental monitoring can support a reduction of inspections if accompanied by political analysis and broader information gathering.

Planning

Further study will be required before environmental monitoring can be fully integrated into the safeguards system. The major uncertainty is over the ability to find undeclared sites. Finding these will be very different than finding undeclared activity at declared sites. Potential proliferants will have greater incentive to build undeclared, covert facilities if they judge that weapon activities co-located with safeguarded facilities will be detected. More field trials are likely to be necessary to develop and test the means to find undeclared sites, especially reprocessing and enrichment plants. By definition, this means long-range monitoring, especially of rivers, which were not intensively examined in the earlier field trials.

The IAEA and several member states, including Canada and several Scandinavian countries, already are discussing field trials specifically designed to detect such activities.[10] Tests in weapon states would be particularly useful, but—given the power of environmental monitoring—they would have to be carefully designed to avoid compromising national security. The United Kingdom might be a possibility.[11]

A detailed plan for implementing environmental monitoring will be needed. An outline was prepared for the June IAEA Board of Governors meeting, but the full plan will not be ready before 1996. The full plan should cover topics such as facilities that will be subjected to environmental monitoring, the level of effort of inspectors at each kind of site, the training and equipment they will need, the labs to which their samples will be sent and the type of analysis to be done, procedures for quality assurance, how current safeguards will be modified, and the cooperation that may be needed with the host state. This plan will need considerable input from IAEA operations personnel and review and acceptance by member nations.

In addition to the general plan, specific plans will be needed for each site. Sampling must be done on the basis of the known operations at the site and expected signatures, possible undeclared activities, and the specific site characteristics such as topography and environmental conditions. The IAEA is documenting signatures from all relevant activities (see chapter 2). A current project at the Pacific Northwest Laboratory is developing a computer program, the EM

[9] Personal communication with IAEA staff, Apr. 4, 1995.
[10] Briefing by IAEA staff, Mar. 31, 1995.
[11] Personal communication with IAEA staff, Apr. 4, 1995.

Assessment Tool, which helps the user to plan inspections based on site and operational characteristics, and on safeguards needs and capabilities. This tool could be quite useful for wide area monitoring.

Initially, the plan will have to focus on sampling at declared sites. It will be very expensive if every safeguarded site has to be exhaustively sampled and analyzed. Nevertheless, the IAEA has to be sensitive to member concerns about being unfairly singled out for closer examination. Perhaps a list of critical facilities can be selected initially, with the number (e.g., 50) large enough to avoid charges of discrimination but small enough to be manageable. Baselines for these facilities could then be established.[12] Baselines will be especially important at research complexes that have a variety of activities that could produce emissions similar to weapons production. (Such places would be logical sites for covert nuclear facilities if remote siting is not possible.) Future samples can then be compared with the baseline to see if any new activities have been introduced.[13]

Data interpretation

Interpreting the information that is developed will be a particularly important function that will also be especially difficult to implement. If a confrontation with an inspected state ensues, it must be based on very strong evidence with virtually no chance of error. The IAEA must spend considerable effort on this area. Confidence is hard to quantify, especially since environmental monitoring is so different philosophically from current safeguards. The United States is providing assistance in this critical area.[14] Russia and the United Kingdom also could provide useful help. However, much of the equivalent work done by member nations is classified and will be difficult to share.

Costs

Adding environmental monitoring to the IAEA's activities should not greatly affect its budget.[15] Over the past nine years, the Safeguards Division budget has been under a zero growth restriction (actually it has been slightly negative when corrected for inflation). Additional costs for inspection and analysis will have to be largely balanced by reductions in other activities, such as material accountancy, and by learning to do more with less. As noted above, the frequency of some inspections may be reduced because of environmental monitoring. The United States paid for most of the field trial laboratory analysis, but the IAEA will have to cover these costs when the activities become routine. U.S. assistance has also included cost-free experts, who are individuals whose services are provided free of charge to the IAEA, but both the United States and the IAEA have limits on how many such experts can be supported. Safeguards in general, and environmental monitoring in particular, cannot be seen as a U.S. operation. Other nations must also be involved both financially and technologically for the IAEA to maintain its credibility. Fortunately, the level and breadth of support from other members has been quite good.

CONCERNS OVER ENVIRONMENTAL MONITORING

The IAEA will have to deal with several concerns on the part of inspected nations. Environmental monitoring is predicated on finding radionuclides released from nuclear facilities to the local environment. Many people are worried about exposure to any radioactive materials. Even though the level of radiation sought by environmental monitoring is far below any that might cause health problems, some people may become concerned that any radioactivity is being

[12] Briefing by IAEA staff, Mar. 31, 1995.

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

[14] Personal communication with IAEA staff, Apr. 4, 1995.

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

found. Public opposition could increase just by the knowledge that inspectors are looking for such radioactivity. Release of information on the material found could increase opposition even more, no matter what the levels are. Plutonium is particularly worrisome, as many people are unaware of how ubiquitous it is. Regulatory problems also are possible if radioactive materials are found.

Some states will have to make significant adjustments to accommodate environmental monitoring. Where operators have downplayed the emissions of plutonium and other radionuclides, new approaches to explaining the results will be needed, especially if standards have actually been exceeded. A compensating factor is that environmental monitoring may be quite useful for the state to achieve its own health and safety goals. Overall, most nations should be able to accept environmental monitoring, though they may want to place some conditions on it, such as the release of information.

A second concern is over competitiveness. Emissions can contain information on the process used in an enrichment or fuel fabrication plant. If this information falls in the hands of competitors, it could be damaging. The IAEA employs nationals of many different states, and it is not impossible that one would pass the information to a company back home. This is a more easily managed problem than public opposition. The IAEA already has access to plants with competitive concerns and is able to maintain confidence. Strict confidentiality of the samples, as will be accomplished by repackaging at the Siebersdorf facility, will help.

In addition, the advanced states may worry about compromising their own national security technology secrets. Some of the analytical techniques that are used for environmental monitoring were developed for national security purposes, and these states may not wish them to become more widely known.

CONCLUSIONS

Environmental monitoring will significantly increase the effectiveness of IAEA safeguards, especially for the detection of undeclared activities at declared sites. Implementation should be relatively straightforward, though considerable planning and consultation with all parties to the activity will be necessary. As the IAEA becomes more proficient, and improved technologies are made available, capabilities should expand considerably. U.S. assistance will be essential in this process.

Future Technologies 5

Research currently under way is developing new technologies that could improve significantly the effectiveness of environmental monitoring. This chapter first discusses how monitoring effectiveness might be improved, next summarizes the general directions of laboratory research, and then describes some specific approaches.

STRATEGIC DIRECTIONS

One significant improvement expected in future environmental monitoring is **remote sensing,** the ability to collect information without physically collecting samples. Remote sensing could be valuable for rapid screening of many sites, especially if that must be done without the cooperation of the state. (Note that this is not an IAEA function.) Two approaches are possible: aerial and ground-based, remote surveillance. Spacecraft already have unrestricted overflight rights, and flights for aircraft at altitudes above 10 kilometers could become acceptable under an Open Skies Treaty.[1] Sensors that operate at these ranges could be used routinely to monitor areas of concern without on-site inspection. The other approach is for shorter-range technologies that could sample a stack plume from a convenient nearby ground location.

The schemes for remote environmental monitoring discussed here are optical in nature, using ultraviolet or infrared light. There are active schemes that supply the light they need, and passive schemes that use existing sunlight or thermal radiation. In either case, the monitoring instrument views the area under investigation and analyzes light received from it.

As discussed in the section *Improving Technology,* such techniques can identify airborne molecules, but with only moderate accuracy and specificity. They are much less sensitive to solid material, and—in most cases—they cannot differentiate among isotopes of the same element. However, they are naturally adapted to surveying a large area for suspicious occurrences, and can select and focus on a small spot within that area.

Another direction of significant improvement will be instruments of **greater sensitivity and/or selectivity**. This would allow future investigations to use smaller samples (possibly down to

[1] U.S. Congress, Office of Technology Assessment, *Verification Technologies: Cooperative Aerial Surveillance in International Agreements,* OTA-ISC-480 (Washington, DC: U.S. Government Printing Office, July 1991).

single dust particles), process samples that are more dilute, measure additional contaminants, or provide higher-quality answers. The most dramatic improvement comes from combining two different technologies so that the substance being measured passes two independent selection screens. **Greater speed** would allow the processing of more samples for greater statistical accuracy.

Portability is also desired. Some new technologies are intrinsically small-scale, and other technologies are being miniaturized. An instrument that can be hand-carried at an inspection site can provide immediate results to guide the inspection strategy. At present, an instrument weighing 61 pounds can be called portable;[2] standards will change with further miniaturization.

Features of remote sensing and portability could be combined by mounting instruments on an aircraft and flying it to the inspection site. There it can fly through an exhaust plume, collect samples, and analyze them immediately.

IMPROVING TECHNOLOGY

Two kinds of technologies are under development for environmental monitoring to detect a clandestine weapon program. One kind looks for suspicious isotopes, such as uranium-235 or technetium-99 (as does most current technology as discussed in chapter 3). The other kind looks for suspicious molecules, such as HF (hydrogen fluoride) or TBP (tributyl phosphate). (See the end of this chapter for another possibility: looking for waste heat from a concealed nuclear reactor.)

Identifying Molecules

The chemist has many analytical tools to identify molecules of interest: infrared spectroscopy, mass spectrometry, chromatography, and benchtop chemical analysis. Finding specific molecules in the environment is useful for many missions, from detecting a chemical weapon program to measuring pollution; new technologies may be truly dual-use, with application both to national security and to civilian needs. On the other hand, a specific molecule may have many legitimate sources, and its presence only indicates an anomaly worth further investigation; also, some signature molecules will not be found in the environment for reasons such as low vapor pressure or because they react with atmospheric gases to form other species.

Infrared Spectroscopy

Infrared "fingerprints" observed by infrared or Raman spectroscopy (box 5-1) can be used to identify particular molecular species. Chemists usually record the entire fingerprint spectrum using a laboratory infrared source, and laser Raman spectroscopy[3] naturally gives the entire spectrum. Another technique under development that will cover a broad spectral region is a passive airborne infrared spectrometer.[4] Alternatively, it may be possible to look at individual spectral features using DIAL (differential absorption LIDAR), in which a laser alternates between the frequency of a characteristic molecular absorption line and a different nearby frequency, and the receiver looks for a corresponding change in return signal due to the presence of the absorbing molecule.[5]

Infrared or Raman spectroscopy is able to detect molecules as vapor in air, and in solution.[6] For the lighter elements (hydrogen to oxygen), different isotopes of the same element are distinguishable because their vibration frequencies, which depend on mass, can be differentiated. It is less suited for analyzing solid material, or for

[2] Brian Andresen et al., "A Small, Portable Gas Chromatograph-Quadrupole Mass Spectrometer for On-Site Analysis," Lawrence Livermore National Laboratory, UCRL-ID-116939, April 1994.
[3] Briefing by C. Chen, Brookhaven National Laboratory, Jan. 10, 1995.
[4] Briefing by A. J. Ramponi, Lawrence Livermore National Laboratory, Mar. 1, 1995.
[5] Briefing by Lyn Pleasance, Lawrence Livermore National Laboratory, Mar. 1, 1995.
[6] Briefing by C. Chen, Brookhaven National Laboratory, Jan. 10, 1995.

BOX 5-1: Infrared Spectroscopy

Different colors of light are waves with different oscillation frequency and correspondingly different wavelength (which is the speed of light divided by the frequency). The characteristic vibration frequencies of molecules are in the infrared (IR) spectrum; the much higher frequencies to which atoms respond are in the ultraviolet (UV). Visible light occupies the intermediate region between UV and IR, to which many substances do not respond. These substances are therefore transparent.

Light frequencies are so large that the numbers are hardly meaningful. For example, water vapor makes air essentially opaque to a broad band of infrared radiation near a wavelength of 6.2 micrometers. This corresponds to a frequency of 5×10^{13} Hertz (cycles per second). It is more usual to specify this frequency as 1600 waves per cm, or a wave number of 1600 cm^{-1}.

Most molecular species have a number of infrared absorption lines, or characteristic frequencies that they absorb strongly. The spectrum of light that has passed through air containing these molecules will show narrow dark bands (spectral lines) due to that absorption. The pattern of lines forms a "fingerprint" that identifies that molecular species.

Raman spectroscopy is another way to measure a molecule's infrared fingerprint. Here light of high frequency (visible or UV) interacts with the molecule, and re-emitted (scattered) light is analyzed. Due to the Raman effect, some of the scattered light will be shifted in frequency by an amount equal to a characteristic frequency of the molecule. With a single-frequency laser source, the returning light will display a spectrum with bright lines corresponding to molecular frequencies—also a fingerprint.

SOURCE: Office of Technology Assessment, 1995.

distinguishing among isotopes of heavy elements, for which the relative changes in vibration frequency due to mass differences are much smaller.

In a mixture of many different molecular species the fingerprints are likely to overlap, obscuring some of the lines and complicating identification. Also, large and interesting molecules tend to have poorly defined spectral features with broader absorption lines than smaller simple molecules. As a result, infrared or Raman spectroscopy can be expected to identify and characterize simple molecules but may not be very sensitive to the large complex molecules important to environmental monitoring. Remote sensing by infrared spectroscopy does offer the possibility of rapidly screening large areas to locate any regions of suspicious activity.

Mass Spectrometry

Chemists use mass spectrometry to identify molecules based on their masses, and are therefore concerned with measuring a wide range of masses. The heaviest atoms weigh about 250 atomic mass units (amu—approximately equivalent to the mass of a hydrogen atom). Interesting organic molecules range up to 10 times this value, and biological molecules can reach several hundred thousand amu. Molecules are broken up by the ionization necessary for mass spectrometry. Thus a single molecular species can give rise to a spectrum of different fragment masses, which define the mass spectral fingerprint for that molecule.

Conventional mass spectrometers (see chapter 3) separate and identify atoms or molecules according to their response to magnetic forces. Ionized molecules are accelerated electrically and follow a narrow path through a magnet, which bends their trajectories: the larger the mass, the smaller the deflection angle. These systems need relatively large samples because the ionizer and molecular throughput are not very efficient.

Improvements in magnetic spectrometer efficiency are discussed below in connection with identifying particular nuclei. In addition, more

sensitive technologies using electric forces have been demonstrated. Ion trap[7] and time-of-flight[8] mass spectrometers do not need a narrow path, and the latter can measure molecules of biological size. These technologies can be combined with laser ionization, which causes less damage to molecules and further increases the efficiency, compared to other types of ionization.

Future mass spectrometers will have greatly improved sensitivity, making this already useful technique much more useful.

Other Tools

Molecules can also be identified by their chemical properties. Liquid and gas chromatography are techniques where the sample is mixed with a carrier that flows through a column to separate mixtures of chemicals; different substances are carried through the column at different rates, depending on how they interact with the surface coating of the column. Thus chromatography separates the sample into fractions that come out of the system at different times. The molecule of interest comes out at a time established by previous tests, and that fraction can be analyzed further. Electrophoresis is a similar technique, where substances are mobilized by an electric current instead of a flow of material across a surface.

These analysis techniques are considered destructive in that they use up the sample, precluding further tests. However, it is possible to construct a surface which selectively attracts some molecules and holds onto them without destroying them. A micromechanical surface-acoustic-wave sensor[9] can compare the density of molecules held by different surfaces, providing still another "fingerprint" that can be used to identify the molecular species (see *Specific Approaches*).

Tools that use chemical interactions require the development of a "cookbook" of recipes, specifying reactants and procedures, before they can be used in practice. Chromatography is an established technique that is already in use for environmental monitoring.

Identifying Isotopes

The nuclear scientist also has a number of tools with which to find nuclides of specific interest. Most of the key nuclides are radioactive, and the radiations they emit can be detected and analyzed (see chapter 3). Mass spectroscopy can determine the relative abundance of different isotopes. Other techniques involve inducing the atoms to emit characteristic radiations. Activation analysis (neutron bombardment resulting in new radioactive isotopes due to transmutation or fission) was covered in chapter 3. There are also optical resonance (scattering of characteristic light frequencies) and induced radiation (emission of characteristic x-rays or gamma rays).

These tools are more specific than those used to identify molecules, and the nuclei they detect are much less ambiguous indicators of a clandestine weapons program. The tools are also less suitable for dual-use. However, some of them have been applied to studying the fossil record of the earth's history by measuring elemental and isotopic abundance ratios, yielding information on various geological and meteorological processes.

Measuring Radioactivity

Counting atomic disintegrations is a mature discipline (see chapter 3), and this chapter is concerned only with advanced state-of-the-art means of doing so. Radioactivity provides a highly sensitive means of detecting atoms or molecules that have previously been selected and concentrated. Its application to capillary electrophoresis and tritium measurement will be mentioned later (see *Specific Approaches*).

[7] Briefing by Aaron Garrett, Los Alamos National Laboratory, Feb. 27, 1995.
[8] Briefing by Charles Wilkerson, Los Alamos National Laboratory, Feb. 27, 1995.
[9] Briefing by Tony Ricco, Sandia National Laboratories, Feb. 28, 1995.

Mass Spectrometry

As already mentioned, the weak point of conventional mass spectrometers is collection efficiency. An instrument with greatly improved efficiency has been developed by combining state-of-the-art technology in isotope separation and ion optics.[10] It has a better ionizer and a broader beam path.

Optical Resonance

Individual atoms can respond strongly to light that is tuned to an atomic transition frequency. For example, extremely small quantities of sodium can be measured by resonant excitation and re-emission of yellow light at 589 nanometers wavelength. This method is very selective for atomic species (nuclear charge); combining it with mass spectrometry to select for nuclear mass provides an absolutely unambiguous identification of a particular isotope. Unfortunately, the strongest responses come from the alkali metals (sodium, potassium, rubidium, and cesium), and the application of this technique to other elements is not obvious.

Induced Radiation

When a solid sample is bombarded by a particle beam (of electrons or ions), or by gamma rays, the atoms and nuclei in it can be stimulated to emit radiations. All except the lightest atoms will produce x-rays characteristic of the atomic species. X-ray tubes use electron bombardment; use of ions or low-energy gamma rays to irradiate samples can give a cleaner signal. In addition, ion bombardment of many of the heavier nuclei causes Coulomb excitation, in which the nucleus is stimulated to rotate and to emit gamma rays characteristic of the nuclear species. These methods are sensitive and selective for most nuclei of interest, and will be applied more often in future instruments.

SPECIFIC APPROACHES

Laser Remote Sensing

The U.S. Department of Energy (DOE) has a program called CALIOPE (Chemical Analysis by Laser Interrogation of Proliferation Effluents). This is a collaborative effort involving five national labs, which are pursuing various ideas for laser remote sensing. It is intended to culminate with an airborne field demonstration of the most promising ideas. Current research efforts include:

- **Enhanced UV Raman Spectroscopy:** A characteristic of the Raman effect is that the return signal is weak. Using ultraviolet instead of visible light can increase the signal by a factor of 1000, and changing (modulating) the laser output and looking for changes in the return can result in suppressing the noise by a similar factor.[11] Ultraviolet light does not travel well through the atmosphere, limiting this technique to distances less than 1 kilometer. Its best application may be on-site characterization (of stack plumes, for example) instead of airborne use.
- **MWIR (Mid-Wave Infrared) DIAL (Differential Absorption LIDAR):** Recent advances in tunable infrared laser technology have made it possible to probe the spectral fingerprint lines of many molecules with a single instrument covering the spectral range from 2.5 to 4.5 micrometers.[12] This instrument could be used at airborne ranges of many kilometers.

Early tests are encouraging, but much development work remains before any of these concepts could be used routinely in the field. The ultimate sensitivity and specificity of these technologies are still to be determined; it is not easy to detect the fingerprints of molecules in trace concentrations, and most molecules, even if detected, are uncertain indicators of clandestine activity.

[10] Briefing by Jane Poths, Los Alamos Scientific Laboratory, Feb. 27, 1995.
[11] Briefing by C. Chen, Brookhaven National Laboratory, Jan. 10, 1995.
[12] Briefing by Lyn Pleasance, Lawrence Livermore National Laboratory, Mar. 1, 1995.

Airborne Measurements

The Department of Energy also has a program called AMPS (Airborne Multisensor Pod System). Three different sensor pods have been built and are being tested on a P-3A aircraft:[13]

1. Synthetic aperture radar;
2. Optical imaging sensors
3. Effluent species identification.

While all three pods are intended to gather information on clandestine activity, only pod 3 will demonstrate technologies for environmental sampling and analysis. Three instruments will sample the air through which the plane flies (which can include an exhaust plume of a suspected facility), with high sensitivity due to the large volume of air sampled, and two additional instruments will view the area within several kilometers of the plane:

- **Real-time radionuclide analyzer:** Collects particles and some vapors on filter cartridges and promptly records their high-resolution gamma-ray spectra. With a cycle time of 1 minute, it can detect short-lived radionuclides with a sensitivity that can reach 5000 atoms per cubic meter of air (for substances with a 4-hour half-life).
- **Ion trap mass spectrometer:** Collects chemical vapors and analyzes them, with a sensitivity that can reach parts per trillion.
- **Whole air sampler:** Collects air components for later analysis.
- **Echelle grating spectrometer:** Analyzes sunlight reflected off the ground to determine the molecular absorption in an exhaust plume.[14] It covers a wide spectral range, from 2.0 to 4.2 micrometers, records continuously over the entire range with very high spectral resolution, and is sensitive to parts per million in a 1-meter-wide plume.
- **IR tracking system:** Correlates an IR signature with the other sensor data.

This program will reduce to practice several technologies already available in the laboratory. A pod carrying multiple instruments is an excellent way to collect airborne data. Experience will tell whether the instruments chosen are the right ones—for example, whether the molecule detectors (ion trap and echelle grating) see any useful signals.

Chemical Identification

Several different advanced ideas for identifying molecules are being developed and improved. They include:

- **Ion trap mass spectrometry:** Catches ionized atoms or molecules in an electrostatic trap where they can be held and measured precisely. Sensitive to parts per billion or less (see above).[15]
- **Time-of-flight mass spectrometry (TOFMS):** Electrically accelerates molecules and measures how rapidly they move, with heavier molecules moving more slowly than lighter ones for a given accelerating voltage. TOFMS can measure the weight of molecules up to several hundred thousand amu in mass.[16]
- **Microsensor arrays:** A micromechanical surface-acoustic-wave sensor is built with a coated chemical reagent that selectively attracts some molecules and holds onto them without destroying them. Since the propagation of sound waves along the surface depends on the density of adsorbed molecules, such a sensor can measure the concentration of the substance that binds to it. An array of these with different chemical surfaces will show different densities of attracted molecules, and

[13] Briefing by Bruce Roberts, Pacific Northwest Laboratory, Mar. 3, 1995.
[14] Briefing by A. J. Ramponi, Lawrence Livermore National Laboratory, Mar. 1, 1995.
[15] Briefing by Aaron Garrett, Los Alamos National Laboratory, Feb. 27, 1995.
[16] Briefing by Charles Wilkerson, Los Alamos National Laboratory, Feb. 27, 1995.

thus offers some hope of identifying the molecular species.[17]

The two electrostatic mass spectrometers have been demonstrated in the laboratory, and they offer significant performance benefits. They also may offer easier portability, because they do not contain magnets. The microsensor array is in the research stage; it seems better suited for monitoring changes in abundant molecules (that is, process control) than for identifying rare ones.

Nuclear Mass Spectrometry

Various improvements to ordinary magnetic mass spectrometry (chapter 3) are under development. They include:

- **Laser ablation source:** Laser light is focused on a single dust particle or a small region, vaporizing only the selected material for input to the mass spectrometer.[18]
- **Improved design:** A mass spectrometer several thousand times more efficient and more sensitive was constructed using modern advances in isotope separator and ion optics technology. It can measure the isotopic composition of atmospheric Kr and Xe from 10-cc air samples, and it can do so accurately enough not only to detect reactor emissions (natural and fission-product Kr and Xe have different isotopic ratios), but to distinguish between uranium and plutonium reactor sources.[19]
- **Accelerator mass spectrometry:** This extremely sensitive technique (chapter 3) is being extended to a large part of the periodic table.[20]
- **Projectile x-ray detection:** The accelerator mass spectrometer can be modified to select for atomic species as well as nuclear mass, by observing the x-rays that result when the selected atoms hit a target at the end of the machine. The result is an unambiguous identification of the nuclide.[21] (Ordinary mass spectrometers do not produce x-rays because the atoms have too low a speed.)

Mass spectrometer designs of the future will draw on these technologies, as well as electrostatic mass spectrometer (see above) and optical detection (see below) technologies, to produce the best instrument for each application. Greater sensitivity and selectivity is the major benefit.

Optical Resonance Spectrometry

Different isotopes of the same atomic species absorb and scatter light at slightly different characteristic frequencies. With extremely precise frequency control, a laser can be tuned to interact selectively with one isotope and detect it. Practical applications include:

- **Photon burst detection:** The atoms pass through a series of detectors while being illuminated with light at the desired isotope's characteristic frequency. The correct isotope will scatter light into each detector as it passes by. The wrong isotope may occasionally scatter light, but will not trigger all of the detectors. This method has demonstrated the ability to detect a rare isotope at a dilution of 10 parts per billion in ordinary krypton, and the current goal is 10 parts per trillion (the natural abundance of radioactive krypton-85). The method may apply to 40 or 50 atomic species, including many important fission products.[22]
- **Optical trap detection:** The atoms are caught and held in a magneto-optical trap, due to the combined effects of resonance light and a magnetic field. Their presence is measured by

[17] Briefing by Tony Ricco, Sandia National Laboratories, Feb. 28, 1995.
[18] Briefing by Brian Andresen, Lawrence Livermore Laboratory, Mar. 1, 1995.
[19] Briefing by Jane Poths, Los Alamos National Laboratory, Feb. 27, 1995.
[20] Briefing by Ivan Proctor, Lawrence Livermore Laboratory, Mar. 1, 1995.
[21] Ibid.
[22] Briefing by H. Oona et al., Los Alamos National Laboratory, Feb. 27, 1995.

the intensity of scattered light. This technique may be limited to the alkali metals.[23]

In the laboratory, both of these methods have been applied to atoms coming out of a mass spectrometer, with the promise of much greater sensitivity and selectivity.

Nuclear Identification

Several miscellaneous ideas for identifying nuclei are being researched. These include:

- **Capillary electrophoresis:** A small instrument that analyzes nanoliter quantities of liquid in minutes; it has been used to separate fission products. In this application, a counter detects the radioactivity of different fractions; other detection schemes are possible. The system offers small size and speed; may be more adapted to process control than to trace element detection.[24]
- **Particle-induced x-rays:** Bombards a filter or swipe containing particles of interest with ions from a high-voltage accelerator. The system can locate a particle and then focus on it. The resulting x-rays and gamma-rays give a nondestructive simultaneous measurement of all atomic species from sodium to the high end of the periodic table, and can also provide some indication of isotope ratios in heavy nuclei.[25] Relatively costly.
- **Laser ionization mass spectrometry of iodine-129:** Improves the efficiency of a mass spectrometer to detect I^{129} by a factor of 1,000 over conventional techniques, by using a laser to selectively ionize the iodine atoms.[26] A special-purpose tool.
- **Automated hydrogen isotope system:** Automated system to extract tritium from air and count it. One measurement every three hours, expected to be sensitive to tritium at background levels (the abundance at which it is found in the absence of a local source: two disintegrations per minute per cubic meter of air).[27] Not yet built.

In addition to technology development, new methods of data analysis are being pursued. "Nuclear Archeology" analyzes the isotopic ratios of impurities which have been irradiated in reactor components. Given a sample of graphite moderator taken from a nuclear reactor (i.e., a research reactor or a gas-cooled power reactor), the method measures the isotopic ratios of impurities such as barium and titanium. This determines how many neutrons were generated over the reactor's lifetime, and therefore its power history. Discrepancies with reported use can indicate that the reactor was used for undeclared purposes, possibly including the production of plutonium.[28] This is a new application of relatively conventional technology.

Thermal Imaging

DOE is also building an experimental satellite that will view the earth in 15 different spectral bands covering the visible and infrared.[29] It will measure local ground and water temperatures to an absolute accuracy of 1 degree K (Kelvin). This should be sufficient to detect the waste heat from a clandestine plutonium production reactor. For example, the flow of cooling water from such a reactor into a river or lake has been modeled and would appear as an easily detected plume of higher water temperature. This is a demonstration program, not a monitoring program, and the satellite is expected to operate for only one to three years.

[23] Briefing by Dave Vieira, Los Alamos National Laboratory, Feb. 27, 1995.
[24] Briefing by Brian Andresen, Lawrence Livermore Laboratory, Mar. 1, 1995.
[25] Briefing by Arlyn Antolak, Sandia National Laboratories, Feb. 28, 1995.
[26] Briefing by Bret Cannon, Pacific Northwest Laboratory, Mar. 3, 1995.
[27] Briefing by Paul Reeder, Pacific Northwest Laboratory, Mar. 3, 1995.
[28] Briefing by Bruce Reid, Pacific Northwest Laboratory, Mar. 3, 1995.
[29] Personal communication with Bob Courtney, Sandia National Laboratories, Apr. 6, 1995.