

LA-UR-13-28412

Approved for public release; distribution is unlimited.

Title: Incorporation of New, Automated Environmental Sampling Systems into Safeguards Approaches

Author(s): Boyer, Brian D.
Anheier, N.
Cable-Dunlop, P.
Sexton, L.

Intended for: Report

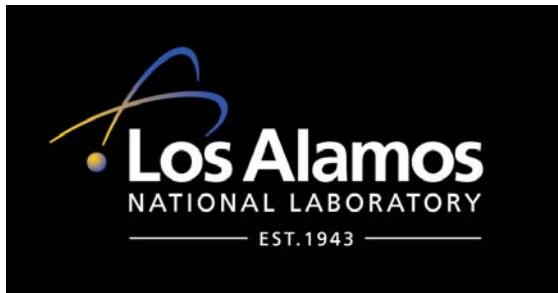
Issued: 2013-11-01



Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.



LA-UR-13-YYYYY

Incorporation of New, Automated Environmental Sampling Systems into Safeguards Approaches

October 2013

B Boyer
Los Alamos National Laboratory
N Anheier
Pacific Northwest National Laboratory
P Cable-Dunlop
Oak Ridge National Laboratory
L Sexton
Savannah River National Laboratory

Sponsored by the
NNSA-NA-241 Office of Nuclear Safeguards & Security: Safeguards Policy

ACKNOWLEDGMENTS

The United States Department of Energy, National Nuclear Security Agency Office of Nuclear Safeguards and Security: Safeguards Policy (NA-241), under the Safeguards-by-Design Project, provided the funding from the Next Generation Safeguards Initiative to sponsor this endeavor. The authors wish to thank NA-241 for its input, support, and guidance in preparing this final report.

Contents

ACKNOWLEDGMENTS.....	3
Executive Summary	6
ACRONYMS AND ABBREVIATIONS	9
1 INTRODUCTION	11
2 Current Safeguards Approach at GCEPs	13
2.1 Current Environmental Sampling at GCEPs	15
2.1.1 ES Swipe Sampling Operations	19
2.2 Current DA at GCEPs	23
3 Description of New Technologies: Their Safeguards Niches.....	27
3.1 Introduction to TRI-ACE	27
3.1.1 TRI-ACE Operational Description	28
3.1.2 Tamper Resistant/Indicating Components.....	29
3.1.3 Evaluation of Active and Passive Collection.....	32
3.1.4 TRI-ACE Safeguards Significance	32
3.2 Introduction to LAARS-ES and LAARS-DA.....	33
3.2.1 Laser Ablation, Absorption Ratio Spectrometry Technology	33
3.2.2 LAARS-ES Sample Collection.....	35
3.2.3 LAARS-DA Sample Collection.....	39
3.2.4 LAARS-ES and LAARS-DA Safeguards Significance	40
4 Safeguards Qualities of the Unattended Systems	41
4.1 TRI-ACE.....	41
4.2 LAARS ES.....	42
4.3 LAARS-DA	43
4.4 Instrument Deployment Strategies.....	44
4.4.1. TRI-ACE Deployment Strategies	44
4.4.2 LAARS-ES Deployment Strategies.....	45
4.4.3 LAARS-DA Deployment Strategies.....	48
4.4.4 Safeguards by Design Issues in GCEPs for TRI-ACE and LAARS Installations	48
4.5 Evaluation of Proposed TRI-ACE and LAARS Use in GCEPs Safeguards vs. Status Quo	48
4.5.1 ES Effectiveness	48
4.5.2 DA Effectiveness	49
5 REFERENCES	51

Figures

Figure 2-1. The Environmental Sampling Process	16
Figure 2-2. Example of a Point ES at a Beam Stop at a Suspected Enrichment Laser Facility in a Training Exercise at Brookhaven in 2010	17
Figure 2-3. Training Example of a Composite ES at a Laser Source at a Suspected Enrichment Laser Facility in a IAEA Training Exercise at Brookhaven in 2010.....	17
Figure 2-4. Cotton Swipe ES Kit	19
Figure 2-5. Assistant Prepares Working Surface (left), and Receives Samples (right)	20
Figure 2-6. Pulling a DA Sample.....	23
Figure 2-7. Temporary Paper Seals on the Sample Cabinet.....	24
Figure 3-1. Standard ACE Sampler	27
Figure 3-2. ACE Two-stage Collection Tube	29
Figure 3-3. Example of a Typical TRI-ACE Collection Plate.....	29
Figure 3-4. TRI-ACE Conceptual Design	30
Figure 3-5. TRI-ACE Prototype Outer Enclosure (left), Components Inside the TRI-ACE Prototype (right).....	30
Figure 3-6. ACE Collection System In-line with the Dispersal System at ORNL	32
Figure 3-7. The Basic Components of the LAARS Instrument (Including a Compact Nd:YAG Laser, Two External Cavity Diode Lasers, the LAARS Sample Chamber, and the Absorbance Detectors. The diagram shows the sample chamber configured to characterize planar samples. The sample chamber can be configured to measure particles directly on the aerosol collector drum).	34
Figure 3-8. LAARS-ES Sample Analysis of a Dissolved Uranium Mineral Containing 2.7 μg Total Uranium. (The spatial map is an XY intensity plot, where the color scale is mapped to combine laser absorbance. The data are discriminated, processed into a histogram plot, and then fit with a Gaussian function. The analysis results provide the number of valid measurements, ^{235}U relative abundance estimate, as well as the standard deviation.)	36
Figure 3-9. The Conceptual Rotating Drum Impactor.....	36
Figure 3-10 RDI Design Parameters Used to Optimize Performance	38
Figure 3-11 Prototype LAARS-ES Rotating Drum Impactor Aerosol Collector	38
Figure 3-12. Conceptual LAARS-DA Gaseous UF_6 Sampling Device Attached to a Conventional Sampling Tap (left). A Modified Commercial Ball Valve (right) and Magnetic Sample holder (below, right) Serve as a LAARS-DA Sampler.....	39
Figure 4-1. TRI-ACE Removable Sample Cartridge.....	45
Figure 4-2. The LAARS-ES Instrument Uses a Stand-alone Environmental Aerosol Sample Collector (right), Housed within a Tamper-indicating Enclosure (left) --.....	46
Figure 4-3. The LAARS-ES Instrument Used to Analyze Uranium Enrichment in Collected Environmental Samples	47

Tables

Table 2-1. ES Laboratory Analytical Techniques.....	21
---	----

EXECUTIVE SUMMARY

The three technologies—tamper resistant/indicating aerosol contaminant extractor (TRI-ACE), laser ablation laser absorbance ratio spectrometry-environmental sampling (LAARS-ES), and LAARS-destructive assay (DA)—have the potential to revolutionize aspects of sampling at gas centrifuge enrichment plants (GCEPs). The environmental sampling capabilities of TRI-ACE and LAARS-ES have the ability to provide unattended and remote monitoring equipment at a GCEP that constantly samples the environment for anomalous uranium particles that are indicative of clandestine enrichment to highly enriched uranium (HEU). This ability would allow the International Atomic Energy Agency (IAEA) to have a nearly continuous scanning presence for potential HEU production at a facility.

LAARS-ES has on-site sample verification possibilities, with an analysis unit concept designed to fit in a standard IAEA tamper-indicating equipment rack. The LAARS-ES aerosol collector will need development of a tampering indicating technology, as planned for the future. This approach can save significant time and effort to obtain assay results from the samples collected on-site, compared to shipping and analyzing samples off-site. An effort to determine if the LAARS-ES analysis unit could analyze the TRI-ACE samples would also be of value. Having the capability to analyze samples on-site and have results in real time for inspectors to formulate actions while on-site would be a big advantage of these systems.

LAARS-DA provides an ability to collect DA samples and analyze them on-site. This is a significant improvement over the present DA sampling process because the storage, tamper-indicating sealing, shipment to analytical laboratories, and delays in analysis beyond the declared timeliness for depleted, natural, or low enriched uranium would be eliminated. The current LAARS-DA concept relies on a LAARS-DA sampler assembly that connects directly to a sampling tap. DA sampling is conducted using steps similar to sampling with a conventional sample bottle. The key issues for success will be the ease of sampling for the facility operator and inspector, collecting a DA sample that is representative of the cylinder enrichment, and transferring and interfacing to the LAARS assay instrument. During on-site inspections, the inspector could retrieve the cartridge and perform enrichment analysis using the on-site LAARS assay instrument. Unique identification of the cylinder and cartridge sample substrate and authentication of the sampling process would need development, too. Such an approach could be considered after the LAARS-DA concept that mirrors conventional bottle sampling was vetted.

The TRI-ACE collection system provides an air monitoring approach that may complement current environmental sampling protocols for IAEA-safeguarded uranium or plutonium bulk-handling facilities. Additionally, this method offers timely collection of airborne materials before they settle on surfaces (that can be decontaminated prior to collection), taken into existing duct work (that can be filtered by plant ventilation), or escape via alternate pathways (i.e., drains or doors). An aerosol collection system such as the TRI-ACE, which could operate in a mode to collect discrete, timed samples or continuously draw samples from the atmosphere of uranium handling areas of bulk

facilities, could improve the probability of detecting undeclared activities with continuous sampling capacity and provide a way to alarm the IAEA to do more expansive swipe sampling to validate the TRI-ACE findings. The TRI-ACE plate will however, need to be sent to an on-site lab or shipped off-site for analysis..

The effectiveness of TRI-ACE and LAARS-ES depends on cost benefits arising from reductions in time to obtain results, lower probability of cross-contamination, increased samples over the course of a year, and sampling at the best locations to detect undeclared HEU production. The key concern with current ES measures has been the cost of sample analysis, cross-contamination issues, and timeliness. If TRI-ACE and LAARS-ES can capture target aerosol particles and the LAARS instrument provides the on-site data that are required to detect undeclared enrichment, then the cost-effectiveness lies in improvements in performance. TRI-ACE samples will need to be shipped to a laboratory off-site, unless adapted for on-site LAARS analysis. However, the tamper-indicating enclosure helps ensure that there are authentic samples that have not been tampered with in the collection plates. While the LAARS analysis instrument will be secured in an IAEA room within the facility, the LAARS-ES stand-alone sampler has yet to develop a tamper-indicating enclosure. Hence, LAARS needs this feature to be effective in an unattended mode. However, the LAARS analysis provides direct on-drum particle measurements, which sidesteps painstaking particle handling and fission track analysis. Since TRI-ACE and LAARS-ES can provide samples tied to location and date and could be analyzed without shipping and contamination, there is a real cost benefit for having samples taken in an unattended mode. The timeliness issue is also a key benefit with LAARS-ES, since this approach eliminates the many months delay before the verification results are known. The current safeguard measures could be better, especially when considering the timeliness for detection of undeclared HEU production is one month. The other key benefit in both systems is the suite of samples, time-dated so that any changes in enrichments could be tracked over time.

A key point about the random and systematic uncertainties in the three systems is that they need to be analyzed in the context of the deployment in an integrated safeguards system. They may turn out to be less accurate than ES swipes and laboratory analysis and provide little improvement on nondestructive assay (NDA) and DA techniques in present GCEPs safeguards. However, if they can provide more timely information and more data points on uranium isotopes in a GCEP, then they may have greater value than ES swipes to the safeguards regime. If these systems act as trip wires, alerting the IAEA to possible GCEP misuse, then a campaign of ES swipes can be planned and a rush analysis at one of the laboratories in the IAEA's network of analytical laboratories (NWAL) can be completed. Furthermore, if the LAARS-DA shows promise in obtaining authentic samples from uranium hexafluoride (UF_6) cylinders, it could make DA samples timely and accurate, avoid sample transport off-site, improve and supplement the present NDA and DA regime and possibly replace the DA sampling mode used now.

The automated systems described in this report should be less intrusive than the continuous enrichment monitor (CEMO), which was on a number of the product cascade headers at the Capenhurst GCEP operated by URENCO. Furthermore, URENCO

has shown more confidence in ES than in the CEMO system because CEMO had a few episodes of delivering false alarms.¹

¹ Conversation of Peter Friend from Brian Boyer of Los Alamos National Laboratory, “Environmental Sampling and CEMO Operation,” April 20, 2012, Capenhurst, UK.

ACRONYMS AND ABBREVIATIONS

AEM	Advanced Enrichment Monitor
AP	Additional Protocol
CEMO	Continuous Enrichment Monitor
CofK	continuity-of-knowledge
DA	destructive assay
DIV	design information verification
DNLEU	depleted, natural, or low enriched uranium
DU	depleted uranium
ES	environmental sampling
GCEPs	gas centrifuge enrichment plants
HPGe	high-purity germanium detector
HEU	highly enriched uranium
IAEA	International Atomic Energy Agency
ITV	International Target Values
LAARS-DA	laser ablation, laser absorbance ratio spectrometry-destructive assay
LAARS-ES	laser ablation, laser absorbance ratio spectrometry-environmental sampling
LEU	low enriched uranium
LFUA	low frequency unannounced access
LPM	liters per minute
NaI	sodium iodide
NDA	nondestructive assay
NWAL	network of analytical laboratories
NU	natural uranium
ORNL	Oak Ridge National Laboratory
PIV	Physical Inventory Verification
PNEM	passive neutron enrichment meter
PNNL	Pacific Northwest National Laboratory
RDI	rotating drum impactor
SAL	Seibersdorf Analytical Laboratory
SEM	scanning electron microscope
SEM/EDS	scanning electron microscope with x-ray microanalysis
SIMS	secondary ion mass spectrometry
SRNL	Savannah River National Laboratory
SQ	significant quantity
TIMS	thermal ionization mass spectrometry
TRI-ACE	tamper resistant/indicating aerosol contaminant extractor
UCVS	Unattended Cylinder Verification System
UF ₆	uranium hexafluoride
UO ₂ F ₂	uranyl fluoride

1 INTRODUCTION

The International Atomic Energy Agency (IAEA) has as at its top level, the following three safeguards objectives. The IAEA strives to detect:²

- undeclared nuclear material and activities anywhere in a State (through environmental sampling [ES] and destructive assay [DA] sample collection)
- undeclared production or processing of nuclear material at declared facilities through ES and DA sample collection
- diversion of declared nuclear material (through DA sample collection).

The IAEA attempts to meet these goals under the framework of the State-Level Approach using safeguards that are fully information-driven.

The fact that every nuclear process, no matter how leak tight, emits small amounts of process material to the environment is the basis for ES for safeguards. With the knowledge that this material can settle on equipment and surfaces within buildings, and can diffuse out of the buildings to deposit on vegetation or soil, or be carried into the water systems, the IAEA and supporting Member States have developed methods to collect and analyze samples. The purpose of ES is to collect samples containing nuclear material emitted from processing operations. Collected samples generally contain only trace nuclear material, below activity thresholds of safety concern. However, analytical laboratory techniques are available with sufficient sensitivity to detect and quantify these extremely low levels of nuclear material at confidence levels required to make safeguards conclusions. The IAEA also attempts to collect nuclear material from declared processes as DA during inspection activities. In some cases these collection modes have been automated for unattended collection and analysis.

Ideally, ES and DA collection and analysis are comprehensive, and are completed at regular intervals or continuously to provide information about current gas centrifuge enrichment plant (GCEP) production activities and to provide peaceful use verification assurances. Manual ES and DA sample collection consumes much inspection effort and expense, which tends to self-limit the sampling dataset. Furthermore, timely detection of anomalies that could indicate the diversion of nuclear material or illicit production is difficult when samples must be shipped off-site to the network of analysis laboratories (NWAL) for analysis. For GCEPs, the timeliness goals are one year for diversion detection of a significant quantity (SQ) of depleted, natural, or low enriched uranium (DNLEU) from the plant and one month for the detection of the undeclared production of highly enriched uranium (HEU). Restrictions on ES and DA sampling because of cost are unlikely to change without increases in IAEA operations budgets or the development of new safeguards technology and adoption of new safeguards approaches.

² V. Bytchkov, “Panel - Challenges Relating to the Implementation of Technologies For Detecting Undeclared Materials and Activities,” *Symposium on International Safeguards Preparing for Future Verification Challenges*, November 3, 2010, Vienna, Austria.

In this study we examined current GCEPs safeguards measures using ES and DA and their ability to meet the safeguards objectives listed above. We examined the tamper-resistant/indicating aerosol contaminant extractor (TRI-ACE) and the laser ablation, laser absorbance ratio spectrometry (LAARS) for environmental sampling (-ES) and for destructive analysis (-DA) technologies and their potential impact to current GCEP safeguards if they were adopted as a future safeguards tools. We analyzed the ability of each technology to meet safeguards objectives, provide gains in effectiveness and efficiency, and affect deployment issues such as intrusiveness or vulnerabilities.

2 CURRENT SAFEGUARDS APPROACH AT GCEPS

The IAEA currently safeguards large low enriched uranium (LEU, <20% enriched uranium-235) GCEPs in several countries. The IAEA has used the same basic approach to safeguard GCEPs that the Hexapartite Safeguards Project recommended in 1983 (Menzel 1983). This approach has seen a few small enhancements for the large European URENCO facilities sited in Great Britain, Netherlands, and Germany, and evolving use of State Level Concepts for these URENCO sites reflecting State-Specific Factors (Everton et al. 2011). Several new GCEPs are under construction and beginning operations in France and the United States, and the three aforementioned URENCO plants in Europe are undergoing continual capacity expansion. The Treaty of Washington (United Kingdom Foreign & Commonwealth Treaty Section 2000) and the Treaty of Cardiff (United Kingdom Foreign & Commonwealth Treaty Section 2007) respectively stated that the URENCO USA plant in Eunice, New Mexico and the Georges Besse II plant in France should be under IAEA safeguards. Furthermore, both treaties stated that the IAEA safeguards should be “equivalent to those applied at the commercial gas centrifuge uranium enrichment facilities under jurisdiction of the Three Governments” (i.e., Great Britain, Netherlands, and Germany).

Implementing the IAEA safeguards objectives at a GCEP facility can be distilled into three principal production and divisions concerns:

- production and diversion of a significant quantity of uranium with enrichment greater than declared (in particular, HEU, where uranium-235 abundance is greater than 20%)
- diversion of a significant quantity of declared uranium (particularly in the form of LEU product)
- production and diversion of LEU in excess of declared amounts (e.g., using undeclared feed).

The detection of undeclared HEU production is of greatest concern, because HEU can be used directly in nuclear weapons. Detection of the diversion or production of undeclared quantities of LEU is also crucial for two reasons. First, LEU can be quickly enriched to HEU either clandestinely in a LEU GCEP or in a separate undeclared facility, both of which are a major concern of the IAEA. Second, natural uranium or LEU can be used to fuel a reactor for the production of plutonium. The Hexapartite Safeguards Project safeguards approach explicitly addresses the first two diversion concerns but does not adequately address the third concern, which centers on undeclared feed. In this scenario, an operator would bypass IAEA inspection and introduce undeclared uranium hexafluoride (UF_6) feedstock into a GCEP. The operator would then remove the undeclared product for use in an undeclared HEU cascade in the same facility or in a separate clandestine HEU enrichment facility. The operator would ensure that his material accountancy would not reveal the undeclared feed, undeclared product, and depleted tails by falsifying the books and ensuring any discrepancies in enrichment values or material amounts would be undetectable by IAEA safeguards (Boyer and Swinhoe 2009). The IAEA moved in recent years to cover this gap in the GCEPs

safeguards approach by rolling out a new model safeguards approach, which includes randomized inspections and the use of the “mailbox” concept as safeguards tools to enable the detection of undeclared LEU operations (Tsvetkov et al. 2007, Boyer and Swinhoe 2010).

The IAEA pioneered the use of the mailbox declaration in the last two decades to verify receipts, production, and shipments at some bulk-handling facilities (e.g., fuel-fabrication plants) (Gordon and Sandborn 1984, Fishbone et al. 1995). The operator declares the status of his plant to the IAEA on a daily basis using a secure mailbox system, such as a secure tamper-resistant computer. The operator agrees to hold receipts and shipments for a specified period of time, along with a specified number of annual inspections, to provide inspector access to a statistically large enough population of UF_6 cylinders and fuel assemblies to achieve the desired detection probability. The inspectors can access the mailbox during randomly timed inspections and then verify the operator's declarations for that day.

A key point related to the technologies under review is that mailbox declarations could hold information on the uranium material properties such as chemical composition and enrichment that automated ES could verify. Commercial enrichment plants exclusively use UF_6 as the feedstock for production of low enriched UF_6 product. While the entire system is at reduced pressure, some UF_6 material is released by processing, failures, and mistakes or the order of $\sim 0.1\text{kg}$ of UF_6 per year at the large GCEPs (Kemp 2010). Trace UF_6 process emissions (i.e., minor leaks within the cascade hall, process service area, and feed and withdrawal area) quickly hydrolyze with water vapor, resulting in micron-size and smaller uranyl fluoride (UO_2F_2) aerosol particles detectable by ES. Inspectors could compare ES data to inspection data and operator mailbox declarations.

IAEA inspectors aim to determine both the uranium-235 enrichment as well as the uranium mass. On-site safeguards verification activities focus on UF_6 cylinders in the declared inventory, since conventional safeguards verification practices can be conducted using gamma enrichment and load cell mass measurements in the cylinder storage yard. Inspectors use nondestructive assay (NDA) and DA sampling plans to verify UF_6 cylinders at prescribed intervals to achieve sufficient statistical coverage of the UF_6 flows over one year of plant operation.

Another focus is on verifying the declared operations of the centrifuges themselves. Through low frequency unannounced access (LFUA) inspections, inspectors access the cascade halls and verify that the designed centrifuge plumbing and layout has not been tampered with to make uranium-235 enrichments outside the declared range. Starting in the 1990s as part of Strengthened Safeguards System measures, the IAEA introduced the use of ES in the GCEP inspection regime to detect if any particles indicative of uranium enriched beyond the declared upper bounds of uranium-235 declared for nominal operation existed at the facility. The IAEA conducts ES using manual swipe sampling and targets UO_2F_2 formed during GCEP process emissions. In this study we will evaluate how TRI-ACE and LAARS-ES could enhance or replace present ES practices as a main goal and whether these technologies can enable new approaches for DA sampling and analysis. Safeguards experts (Hase and Boyer 2012) also see technologies (Miller et al. 2010) such as the Advanced Enrichment Monitor (AEM) passive neutron

enrichment meter (PNEM), and Unattended Cylinder Verification Station (UCVS) combined with load cell monitoring as capable of adding coverage of GCEP uranium processing activities in an unattended mode to verify the mailbox on a random and remote basis. The unattended ES systems could act as a background check on anomalous unreported enrichment beyond declared levels seen by the systems described above. The LAARS-DA could provide bias defect level verification that improves on present IAEA capabilities.

2.1 Current Environmental Sampling at GCEPS

Environmental sampling came into use at the IAEA in the 1990s under Strengthened Safeguards System measures (IAEA 2013) beginning with the collection of environmental samples in facilities and at locations where inspectors have access during inspections and design information verification (DIV) visits. The sample analysis occurred at the IAEA Clean Laboratory and/or at certified laboratories in Member States. With the adoption of the Additional Protocol (AP), IAEA collection of environmental samples at locations beyond declared facility locations, when deemed necessary by the IAEA, became possible. We will focus on the ES at a GCEP as part of the inspection regime and not AP-complementary access at the site. Our tools are envisaged to be part of ES of the GCEP at declared locations and not an AP tool for use in areas of the plant not accessed in DIV visits, low frequency unannounced access (LFUA), or routine inspections. The ES process encompasses the following:

- sampling plan preparation
- collection of samples according to the sampling plan and established procedures
- analysis of the samples to identify any nuclear signatures present
- evaluation of the analytical results to ensure consistency with declared nuclear material or activity.

Figure 2-1 shows the process for ES and how it is linked with the entire safeguards approach for a facility. The two methods of ES collection are known as the point and the composite samples.

The IAEA describes present ES safeguards practices in *Environmental Sampling for Safeguards*, STR-348 (IAEA 2005), for general applications showing proper ES techniques. STR-348 also notes specific GCEP safeguards practices including locations and frequencies. It also describes the use of the Koshelev filter, a special particulate sampling method using installed sample filters (Pasasyuk et al. 2001), designed for the Russian style Shaanxi GCEP (Bukharin 2004) in China.

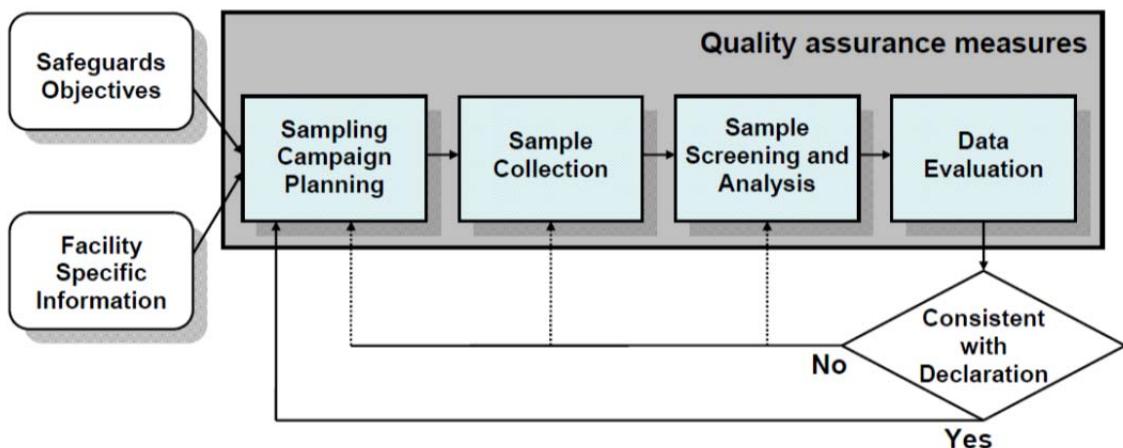


Figure 2-1. The Environmental Sampling Process

The initial IAEA implementation of ES focused on swipe sampling in enrichment plants and in installations with hot cells. The general practice is to implement ES at a facility in two phases—baseline and routine sampling. The baseline phase serves to establish the nuclear signature for a facility and allows for clarification of the nuclear material or activity at the sampling location. The observed nuclear signature should be consistent with the declared activities of that facility, and one or more baseline sampling campaigns may be necessary to adequately characterize the signature of a facility. After the completion of the baseline sampling phase, the sampling at the facility moves to the second phase, routine sampling. Routine sampling occurs periodically at a facility and is used to provide assurances that operations are as declared. Fewer samples are collected during normal routine sampling inspections than during baseline sampling inspections. Routine samples may be taken from locations that were previously sampled during the baseline phase and also from new locations that were not previously sampled.

The point swipe sample, shown in Figure 2-2, involves collection of replicate swipes from contiguous areas at a single location. Point swipe samples are used to characterize activities at a specific location in the facility, such as a glove box or a single piece of equipment. For example, if an inspector enters a GCEP, he could choose to take three point swipe samples: one from the feed and withdrawal area, one from the cascades, and one from the operator's load scale. The IAEA *Safeguards Glossary* defines a point sample as an environmental sample taken in one particular area to characterize one source of released material, which can be found in a contiguous area adjacent to a release point (IAEA 2001, p.73).



Figure 2-2. Example of a Point ES at a Beam Stop at a Suspected Enrichment Laser Facility in a Training Exercise at Brookhaven in 2010

A composite sample, shown in Figure 2-3, involves collection of replicate swipes from contiguous areas at multiple locations. Composite samples are used to characterize operations over a wider area than a point sample. Composite sampling increases the chances of collecting traces of nuclear material that can be analyzed, but information on precisely where the nuclear material is located is lost. Using the GCEP example given, the inspector would take one sample swipe at the feed and withdrawal area, from the cascades, and from the operator's load scale. The IAEA *Safeguards Glossary* defines a composite sample as a sample taken in several areas to characterize multiple sources of a released material that is expected to be found in separate areas more than a few meters apart (IAEA 2001, p. 73).



Figure 2-3. Training Example of a Composite ES at a Laser Source at a Suspected Enrichment Laser Facility in a IAEA Training Exercise at Brookhaven in 2010

The frequency of swipe samples defined for a facility's safeguards approach depends on the facility and the material being handled. Environmental sampling may be used during the following:

- ad hoc inspections at locations where the initial report, and inspections carried out in connection with this report, indicate that nuclear material is present
- routine inspections at strategic points
- DIV during initial visits and periodically as part of inspection activities
- complementary access provided by the State in accordance with the provisions of its additional protocol.

Our analysis focuses on ES conducted during routine inspections at strategic points, since the TRI-ACE and LAARS-ES are designed to be installed at strategic points with a GCEP. According to STR-348 (IAEA 2005), inspectors should consider taking one to three swipe samples per month at large GCEPs like the URENCO facilities in Europe, with less sampling for smaller pilot facilities the size of Iranian and Brazilian facilities.

ES at enrichment facilities strives to provide increased assurance of the absence of undeclared operations (involving enrichments higher than the maximum declared) and handling of undeclared material. In particular, the IAEA desires increased assurance of the absence of undeclared production of HEU. For GCEPs safeguards approaches, ES can be conducted during routine inspections, at strategic points, and for LFUA, where samples can be collected along the LFUA route inside the cascade halls. The TRI-ACE and LAARS-ES deployment requires installation in strategic points most likely to generate target aerosols or at the following key swipe sampling locations with the highest potential to contain traces of past and current operations:

- on or near breakable cascade connections (e.g., sampling stations, service connections, vacuum flanges)
- surfaces around chemical traps
- surfaces in and around sampling stations
- cylinder connection points in cylinder handling areas (e.g., feed stations, tails and product withdrawal stations, sampling, homogenization, blending)
- surfaces of carts used to service the cascades (e.g., sampling, vacuum, calibration, and feed/withdrawal)
- work surfaces and surfaces of tools and equipment (including dismantled equipment) in maintenance and decontamination areas.

During baseline sampling for routine inspections, including LFUA through the cascade hall, swipe samples are collected from the locations listed above. The number of samples taken and the exact sampling points are facility-specific, but should include samples from the cascade hall and areas where nuclear material is handled. During subsequent inspections, swipe samples may be collected at locations randomly selected from the former selected locations. As a general guideline, one to three samples should be collected during a subsequent inspection. Inside the cascade area, samples can be taken during every LFUA. Outside the cascade area, samples can be taken during every inspection. The sampling frequency can be reduced for plants with low capacity; the

number of samples collected can be reduced where extensive baseline data exists. Keep in mind that taking composite swipe samples can be an efficient way to sample several areas, yet minimizes the number of samples collected. For example, an inspector could swipe several points along the LFUA route using one swipe kit.

2.1.1 ES Swipe Sampling Operations

Once the sampling plan is complete and the number and type of samples to be taken have been determined, the inspector requests the sampling kit from two standard sampling tools for swipe sample collections: the cotton swipe and the cellulose swipe.

The cotton swipe is the most commonly used and versatile of the sampling tools. The cotton swipe is a Texwipe® cloth used for swipe sampling at all areas other than inside hot cells or glove boxes. The cellulose swipe is a paper disc with folded tab used for sampling inside hot cells or glove boxes. The cellulose swipe is referred to also as a hot cell swipe, J-type swipe, or J-swipe. This swipe is designed for inserting in a hot cell and handling by remote manipulators. Figure 2-4 shows the cotton swipe kit.

A third standard sampling tool is the Koshelev filter, which is specifically designed for use at the enrichment facility at Shaanxi, China. The Koshelev filter is simply a cellulose swipe that is placed inside a specially designed holder installed at the facility.

TRI-ACE and LAARS-ES will be evaluated against these swipes.

- 1 large outer bag (30×30 cm), with a CL identification number and security seal
- 6 cotton swipes (10×10 cm Texwipes®) bagged in a small mini-grip bag (15×15 cm)
- 6 small mini-grip bags (15×15 cm) to bag the swipes in individual bags
- 6 medium mini-grip bags (20×20 cm) for double-bagging the swipes
- Peel-off labels for samples that are stored in the facility or given to the Facility/State Authorities
- 2 pairs of latex or plastic clean-room gloves (medium size)
- 1 blank working paper *WP EM1* (see Annex 2)
- 1 pen



Figure 2-4. Cotton Swipe ES Kit

The cotton swipe is a piece of cloth used for swipe sampling at all areas other than inside hot cells or glove boxes. The equipment needed for swipe sampling (point or composite swipe sample) consists of one cotton swipe sampling kit, and 0.5 m² of clean aluminum foil or other clean material. The kit's outer bag has a security seal and carries a label indicating the time limit for using the kit. If the seal is broken or if the time limit is exceeded, the kit should not be used.

IAEA ES follows a precise procedure to ensure that the inspectors do not cross-contaminate the samples themselves and that the sample swipes do not get

contaminated during the sampling procedure, storage, and subsequent shipment of the samples to the laboratory.

The inspectors must sample themselves in a pre-inspection check so that swipes of the inspectors' clothes can be checked against the samples taken in the facility. The goal is to have the inspectors go into a facility or site "clean" to do ES. However, an inspector could have been in a research reactor verifying declared 80%-90% enriched metal HEU plates the day before. The inspector could then carry HEU particles on his skin, clothes, working papers, or equipment into a LEU GCEP. These HEU particles could end up on swipes from the LEU GCEP, triggering an anomaly investigation of how and why 80%-90% HEU got in the GCEP cascades. The pre-inspection swipe showing such HEU would explain the anomaly as cross-contamination from a different inspection in another facility using declared HEU.

When the inspectors take their samples in the facility, the team assumes roles. One inspector is the assistant, who is deemed "clean," and the other is the collector, who is deemed "dirty." Both wear gloves. The assistant never comes in contact with the used swipe and the collector's gloves. The team chooses a flat surface as a working surface and puts the kit's aluminum foil down as a work area as shown in Figure 2-5. The collector takes a minimum of four swipes per sampling point or composite, which are returned to the IAEA. The kit holds additional swipes to provide samples for the State authority, if required. Hence, each swipe sampling exercise should collect replicate samples of the site and area swiped for the State if requested. The assistant hands clean swipes to the collector and then holds the small bag to let the collector put the used swipe in the bag as shown in Figure 2-5. The assistant closes the bag and puts the bagged swipe into a medium bag that will be sealed and not opened until received by the analytical laboratory. At the conclusion of the sampling, the assistant places the bags into the original sampling kit bag and the assistant closes that bag. The collector discards all used and unused supplies to prevent cross-contamination. The kit bag is checked for radioactivity and then the inspector transports the samples back to Vienna, or ships them under seal from the site.



Figure 2-5. Assistant Prepares Working Surface (left), and Receives Samples (right)

Observations at the Additional Protocol Exercise course at Brookhaven and conversations with inspectors indicate that it can take the inspectors 15-30 minutes to perform ES depending on the complexity and size of the sampling area combined with the sample-taking plan. The inspectors can take composite samples of a room or facility or multiple samples of specific places in the facility depending on what materials and activities they hope to detect. Seibersdorf Analytical Laboratory (SAL) receives swipe samples, where, depending on the type and number of samples, they are parsed, screened, and shipped to various laboratories in the NWAL. The samples are then analyzed for the presence of radionuclides of interest using the analytical techniques in Table 2-1. The NWAL can process the swipes within two weeks of receipt. However, depending on the backlog and priority of swipes at the NWAL, it can take over a month to get the results back to the IAEA. Hence, a minimum of a month would be needed to ship, process, and analyze ES swipes. Instruments capable of on-site analysis would improve timeliness of analysis and should be cheaper than the above process.

Table 2-1. ES Laboratory Analytical Techniques

Method	Measurement Technique	To Quantify
Bulk	High-resolution gamma spectrometry (HRGS) Liquid scintillation counting Delayed neutron counting Laser-induced luminescence analysis Isotope dilution mass spectrometry (IDMS), thermal ionization mass spectrometry (TIMS) Inductively coupled plasma mass spectrometry (ICP-MS) Accelerator mass spectrometry (AMS)	Gamma emitters Alpha and beta emitters U quantity U quantity U, Pu, Am quantity and isotopes U, Pu quantity and isotopes, other elements ^{129}I , ^{236}U , ^{240}Pu
Particle	Fission track + TIMS Secondary ion mass spectrometry (SIMS) Scanning electron microscope (SEM) Electron microprobe (EMPA)	U, Pu isotopes U isotopes U, Pu, other elements U, Pu, other elements

The analytical techniques used at SAL and NWAL provide very sensitive means to detect nuclear materials and determine isotopic abundance. For GCEPS the primary interest will be determining the relative abundance of uranium-235, a key parameter for detecting misuse and clandestine production of HEU. Anomalies in uranium minor isotope abundance could also reveal undeclared LEU feed or re-enriched tails processing, since, for example, uranium-234 is enriched and depleted along with uranium-235. Feed originating from reprocessed fuel could be detected by the presence of samples enriched in uranium-236, since nuclear reactors produce uranium-236

prodigiously. The identification of these isotopes would likely require point swipes to detect anomalous feed compositions above the feed, product, and tails isotopic background distributions. Any of the automated techniques reviewed here should be able to at least give results accurate enough to alarm the IAEA to do a thorough ES swipe campaign based in the evidence of the automated system. Furthermore, since the automated system would have alarmed the IAEA to anomalous behavior, there would be a reason to rush the swipes through off-site analysis to obtain high-accuracy enrichment confirmation.

When we examine the evidence for the effectiveness of ES at enrichment plants, the IAEA and industry have made some important and strongly positive statements regarding the effectiveness of ES. In 1999 the IAEA and URENCO jointly presented a paper (Cooley et al. 1999) on ES trials conducted at URENCO's E23 GCEP cascade halls in Capenhurst, England, which started operations in 1997. The study began with pre-operation with only natural uranium in the plant and continued through the full production of 5.1% maximum enrichment with a startup product of 3.4% enrichment in the early months of production. The IAEA was able to use ES to successfully capture the history and stepwise increase in enrichment at the facility and to resolve any anomalous particles. An additional paper by Bush et al., of the IAEA in 2001 reiterated these findings and analysis from the 1998 trials and further ES studies in 2000 using thermal ionization mass spectrometry (TIMS) analysis. Friend (2008) from URENCO stated that, "If URENCO were making HEU (of course, URENCO has no intention of doing so!), then there would be a very high chance of finding this out from environmental samples." The acceptance of the utility, effectiveness, and accuracy of ES by URENCO makes it easier to propose an automated ES system versus on-line enrichment monitoring such as the Continuous Enrichment Monitor (CEMO).

The weakness of present ES hinges on the key problems of cross-contamination and timeliness combined with cost, as noted above. Bush stated that the risk of cross-contamination exists and needs to be taken into account in the sampling protocols, in selecting locations for sampling, and in the evaluation of any possible unexpected particle results. In one example, undeclared enriched particles were detected within the GCEP, but were later traced to cross-contamination from an adjacent decommissioned gaseous diffusion plant. The IAEA realizes that single particle results should not be the basis for drawing conclusions, but could trigger further follow-up actions.

Bush also noted that the IAEA demonstrated it can significantly reduce the time required from taking the sample to evaluating measured particle results. However, on a routine basis the IAEA cannot attain its one-month timeliness goal for the detection of the undeclared production of HEU through the application of ES. Furthermore, the costs of performing ES, including the strain on SAL and NWAL capabilities, make widespread use of ES at GCEPs whose number, country of operation, and separative work capacity continue to grow difficult to manage. The cost of an ES has been estimated at around \$1000 per swipe including labor and lab time.³ Hence, there exists a desire for an

³ Gazze C. 2013. Email from C Gazze, International Atomic Energy Agency, to Brian Boyer of Los Alamos National Laboratory, "Environmental Sampling Costs," July 2, 2013.

on-site, automated ES capability that collects target aerosols as they are generated and provides early warning to IAEA of possible facility misuse. This approach could then trigger subsequent detailed ES with priority for analysis after detecting an anomalous result.

Current ES measures have a number of shortcomings, compelling the development of new technologies and approaches that facilitate automated sample collection with reduced cross-contamination potential, reduced costs, and a shorter time to deliver data useful for detecting undetected enrichment activities. We will evaluate TRI-ACE and LAARS-ES for their ability to provide international and regional inspectorates with timely, reliable, authentic, and independent safeguards-relevant samples.

2.2 Current DA at GCEPs

At GCEPS, NDA and DA samples provide material accountancy verification for the UF₆ flows. Sampling plans provide random and optimized selections of the number of NDA and DA samples to provide the needed verification probability for the UF₆. The IAEA safeguards inspection regime uses random sampling with a corresponding level of confidence in detection probability commensurate with the strategic value of the material. For indirect use material such as DNLEU the probability of detection has been set at medium probability equal to 50%. With the advent of State Level Approaches, the IAEA has floated the concept that efforts in nuclear material verification can be reduced because of gains in the use of information from satellites, open source analysis, the Additional Protocol, and acquisition pathway analysis. Hence, the probability of detection for DNLEU could be reduced to 10%-20%, depending on the IAEA's confidence in other complementary measures of verification and analysis (Erpenbeck et al. 2011). In practice with a 50% probability of detection, the IAEA pulls one to three DA samples from GCEPs at monthly inspections. Figure 2-6 illustrates the sample-taking process.



Photo courtesy of P. Friend, URENCO

Figure 2-6. Pulling a DA Sample

Under current safeguards measures, the inspector requests a DA sample from a particular cylinder, most frequently a product cylinder. The cylinder could be selected from the storage yard, material balance area, or while being filled in the feed and withdrawal area. A facility operator transports the cylinder to an autoclave, where the cylinder is heated to provide a homogenized gas sample. The inspector provides the operator a verified and previously weighed empty 2926 sample bottle. Under the constant supervision of the inspector, the operator connects the empty sample bottle to the sampling tap. Inserting the bottle in liquid nitrogen cools the bottle and causes the pressure to drop in the bottle pulling UF_6 gas in the cylinder to be drawn into the bottle, where it desublimates. When the operator draws a sample of 4-8 g UF_6 (~ 500 mg uranium-235, assuming an LEU sample) the inspector takes custody of the bottle, which is then weighed and the net UF_6 calculated. Sample bottles are packaged and tamper-sealed in a cabinet until shipping time (as shown in Figure 2-7). Then they are packed in an approved shipping container such as a 3919A shipping drum, and stored on-site under IAEA seal until it is shipped to SAL for high-precision enrichment assay. Generally the IAEA and the operator ship a whole year's sample batch, the sample sets for the material balance period, once a year to the analysis laboratory.



Photo courtesy of P. Friend, URENCO

Figure 2-7. Temporary Paper Seals on the Sample Cabinet

The DA samples are shipped off-site. Like ES, the IAEA analyzes DA samples using TIMS to measure the isotopic composition and isotopic abundance of uranium. A sample preparation step prior to the actual measurement is required. This consists of the separation of the element of interest from other elements (e.g., matrix materials or impurities). The sample is then deposited onto a filament where the liquid is allowed to evaporate, leaving behind a residue containing the material of interest prior to being introduced into the mass spectrometer. The hot ($>1600^\circ\text{C}$) filament surface atomizes and ionizes the sample. A high voltage potential separates the species U^+ , and subsequently a magnetic field, an electrostatic field or a quadrupole, separates the mass (e.g., $^{234}\text{U}^+$, $^{235}\text{U}^+$, $^{236}\text{U}^+$, $^{238}\text{U}^+$). An appropriate detection system allows the measurement of ratios of ion beam intensities. The isotope abundances are derived from these ratios. TIMS relies on chemically purified samples in order to avoid isobaric interferences. TIMS is therefore very selective and can measure isotope ratios with low uncertainties. The International Target Values (ITV) for Measurement Uncertainties in Safeguarding

Nuclear Materials (Zhao et al. 2010) show that for TIMS for the bias defect measurement uncertainties, (random = $u(r)$ and systematic = $u(s)$) are:

$$u(r) = u(s) = \pm 0.1\% \text{ for LEU } (1\% < \text{uranium-235} < 20\%)$$

$$u(r) = u(s) = \pm 0.2\% \text{ for NU} = 0.711\% \text{ in the range of uranium enrichments } (0.3\% < \text{uranium-235} < 1\%)$$

$$u(r) = u(s) = \pm 0.5\% \text{ for DU } (< 0.3\% \text{ uranium-235})$$

UF_6 DA sample analysis is significantly more accurate than NDA, as reflected in the NDA ITV values:

$$u(r) = \pm 5\% \text{ and } u(s) = \pm 2\% \text{ for LEUF}_6 \text{ using HPGe detector}$$

$$u(r) = \pm 10\% \text{ and } u(s) = \pm 3\% \text{ for NUF}_6 \text{ using NaI detector}$$

$$u(r) = \pm 20\% \text{ and } u(s) = \pm 8\% \text{ for DUF}_6 \text{ using NaI detector}$$

DA is much more precise, but there are some key disadvantages to DA samples:

- intrusiveness to plant operations
- time-consuming inspection effort
- challenge of maintaining continuity-of-knowledge of the samples, as they are collected, stored on-site, and shipped to SAL
- timeliness of results
- need to ship nuclear materials off-site.

DA sampling is quite intrusive to plant operations and time-consuming to the inspection effort. The entire exercise takes planning and time for both the inspector and the operator. The cylinders that are set for inspection either for flow verification or static inventory (static inventory is done during the Physical Inventory Verification [PIV] and mostly involves verifying the tails cylinders that can accumulate on-site.) Those cylinders are set in locations where access to NDA equipment is possible and need little intervention from the plant staff to arrange the cylinders for inspectors to take measurements. These measurements take the inspectors around five minutes to complete. However, pulling a DA sample can take an hour or more of an inspector's time plus the operator's time to position the cylinder in the sampling station and draw the actual sample under the inspector's gaze.

The challenge of maintaining continuity-of-knowledge (CofK) of the samples is a key issue that is not always reviewed. The inspector must maintain CofK from the time he weighs the empty sample bottle until the sample arrives at the off-site analytical laboratory. The inspector must observe the filling of the bottle by the operator after he hands the bottle to the operator. He will then reweigh the bottle to get the net UF_6 mass in the bottle. The inspector must then store the bottle until the sample shipment to the analytical laboratory. The containment and surveillance measures (tamper-indicating seals in this case) and the procedures of the inspectors for the bottles must be robust enough to avoid loss of CofK. While it may be possible for an inspector to recover a year's worth of DA samples at the PIV numerically, it would not be acceptable statistically because the cylinder population at the PIV would not reflect the cylinder

and material flow from the previous 11 months of operation and inspections. The loss of CofK by either a broken seal or a lackadaisical application of seal (bad knot, improper capping of the seal, mixing up the top and bottom of two different seals, etc.) could void a whole year's DA sample set.

The timeliness and escalating cost to reach a safeguards conclusion is linked to the requirement to ship samples to an off-site laboratory. With one-year timeliness verification goal for DNLEU, there is not the urgency for results as with ES for undeclared HEU production detection within one month. However, because of the time to ship and analyze the DA samples, even a one-year timeliness goal for DNLEU can slip by a few months waiting for laboratory analysis results. With escalating enrichment capacity this situation is unlikely to change without development of new safeguards technology and approaches that significantly reduce the costs and improve the timeliness of GCEP DA sample collection and analysis. Such technologies and approaches must feature less-intrusive and automated sample taking, maintain authentic and tamper-indicating samples, and provide results on-site or facilitate ease of shipping off-site. An attractive concept is on-site DA sampling and analysis. We will evaluate TRI-ACE and a variation of LAARS—LAARS-DA—to determine whether these technologies offer improved GCEP DA safeguard approaches.

3 DESCRIPTION OF NEW TECHNOLOGIES: THEIR SAFEGUARDS NICHES

The two technologies under examination are TRI-ACE and LAARS. This section will examine their function and applicability to safeguards.

3.1 Introduction to TRI-ACE

Savannah River National Laboratory (SRNL) and Oak Ridge National Laboratory (ORNL) researchers have been working on the extended development of an air monitoring approach that would allow for continuous sample collection at IAEA safeguarded bulk handling facilities. The principal sample collection device referred to herein is a patented Aerosol Contaminant Extractor (ACE). This device, shown in Figure 3-1, is roughly the size of a desktop computer, and uses electrostatic precipitation principles to deposit particulates onto selected substrates. This technique is applicable for the quick turnaround analysis of samples collected inside operational facilities and may serve as a complementary method to the gold standard environmental sampling protocol used by the IAEA. This device has already proven effective in the collection of fuel cycle signatures including an operational demonstration conducted at the GCEP in Capenhurst, England.



Figure 3-1. Standard ACE Sampler

SRNL and ORNL researchers have been working to incorporate tamper-resistant and tamper-indicating components into the standard ACE system. Incorporation of tamper-resistant/indicating (TRI) technologies will allow continuous collection of samples at uranium/plutonium bulk handling facilities and enhances the probability that sample integrity has not been compromised during collection in the absence of a safeguards inspector. TRI components will indicate if any tampering has taken place and deter attempts to alter the samples by the operator because of the threat of tamper detection. A TRI-ACE has the potential to be an important addition to the international nuclear safeguards inspector's toolkit when developing advanced safeguards approaches for the IAEA.

3.1.1 TRI-ACE Operational Description

The TRI-ACE operates by pulling air (aerosol) and particles suspended (contaminants) in the air through a non-metallic flow tube via a muffin fan at the rate of approximately 300 liters per minute (LPM). The flow tube has two sections (Figure 3-2). The first section creates a negatively charged particle by applying a negative 8.5 KV to a corona wire mounted axially in the middle of the air flow. The corona wire passes through a grounded metallic tube mounted on the inside perimeter of the section, which creates the negative electrostatic field. The second section collects the negatively charged particles on two positively charged silicon (Si) substrates. The two Si substrates (positive 8.5 KV) are mounted in the air stream with two adjacent ground plates affixed to the inside edge of the flow tube. This creates the second electrostatic field. These ground plates repel the negatively charged particles which deposit onto the positively charged substrates (Figure 3-3). The collection plates may then be analyzed directly for morphological/elemental and isotopic composition without chemical processing of the sample via scanning electron microscope/energy dispersive x-ray spectroscopy (SEM/EDS) analysis or secondary ion mass spectrometry (SIMS) analysis, respectively. Conversely, collection plates may be chemically processed with conventional actinide extraction techniques and analyzed for isotopic concentrations by inductively coupled plasma mass spectrometry or TIMS.

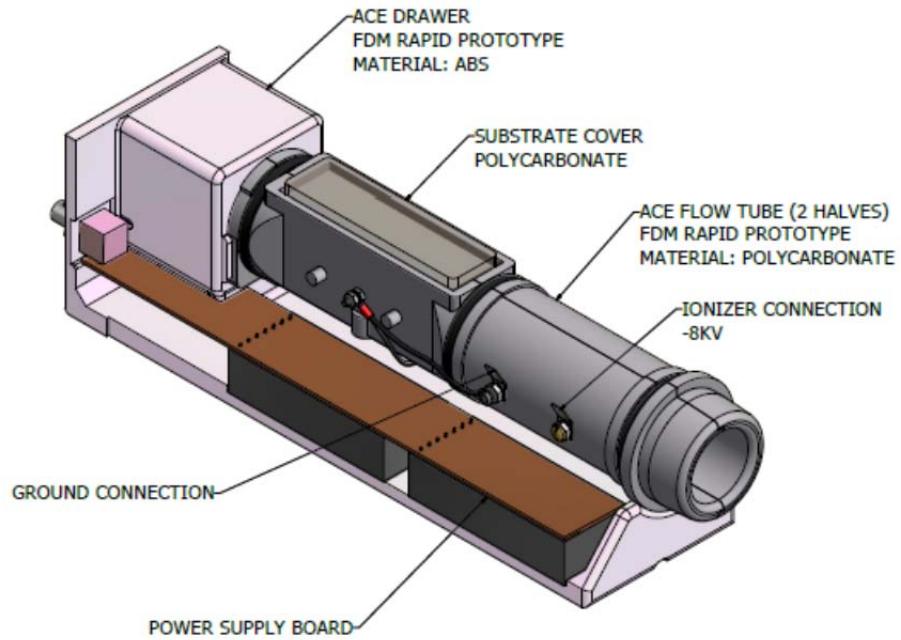


Figure 3-2. ACE Two-stage Collection Tube



Figure 3-3. Example of a Typical TRI-ACE Collection Plate

3.1.2 Tamper Resistant/Indicating Components

TRI-ACE is equipped with many tamper-resistant and tamper-indicating components, which are depicted in the conceptual design in Figure 3-4. The TRI-ACE prototype is shown in Figure 3-5.

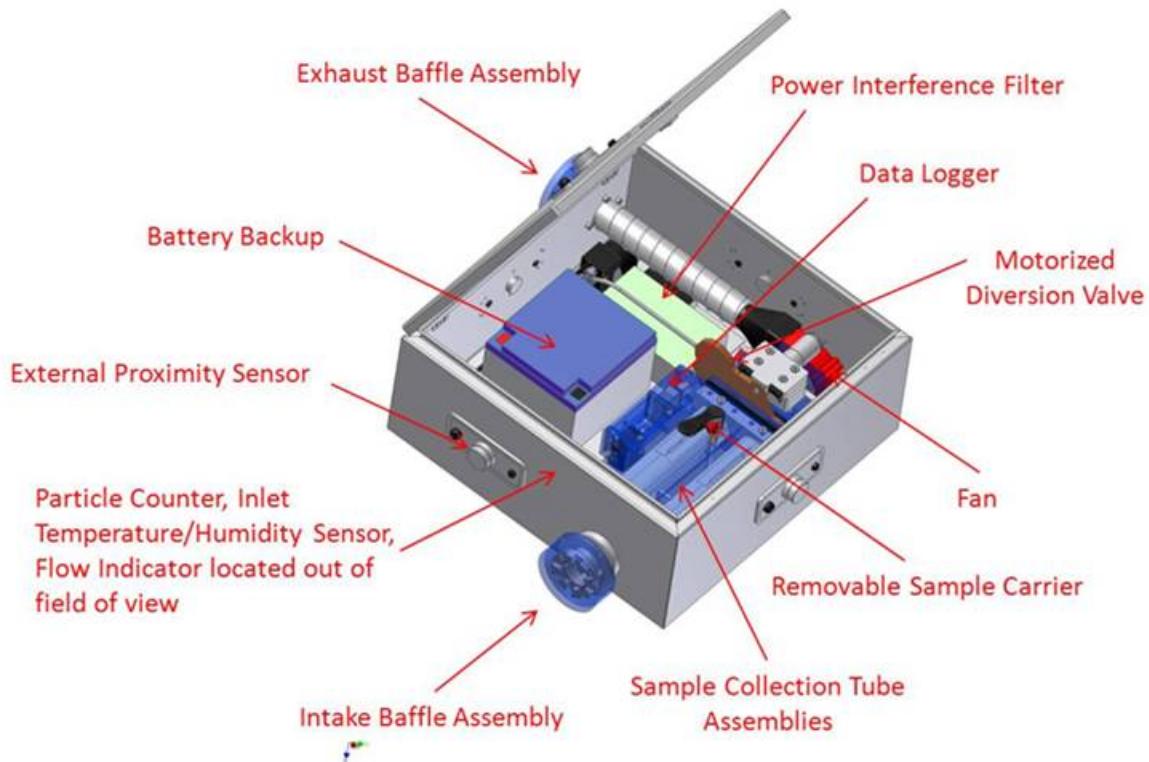


Figure 3-4. TRI-ACE Conceptual Design



Figure 3-5. TRI-ACE Prototype Outer Enclosure (left), Components Inside the TRI-ACE Prototype (right)

The tamper resistant/indicating features are described as follows.

- Particle Counter – A particle counter is installed within the housing at the air inlet prior to the ACE flow tubes. The particle counter will pull a stream of air from the inlet sample flow and will divert sample flow to the alternate ACE flow tube during abnormal particle counts or size.
- Temperature/Humidity Sensor – A temperature/humidity sensor is installed within the housing at the inlet prior to the ACE flow tubes and one internal to the unit. This

sensor detects and records temperature and humidity differences between the interior and exterior of the TRI-ACE housing.

- External Proximity Locator – Several ultrasonic sensors detect and record placement of objects near or around the TRI-ACE.
- Vibration/Tilt Detector – Vibration and tilt detectors within the housing detect and record sampler movement and shock.
- Voltage/Current Data Logger – The voltage and current data logger will record fan power data. Variations of voltage and current will occur if the fan inlet or outlet is obstructed.
- Air Flow Sensor – An air flow sensor at the inlet prior to the ACE flow tubes will detect and record variations in air flow resulting from obstructions on the inlet or outlet.
- Ambient Light Detector – An internal light detector records changes when the enclosure is opened or closed.
- Magnetic Field/Flux Detector – This device detects and records the magnetic field close to the housing. A magnetic field could sabotage electronically stored data.

3.1.1.1 Housing and Size of TRI-ACE Unit

The TRI-ACE sampling system housing is a keyed and sealed stainless steel 51 cm x 51 cm x 20 cm Hoffman enclosure (Sexton et al. 2012). For safety, when the access door is opened, the unit de-energizes. The sampler inlet and outlet barriers installed on the exterior of the enclosure prevent water infusion.

3.1.1.2 Data logging

TRI-ACE has a fully programmable embedded controller that queries, monitors, and time stamps data received from each of the featured devices. Normal parameters will be established and any abnormalities will be flagged. The system stores data on a thumb drive embedded within the flow tube assembly. The thumb drive can be removed easily for analysis.

3.1.1.3 Data Analysis and Reporting

The collection plates located in the flow tube assembly may be analyzed directly for elemental and isotopic composition, without processing the sample, via SEM analysis or with sample processing via mass spectroscopy. Data collected from the thumb drive can also be analyzed for any abnormalities detected by the tamper-indicating components during the collection period.

3.1.1.4 Power Requirements

The input power range for the TRI-ACE is 90-250 VAC, 50/60 Hz, single phase, making for ease of international deployment. The TRI-ACE will normally be powered by the facility. An internal battery backup system will allow the unit to run without facility power for a period of at least 72 hours. Intermittent system operation will be an option. A power interference filter will be installed so the unit will not be damaged in case of power deviations or spikes.

3.1.3 Evaluation of Active and Passive Collection

Recently, the collection performance of the base TRI-ACE system was tested in a controlled uranium dispersal system at ORNL to assess the active deposition collection efficiency relative to that of passive deposition sampling (swipe method) (Trowbridge et al. 2011). An ACE sample collector was placed in-line with the dispersal system (Figure 3-6) for active collection, while layout samples of various materials were placed inside the dispersal system to evaluate swipe sampling. Two distinct uranium exposure scenarios were run to assess collection efficiency. The capture efficiency of the ACE was tested under low humidity (10%) and high humidity (70%) conditions inside the dispersal unit.



Figure 3-6. ACE Collection System In-line with the Dispersal System at ORNL

The active collection sample loadings on the TRI-ACE plates were more than 1000 times greater than the passive deposition samples. The capture efficiencies appear to be slightly greater at higher relative humidity for both TRI-ACE and standard ACE collectors. These findings illustrate the advantage of active collection in pulling in significant samples of materials of interest.

3.1.4 TRI-ACE Safeguards Significance

The TRI-ACE collection system provides an air monitoring approach that may complement current environmental sampling protocols for IAEA-safeguarded uranium or plutonium bulk-handling facilities. Additionally, this method offers timely collection of airborne materials before they settle on surfaces (that can be decontaminated prior to collection), taken into existing duct work (that can be filtered by plant ventilation), or escape via alternate pathways (i.e., drains or doors). An aerosol collection system such as the TRI-ACE, which could operate in a mode to collect discrete, timed samples or continuously draw samples from the atmosphere of uranium handling areas of bulk facilities, could improve the probability of detecting undeclared activities with continuous sampling capacity and provide a way to alarm the IAEA to do more expansive swipe sampling to validate the TRI-ACE findings.

Incorporation of tamper-resistant and tamper-indicating technologies into the ACE system will allow continuous collection of samples at uranium or plutonium bulk-handling facilities in a manner that ensures sample integrity. Tamper resistant/indicating components could deter attempts to alter the samples by the operator and indicate if any such tampering has taken place. A TRI-ACE could be an important addition to the international nuclear safeguards inspector's toolkit when

developing advanced safeguards approaches for the IAEA. In addition, a TRI-ACE could be used by domestic regulatory authorities to confirm the declared enrichment values at GCEPs and verify that enrichment levels are not only within the bounds set by safeguards agreements but also domestic legal regulations for facility operations.

3.2 Introduction to LAARS-ES and LAARS-DA

LAARS is a new verification measurement technology under development at the U.S. Department of Energy's (DOE's) Pacific Northwest National Laboratory (PNNL). LAARS uses three lasers to ablate and then measure the relative isotopic abundance of uranium compounds. An ablation laser tightly focuses on uranium-bearing solids to produce a small atomic uranium vapor plume. Two collinear wavelength-tuned spectrometry lasers transit through the plume. Measurements of the absorbance of uranium-235 and uranium-238 isotopes determine uranium-235 enrichment. The measurement is independent of chemical form and degree of dilution from nuisance dust and other materials. LAARS has high relative precision and detection limits approaching the femtogram range for uranium-235. Scanning and assaying the sample point-by-point at rates reaching 1 million measurements per hour enables LAARS to detect and analyze uranium in trace samples. The spectrometer, assembled using primarily commercially available components, features a compact design and automated analysis.

Two specific GCEP applications of the spectrometer are under development: 1) LAARS-Environmental Sampling (ES), which collects and analyzes aerosol particles for GCEP misuse detection; and 2) LAARS-Destructive Assay (DA), which enables on-site enrichment DA sample collection and analysis for protracted diversion detection. The two applications propose significant technological advances in GCEP safeguards verification.

3.2.1 Laser Ablation, Absorption Ratio Spectrometry Technology

The LAARS instrument uses a miniature commercial pulsed Nd:YAG laser source to vaporize a pinpoint region of the sample, as shown in Figure 3-7. The sample chamber contains windows for the ablation laser beam and entrance and exit of the probe diode laser beams. LAARS samples are acquired as collected aerosol particles, wipes, or adsorbed UF_6 vapor, then loaded into the reduced-pressure sample chamber. The vaporized sample (and substrate) material is ejected from the sample surface to form a high-temperature ($\sim 50,000$ K) plasma through interaction with the intense ($\sim 32 \text{ GW/cm}^2$) pulsed laser radiation. Laser vaporization of the sample serves two important functions. First, the high-temperature plasma effectively atomizes and ionizes most of the uranium sample, regardless of composition (e.g., UO_2 , UO_2F_2). No further manipulation or chemical processing is required. The focused ablation laser spot size also defines the sampling spatial resolution and the subsequent stepwise scanning leads to high spatial resolution isotope analysis across the entire sample surface. This characteristic provides LAARS-ES with the ability to detect and analyze trace assemblages of target particles intermixed in a large excess of background particles (Bushaw and Anheier 2009).

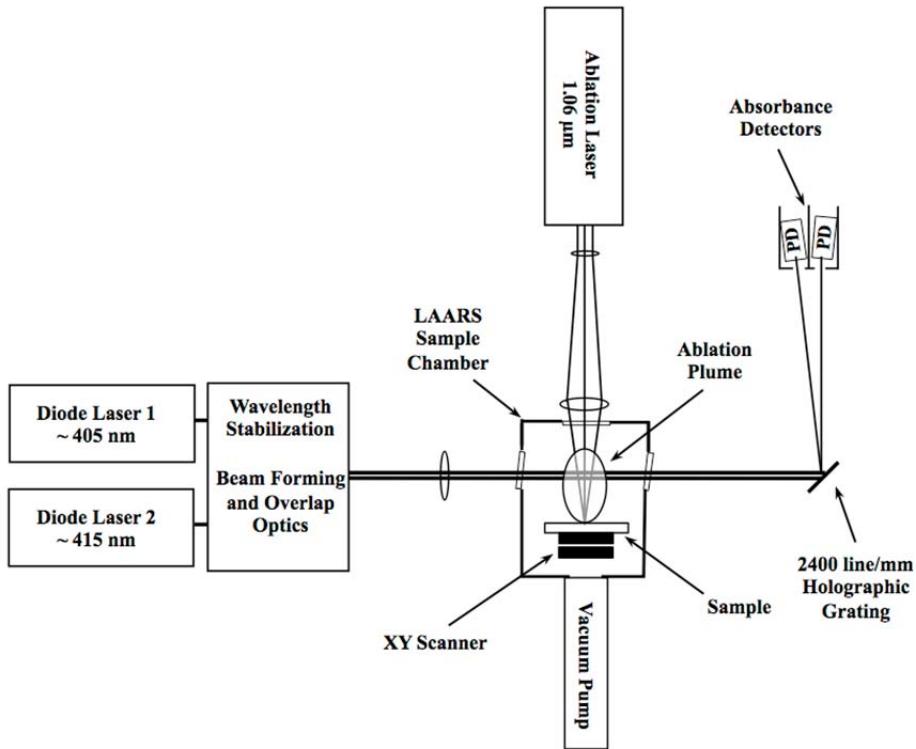


Figure 3-7. The Basic Components of the LAARS Instrument (Including a Compact Nd:YAG Laser, Two External Cavity Diode Lasers, the LAARS Sample Chamber, and the Absorbance Detectors. The diagram shows the sample chamber configured to characterize planar samples. The sample chamber can be configured to measure particles directly on the aerosol collector drum).

Shortly after vaporization, the plasma quickly ($\sim 1 \mu\text{s}$) cools through supersonic expansion and more slowly ($\sim 10 \mu\text{s}$) through conduction with the surrounding cover gas (argon, $\sim 1300 \text{ Pa}$) to form a $\sim 3\text{-mm-diameter}$ hemispherical plume containing neutral uranium atoms. Two wavelength-tunable diode lasers, having linewidths $< 5 \text{ MHz}$, are directed through the plume of neutral uranium atoms and selectively probe (via precise wavelength tuning) the uranium-235 and uranium-238 isotopes. The lasers excite the atoms from the same ground or first excited state to different upper atomic transitions for each isotope. Each laser beam is then directed to a compact photodetector that measures the transmitted laser power. Comparison of the power immediately prior to the ablation pulse with the intensity at the time of maximum atomic column density, typically $15 \mu\text{s}$ after the ablation pulse, yields a precise absorbance signal that is directly proportional to the atom concentration of each isotope. Processing the measured absorbance signals provides the uranium-235 relative abundance determinations each time the ablation laser fires.

Under LAARS pressure and plasma measurement conditions, the isotope linewidths are nominally $\sim 600 \text{ MHz}$ full-width half maximum (FWHM) and the uranium-235 isotope shift is $\sim 11 \text{ GHz}$ relative to uranium-238. The isotope shift is because of the subtle

differences in atomic energy levels that arise from differences in each isotope's nuclear mass and volume. Unlike laser-induced breakdown spectroscopy (LIBS), the narrow probe laser linewidths and large separation between probe wavelengths (two different atomic transitions are excited) are distinctive to LAARS analysis, providing resilience to major-minor isotope channel crosstalk, which would otherwise skew the enrichment result. A laser frequency locking technique (under development) provides frequency stabilization for both probe lasers to <10 MHz, and absolute frequency locking of both lasers to the center of the respective isotopic transitions. This technique can provide uranium enrichment DA to within $\pm 0.1\%$ accuracy.

A custom collector drum (Section 3.2.2) can be integrated with the LAARS assay instrument to directly interrogate the surface of the collector drum. This configuration is called LAARS-ES. Alternatively, a purpose-built sampler assembly (Section 3.2.3) can collect samples, such as DA samples, on planar substrates. The on-site LAARS assay instrument can measure the collected DA samples directly. This configuration is called LAARS-DA. In either mode, the sample substrate (drum, planar) is scanned at the focal plane of the focused LAARS ablation laser. The laser pulse repetition synchronizes to the sample scan to collect ~ 200 sample measurements per second. At this sampling rate, 120,000 discrete enrichment measurements can be completed in 10 minutes. A typical LAARS analysis result is shown in Figure 3-8. Further details of LAARS are available in Anheier et al. (2011, 2012), Bushaw and Anheier 2009, and Anheier and Bushaw 2010.

The LAARS analysis instrument is intended to be deployed on-site within a secure IAEA room. The entire balance of the instrument could easily fit into a standard instrument rack (~ 200 cm x 100 cm x 100 cm) and require <10 amps at 110 VAC single phase.

3.2.2 LAARS-ES Sample Collection

The LAARS-ES environmental aerosol collector, based on a rotating drum impactor (RDI) design, is shown in Figure 3-9. The RDI was originally developed for remote long-term deployment to collect atmospheric particles for subsequent chemical analysis, and it provided atmospheric science interests, such as global climate change with a means to measure chemical concentrations in the atmosphere (Lundgren 1967, Bench et al. 2002, Vanderpool et al. 1990). The RDI has a nozzle width of 1 mm wide by 100 mm long (i.e., the drum width and the particle impaction strip will have about the same dimensions as the nozzle). The circumference of the drum is about 320 mm.

In our design, a rectangular impactor nozzle directs particle-laden air onto the impactor drum surface. Particles having a specific size range are impacted and stick onto the surface of the drum, while the smaller particles are carried by the airflow around the drum to the exhaust.

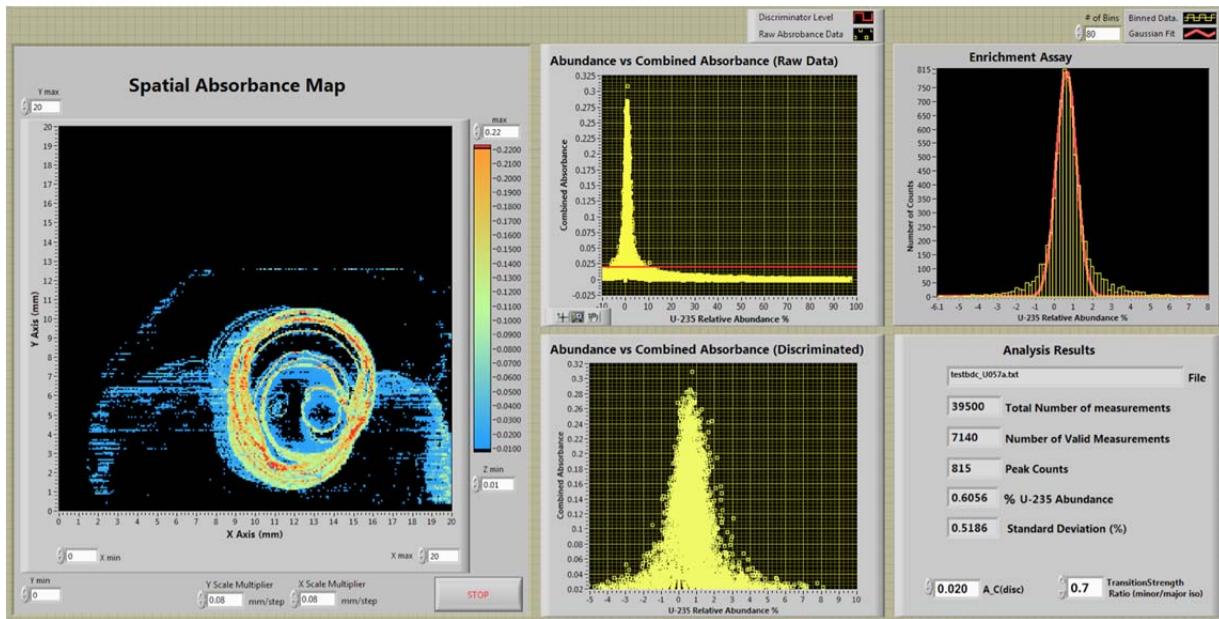


Figure 3-8. LAARS-ES Sample Analysis of a Dissolved Uranium Mineral Containing 2.7 μg Total Uranium. (The spatial map is an XY intensity plot, where the color scale is mapped to combine laser absorbance. The data are discriminated, processed into a histogram plot, and then fit with a Gaussian function. The analysis results provide the number of valid measurements, ^{235}U relative abundance estimate, as well as the standard deviation.)

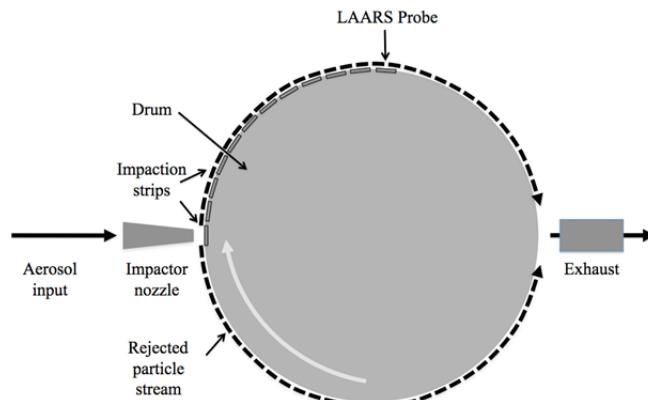


Figure 3-9. The Conceptual Rotating Drum Impactor

The LAARS-ES collector captures airborne particles for a preset time interval, at which point the drum rotates to expose a new rectangular strip on the drum. A drum having sufficient diameter and height could provide time-resolved particle sampling and integration on a daily or weekly basis over a period of one year. The drum could also be segmented along the axis to isolate the collected samples into three equivalent regions, one each for the on-site LAARS measurement, off-site analytical laboratory analysis, and for host state confirmation if required or desired.

The LAARS-ES collector targets UO_2F_2 particles that are produced during atmospheric hydrolysis of trace UF_6 unintentionally released during GCEP operations. These particles are very small (i.e., on the scale of around 100 nm or more) when formed and tend to agglomerate into micron-sized particles over time. It is difficult to capture smaller particles ($< 1 \mu\text{m}$), since they do not have enough inertia to strike the impaction surface, but rather follow the airflow around the drum surface to be exhausted from the collection system. Particles larger than about 2 microns are composed mostly of nuisance dust. These particles can be diverted from the collector using a cyclone trap.

Optimizing several RDI design parameters can increase the capture efficiency of small particles, while decreasing the flowrate requirements. ANSYS FLUENT⁴ computational fluid dynamics (CFD) software was used to optimize the impactor nozzle width, gap between the nozzle exit and the drum surface, and the flowrate through the RDI. Figure 3-10 shows a cross-section of the RDI with these design parameters and Figure 3-11 shows the actual RDI assembly components.

⁴ ANSYS, Inc., Canonsburg, Pennsylvania, <http://www.ansys.com/>

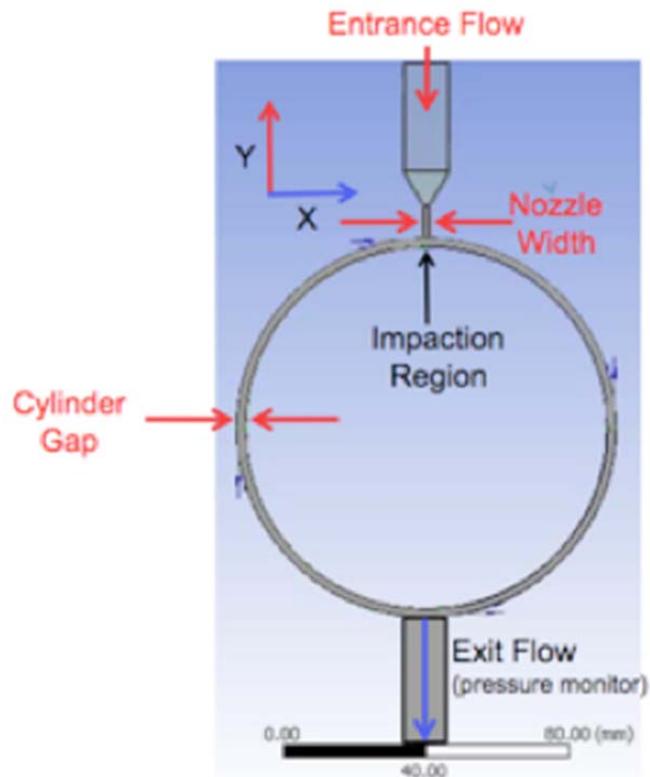


Figure 3-10 RDI Design Parameters Used to Optimize Performance

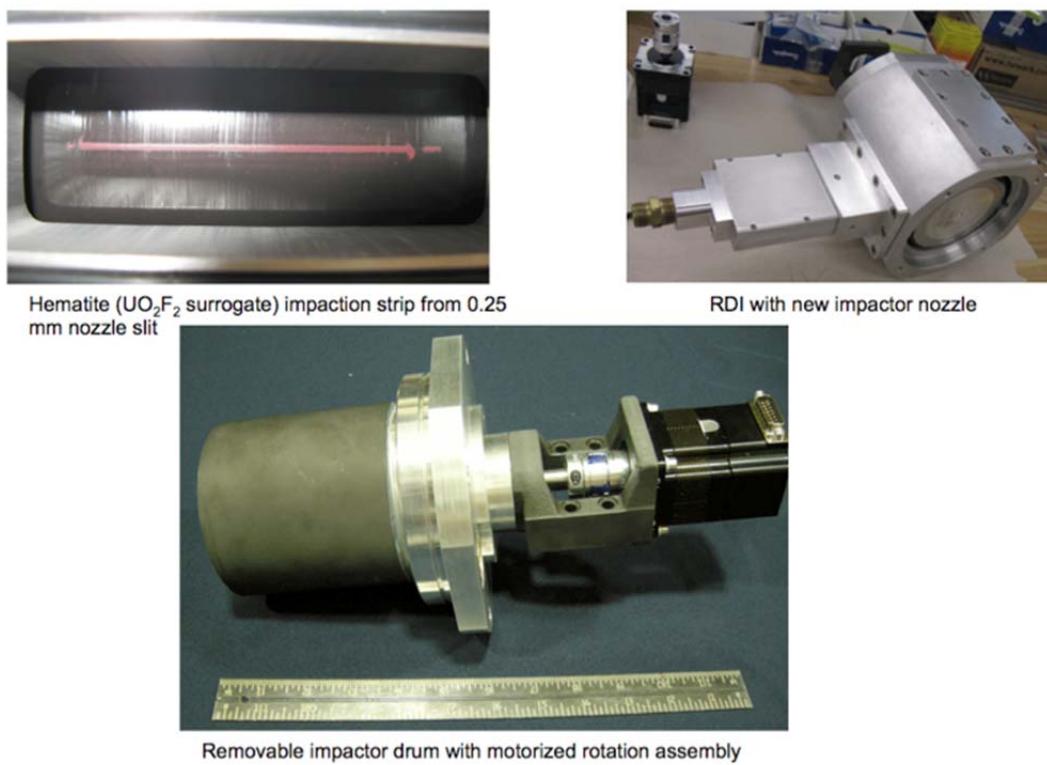


Figure 3-11 Prototype LAARS-ES Rotating Drum Impactor Aerosol Collector

3.2.3 LAARS-DA Sample Collection

The LAARS-DA sampler, shown conceptually in Figure 3-12, is designed to collect DA samples directly from a conventional sampling tap. The LAARS-DA sample assembly is based on a modified commercial vacuum ball valve and can be thought of as a hand-held sampler. The ball valve serves both as an isolation valve and a DA sampling device. A rare-earth magnet retains the DA substrate within the modified valve ball. After connection to the sampling tap, the ball valve is rotated to expose a thin film-coated substrate to gaseous UF_6 . The ball valve is next rotated to isolate the sampler and then removed from the sampling tap.

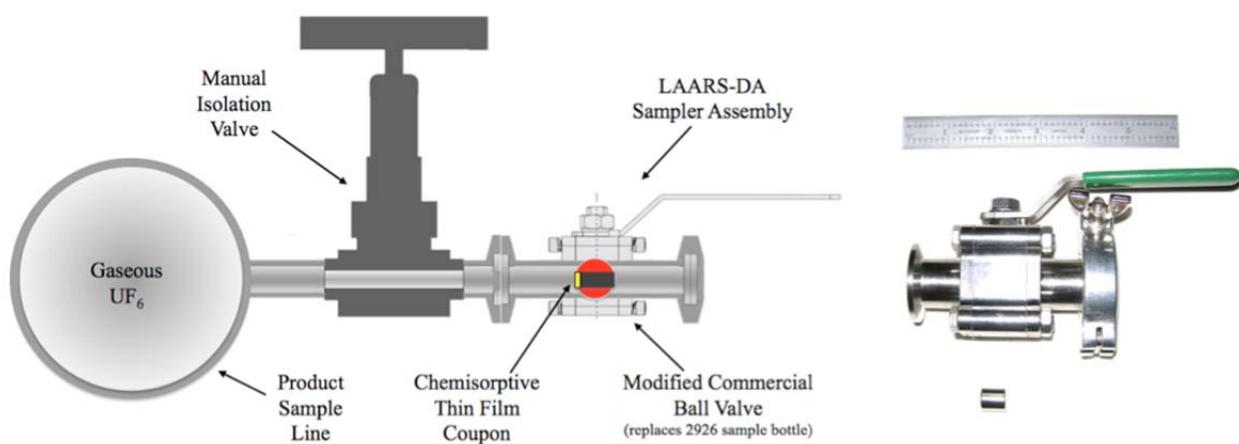


Figure 3-12. Conceptual LAARS-DA Gaseous UF_6 Sampling Device Attached to a Conventional Sampling Tap (left). A Modified Commercial Ball Valve (right) and Magnetic Sample holder (below, right) Serve as a LAARS-DA Sampler

A key component of the LAARS-DA concept is the sorbent thin film. There are several materials that exhibit selective gaseous UF_6 chemisorption, such as sodium fluoride. Gaseous UF_6 quickly reacts with solid NaF to form a stable, solid complex ($2NaF-UF_6$) (Bush et al. 2001, Friend 2008). The sorbent capacity and reaction kinetics are likely different between the granular NaF sorbent bed media and a NaF thin film, but we expect that only short exposure timescales (1 to 5 minutes) and small gas volumes (~ several μ l at 10 torr) are needed to collect microgram sample quantities required for LAARS-DA analysis.

After a short exposure period, an on-site LAARS-DA instrument receives the sampler. The sampler attaches to a sample port and the sample substrate is conveyed into the LAARS instrument using a magnetically coupled sample transporter. The LAARS instrument then quickly scans the DA sample to determine the relative uranium-235 abundance. There may be issues in the authenticity of unattended sample collection from the product line. As noted above, the current IAEA DA sampling relies on inspector presence to provide a chain-of-custody of the sample from the chosen item to the sample container. If an operator has to connect or remove the sample from the autoclave as he does with his own samples without inspector presence, the lack of IAEA knowledge of the authenticity of the 1) cylinder identification and 2) sample history

will be problematic. Furthermore, taking samples from tails and feed cylinders will be more difficult, but necessary. If the LAARS-DA can draw a sample from the same sampling tap as present IAEA samples for all three uranium strata, then the inspector can observe LAARS sample collection as it does present DA sampling. The big advantage for this mode would be the evaluation of samples on-site, which avoids the issues of secure storage, continuity-of-knowledge, and shipping and analysis off-site).

Future LAARS-DA deployment concepts could include an unattended, automatic DA sampling system. For example, a cartridge, containing many multiple sampling substrates, could randomly collect DA samples during product withdrawal. During on-site inspections, the inspector could retrieve the cartridge and perform enrichment analysis using the on-site LAARS assay instrument. This deployment concept would require a higher level of tamper-resistance and tamper-indicating measures. Also it would be more complicated than ES, since it would require a DA sample link to a specific batch of material contained in a specific cylinder. Unique identification of the cylinder, and cartridge sample substrate and authentication of the sampling process would need development, too. Such an approach could be considered after the LAARS-DA concept that mirrors conventional bottle sampling was vetted.

3.2.4 LAARS-ES and LAARS-DA Safeguards Significance

Both LAARS-ES, which collects and analyzes aerosol particles and LAARS-DA, which enables on-site enrichment DA sample collection, provide safeguards technology to cover all three key safeguards goals described in Section 1. LAARS-ES can provide data on undeclared nuclear material and activities at the GCEP by warning if the plant is reaching undeclared enrichment levels. The LAARS-DA provides DA samples that are part of the random sampling process of UF_6 cylinders. These DA samples can tip off IAEA inspectors of undeclared production or processing of nuclear material at declared facilities by detecting small anomalies in the uranium isotopes that NDA cannot detect. These anomalies could suggest that the operator is overstating the tails' uranium-235 concentrations (Gordon et al. 2004) and diverting excess product, while filling the tails cylinders with nickel or steel shot to mask the missing weight of the undeclared product. The weights will be correct, but the minor differences in the tails assay could only be detected by DA sampling and high-precision enrichment assay. LAARS-DA feasibility depends on high-precision assay, which has not been demonstrated in the field by any method on tails cylinders. In this mode LAARS-DA would detect the diversion of the declared nuclear material by showing anomalies in enrichment that would point to missing uranium and uranium-235. Unique identification of the cylinder and cartridge sample substrate and authentication of the sampling process would also need development.

4 SAFEGUARDS QUALITIES OF THE UNATTENDED SYSTEMS

The advantages and disadvantages of the TRI-ACE, LAARS-ES, and LAARS-DA as viable safeguards technology as part of a GCEP safeguards approach are summarized in the following sections.

4.1 TRI-ACE

TRI-ACE strengths are listed below.

- Unattended collection of environmental samples in the absence of a safeguards inspector is possible.
- Automated collection reduces inspector manpower requirements and the possibility of inspector contamination, and provides enhancement over manual swipe sampling.
- Acquires samples when and where they are generated, before loss to the ventilation system or diffusion throughout the facility.
- Active collection has proven to be more effective at capturing particles than passive collection methods.
- Collector can be programmed to collect continuously or intermittently, depending on the facility operation and schedule.
- Tamper-indicating components ensure sample integrity.
- Data logging reports any off-normal conditions detected with tamper-indicating components.
- Data from tamper-indicating components are time-stamped to reconcile any discrepancies.
- LAARS-ES could be adapted to use TRI-ACE-collected samples, recognizing the additional flexibility that the TRI-ACE could provide to the LAARS approach in the broader context of environmental sampling.

Potential vulnerabilities of TRI-ACE are listed below.

- The current TRI-ACE prototype does not include a tamper-resistant locking mechanism. A lock and sealing system has been designed, but not incorporated into the prototype.
- Cross-contamination of plates from residual material in the air-intake baffle could possibly occur. Such cross-contamination issues have not been studied.
- The effectiveness of continuous aerosol sampling within a GCEP is an unproven method to date. Little information is known about the concentrations, frequencies of emission, and transport within a GCEP. Collecting viable ES using TRI-ACE is unproven in an actual GCEP setting.
- The long-term operation and reliability (i.e., several months) of the TRI-ACE has not been tested.
- Field testing is needed to demonstrate operation, reliability, and usefulness of data.
- The acceptance of the device by facility operators has not been approached.

Further studies are needed to determine how well TRI-ACE fits into an integrated GCEPs safeguards approach, taking into account the derived random and systematic uncertainties, ease of sampling, and performance.

4.2 LAARS ES

The strengths of LAARS-ES are listed as follows.

- Standalone on-site aerosol collectors acquire environmental samples when and where they are generated, before loss to a ventilation system or diffusion throughout the facility.
- The programmable collection integration period provides adaptive sample collection, depending on changing facility environmental conditions and production schedules.
- Time-stamped collection allows reconciliation against a declared production timeline.
- Automated collection reduces inspector manpower requirements and the possibility of inspector contamination, and provides enhancement over manual swipe sampling.
- The inspector-portable (handheld, backpack) aerosol sampler concept provides a deployment-flexible tool, which is an advancement over traditional hand swipe sampling methods, and directly addresses the IAEA's emerging focus on detecting undeclared activities during complementary access inspections.
- The aerosol collector is optimized to collect small uranyl fluoride particles, while rejecting larger nuisance dust particles.
- The LAARS analysis provides direct on-drum particle measurements, which sidesteps painstaking particle-handling and fission-track analysis.
- On-site LAARS measurements provide immediate results, which vastly improve the timeliness problems associated with off-site analytical laboratory analysis.
- On-site analysis is more cost-effective, because it removes sample handling, chain-of-custody, loss of continuity-of-knowledge, shipping costs, and logistics problems.
- Rapid results allow an inspector more agility, such as deciding to conduct additional inspections if unexpected assay results occur.

The potential vulnerabilities of LAARS-ES are listed as follows.

- The current LAARS-ES sampler does not include provisions for tamper-resistant or tamper-indicating technology. We anticipate that TRI-ACE methods could be adapted to the LAARS-ES sampler, but this has not been validated.
- Cross-contamination between sequential sampling periods or because of physical handling of the RDI during insertion/removal could possibly occur. Cross-contamination during LAARS analysis may be possible also, but none has been observed to date.
- The effectiveness of continuous aerosol sampling within a GCEP is an unproven method to date. Little information is known about the concentrations, frequencies of

emission, and transport within a GCEP. Collecting viable ES using LAARS-ES is unproven in an actual GCEP setting.

- Further studies are needed to determine how well LAARS-ES fits into an integrated GCEPs safeguards approach, taking into account the derived random and systematic uncertainties, ease of sampling, and on-site analysis performance.

Field-testing and evaluation are required to demonstrate actual LAARS-ES measurement and deployment performance, such as:

- efficient process emission aerosol collection
- high detection sensitivity and low false alarm probability
- noninvasive installation and operations
- modest acquisition cost and low maintenance cost
- simple assay process and intuitive, useful data analysis results for inspectors in the field
- robust and long-term reliability of the LAARS hardware.

4.3 LAARS-DA

The strengths of LAARS-DA are listed as follows.

- LAARS-DA compatible with sampling taps is used to collect DA samples.
- Reduces size of UF_6 DA sample quantity from grams to micrograms.
- LAARS analysis provides direct measurement of DA samples without any handling or chemical processing.
- On-site LAARS measurements provide immediate results, which vastly improve the timeliness problems associated with off-site analytical laboratory analysis.
- On-site analysis is more cost-effective, since it removes sample handling, shipping costs, and logistics problems.
- Rapid results allow inspector more agility, such as deciding to conduct additional inspection activities during LFUA to the cascade halls, environmental swipe sampling, or announced complementary access on the GCEP site beyond areas normally accessed in inspections, if unexpected assay results occur.
- The LAARS-DA concept possibly allows for the collection of significantly more samples, enabling a more comprehensive and robust statistical mass accountancy sampling plan.
- The assay accuracy of $\pm 0.01\%$ may interest facility operators as an indicator for process monitoring or product quality control, as well as for safeguards.

The potential vulnerabilities of LAARS-DA are listed as follows.

- The current LAARS-DA sampler does not include provisions for tamper-resistant or tamper-indicating technology. The maturity of this LAARS application has not allowed considerable thought regarding these possible vulnerabilities during DA sampling operations. Traditional DA bottle sampling is under the observation of the inspector. Similar approaches are possible for LAARS-DA sampling.

- Collecting DA samples from cylinders or sampling taps that are representative of the true sample enrichment has not been demonstrated. Accurate safeguards DA data are dependent on collecting well-homogenized UF₆ gas samples. Will microgram sample quantities, collected by LAARS-DA, provide accurate and representative DA samples? Traditional DA bottle sampling has similar potential vulnerabilities.
- Stable and effective gaseous UF₆ chemisorbent films have not yet been developed, but this work is under way and progressing well.
- Cross-contamination between sequential sampling periods or during the LAARS analysis could be a potential vulnerability. Further study is required here.
- The effectiveness of LAARS-DA sampling and on-site analysis is an unproven method to date. Field testing is needed to verify, authenticate, and compile useful data.

Some questions about LAAR-ES performance remain:

- Will LAARS-DA provide robust and reliable assay accuracy of $\pm 0.01\%$? Further study is needed to fully understand LAARS-DA performance. The random and systematic error bands must be determine, then compare to current DA techniques used by the IAEA.
- Will the LAARS-DA approach be accepted by facility operators or embraced by the IAEA?
- Will deployment be too invasive?
- Will the acquisition and maintenance cost be acceptable?
- Is the approach too complicated to be operated by inspectors in the field?
- Will the long-term reliability of the LAARS hardware be acceptable for the application?

4.4 Instrument Deployment Strategies

This section describes how to install the instruments for use in a GCEP.

4.4.1. TRI-ACE Deployment Strategies

For initial deployment, the fully assembled and programmed-for-use TRI-ACE will be shipped for instrument installation. To set TRI-ACE up for operation, the facility will need to supply power for the device as well as a location for mounting the box. The ideal locations in a GCEP for mounting the instrument box would be near the feed and withdrawal areas. The installers will need to do an initial system check before actual data collection begins. The size of the plant and number of critical material handling areas such as the feed and withdrawal areas would determine the number of TRI-ACE boxes. If the plant has a more complicated and complex design and more places material could be put in and out of the cascades, then more TRI-ACE boxes would be needed.

The nominal TRI-ACE operation is in unattended mode. A safeguards inspector could place the unit in a facility and power it on. TRI-ACE could collect samples continuously or at specified intervals during the inspector's absence. In addition to the many tamper resistant/indicating features, the inspector could take IAEA seals and seal the unit to detect tampering. At subsequent inspections, the safeguards inspector would remove seals, open

the unit, and remove the sample cartridge to collect the safeguards-relevant sampled material. A new sample cartridge would be inserted and the unit seal replaced. The removable sample cartridge is depicted in Figure 4-1 and can also be seen in the TRI-ACE prototype photographs in Figure 3-5. The IAEA inspector would deliver the sample cartridge under IAEA seal to SAL or one of the NWAL for analysis of the collection plates. SEM analysis or sample processing followed by mass spectrometry can directly provide verification of the material on the collection plates.



Figure 4-1. TRI-ACE Removable Sample Cartridge

The removable sample cartridge also contains a thumb drive, which will contain information recorded from the tamper-indicating components during the collection period. The IAEA could encrypt the thumb drive for data authentication. The IAEA could take the data logged onto the thumb drive for analysis performed in conjunction with the collection plates to determine if any off-normal events occurred during collection.

The capabilities of TRI-ACE described herein are for the system currently under development. Future enhancement of the TRI-ACE technology could allow for developing an on-site analysis capability of the collection plates or even analysis by the LAARS-ES system.

4.4.2 LAARS-ES Deployment Strategies

The LAARS-ES approach has two possible deployment options. This includes a fixed, stand-alone collector that could be stationed within a tamper-indicating enclosure at a feed and withdrawal station. Alternatively, it may be possible to develop a handheld or backpack-portable RDI sampler, where an inspector could collect ES on demand, while in route through the enrichment facility. In both cases the RDI could be removed and analyzed by the on-site LAARS instrument. The LAARS-ES uses a stand-alone aerosol collector based on an RDI. An inspector secures these aerosol collectors in tamper-indicating enclosures. The enclosure is part of future developments in collaboration with ORNL and SRNL. The inspector can stage the LAARS-ES at strategic locations within the GCEP near feed and withdraw station locations and areas where access to the cascade halls or exhaust ventilation from the cascade halls could be collected, as shown in Figure 4-2.

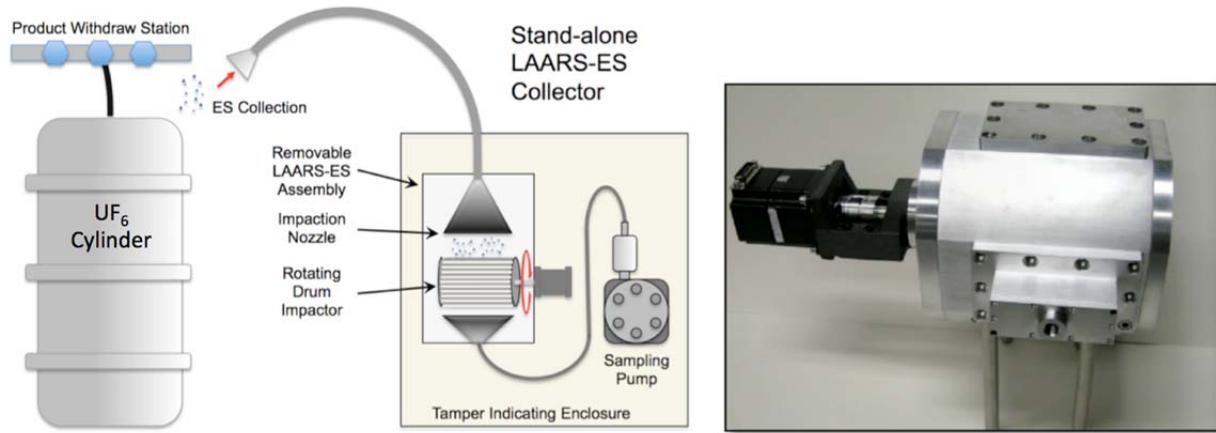


Figure 4-2. The LAARS-ES Instrument Uses a Stand-alone Environmental Aerosol Sample Collector (right), Housed within a Tamper-indicating Enclosure (left) --

Larger GCEP facilities could have multiple feed and withdrawal stations across the facility. It is likely that several collectors would be deployed at each station and possibly several more at other facility locations. The extent of ES collection effort would scale with facility size. Under automated operation, the RDI collects airborne particles over a preset time interval, at which point the drum rotates to expose a new rectangular strip on the drum. Continuous collection serves to acquire samples when and where they are generated (before loss or diffusion) and also provides the ability to time-stamp and pre-concentrate samples prior to analysis (something swipe sampling cannot provide). This approach also significantly relaxes the analysis detection sensitivity requirements and provides a pathway to enable on-site sample analysis. A drum having a modest diameter (~100 mm) could provide time-resolved particle sampling and integration on a daily or weekly basis over a one-year period. The collector is contained within a tamper-indicating enclosure.

During on-site inspections, the inspectors retrieve the aerosol collectors and insert them into the LAARS-ES instrument, shown in Figure 4-3, to rapidly screen for undeclared enrichment levels. The LAARS analysis instrument is intended to be deployed on-site within a secure IAEA room. The entire balance of the instrument could easily fit into a standard instrument rack (approximately 200 cm x 100 cm x100 cm) and require <10 amps at 110 VAC single phase. A 40-mm length, at the center of the 63.5-mm-long rectangular impaction strip, is raster scanned using LAARS-ES to characterize the collected particle distribution. The drum is rotated and the next row is scanned. This process is repeated until the entire impaction strip is analyzed. Within 10 to 20 minutes, each impaction strip is completely scanned and the uranium particle count and uranium-235 abundance distribution are determined. Each collected strip is sequentially analyzed until all impaction strips are assayed. On-site analysis display would show counts above a preset alarm threshold (i.e., 10% enrichment), providing immediate feedback for follow-up manual swipe sampling if required. If the IAEA and the operator and State conclude transmission protocol, the inspectors could electronically transmit raw and analyzed data to IAEA headquarters or regional offices for further review. The inspectors could archive

the unmeasured impaction strip regions or have these original samples analyzed off-site in a laboratory at a later date.

A second deployment scenario offers a new safeguards tool that could either enhance or possibly replace traditional swipe sampling in the future. Based on a well-proven RDI design, the aerosol sampler has specialized features that enable low-flow rate operation and handheld or backpack deployment. This design enables the inspector to quickly conduct ES on virtually any facility surface, without handling swipes, plastic zip lock bags, or the many other items in the swipe sampling kit. Aerosol sampler drum rotation provides the ability to physically separate samples taken at a large number of facility locations. Sampling conducted without handling the particles or the substrate should reduce potential sample loss or cross-contamination by the inspectors.

A battery-powered compact sampler is expected to allow collection of hundreds of samples per inspection day. Hence, each sample could be measured by the LAARS analysis instrument in just a few minutes. This design fosters agile inspections, allowing the inspector to shift verification focus as needed while in the field, regardless of the type of facility under inspection. On-drum particle capture has several advantages. The drum can be removed from the sampler and laser-based enrichment measurements using LAARS can be conducted on-site, directly on the impacted particles. No sample handling or preparation steps are required. Immediate on-site assay allows inspectors to refocus ES on select areas of interest within the facility, given an unexpected assay result. In the case of off-site analytical laboratory assay, particles can be directly removed from the drum using a conventional vacuum impactor. This concept embraces the IAEA's long-term strategy to develop portable technologies with ample design flexibility to detect undeclared activities within the expanding diversity of nuclear fuel cycle facilities.

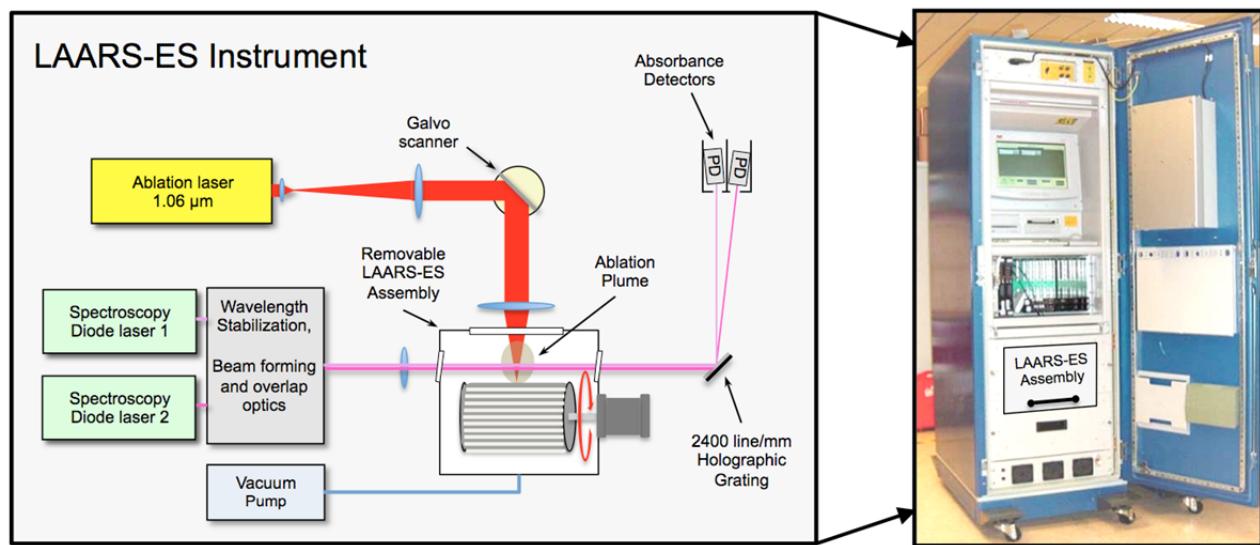


Figure 4-3. The LAARS-ES Instrument Used to Analyze Uranium Enrichment in Collected Environmental Samples

4.4.3 LAARS-DA Deployment Strategies

The LAARS-DA deployment strategy follows current DA sampling protocol, but also enables on-site enrichment analysis with enough precision to measure sample with bias defect level of uncertainty ($\pm 0.1\%$ uncertainty). In practice, the inspector requests the operator draw a DA sample from a particular feed, tails, or product cylinder. The cylinder could be selected from the storage areas or while being filled in the feed and withdrawal area. A facility operator connects the LAARS-DA sampling assembly (Figure 3-12) to a gaseous UF_6 sample tap, while under observation by an IAEA inspector. After connection to the sampling tap, the operator rotates the LAARS-DA sampler ball valve to expose a thin film-coated substrate to the gaseous UF_6 . After a short exposure period, the operator rotates the ball valve to isolate the sampler and then removes the sampler from the sampling tap. The operator transfers the sampler to the inspector's custody who takes it to an on-site LAARS-DA instrument. The inspector attaches the sampler to the LAARS instrument. The magnetically coupled sample transporter conveys the sample substrate into the sample chamber. The LAARS-DA measurement follows the same steps discussed previously in the LAARS-ES section, providing analysis results in about 10 minutes.

4.4.4 Safeguards by Design Issues in GCEPs for TRI-ACE and LAARS Installations

When investigating the utility of these instruments for operation at a GCEP, we need to take into account any issues with installation that Safeguards By Design (SBD) guidance (Laughter et al. 2012) gives us. The GCEP will need to have space for safeguards equipment, storage space for IAEA equipment, and office workspace with telecommunication equipment. The SBD guidance noted automated sampling devices should be designed to facilitate detection of tampering and the use of electrically operated sample valves that shut off after a period of time necessary to acquire enough sample material for the required analysis. If the operators share the instruments with the IAEA, the operator and State should ensure the protection of their national security and commercially sensitive data.

Hence, a plant being designed with safeguards in mind will need space for the TRI-ACE, LAARS-ES, or LAARS-DA collection units near areas where material is likely to be detected most easily such as the feed and withdrawal areas, cascade halls, and indoor storage areas. The versatility and small size of the TRI-ACE, LAARS-ES, or LAARS-DA collection units and the analysis units should make installation in any GCEP, even ones without SBD, a rather straightforward endeavor.

4.5 Evaluation of Proposed TRI-ACE and LAARS Use in GCEPs Safeguards vs. Status Quo

The goal of this study is to see if TRI-ACE and LAARS provide significant improvements and new dimensions to safeguards at GCEPs and identify the strengths and weaknesses of these technologies in a GCEP safeguards approach. In this section we will look at how effective these instruments are in ES and DA applications.

4.5.1 ES Effectiveness

The present mode of collecting ES swipes by IAEA inspectors depends on the inspector's on-site presence and skill in selecting locations for the swipes, collecting samples via

swipes, and securing those swipes and sending them back to a certified laboratory. The inspector must avoid cross-contamination. Both TRI-ACE and LAARS-ES will collect samples in the absence of an inspector and at intervals determined by the IAEA. The ability of a State to beat the system by running campaigns to produce HEU when inspectors are not present probably is detectable with campaigns of ES swipes being taken by inspectors. However, there is concern from the IAEA that a dedicated proliferator may run such clandestine operations between on-site inspections, then either avoid leaks or clean up any trace production signatures. A system that is constantly on and capable of detecting tampering with the system or environment would reduce concerns about the clandestine operations problem, by providing snapshots or conglomerate indications of the trace uranium isotopes over the course of the operation of the plant. Such a database could show anomalies pointing to possible plants misuse.

The effectiveness of TRI-ACE and LAARS-ES depends on cost benefits arising from reductions in time to obtain results, lower probability of cross-contamination, increased samples over the course of a year, and sampling at the best locations to detect undeclared HEU production. The key concern with current ES measures has been the cost of sample analysis, cross-contamination issues, and timeliness. If TRI-ACE and LAARS-ES can capture target aerosol particles and the LAARS instrument provides the on-site data that are required to detect undeclared enrichment, then the cost-effectiveness lies in improvements in performance. TRI-ACE samples will need to be shipped to a laboratory off-site, unless adapted for on-site LAARS analysis. However, the tamper-indicating enclosure helps ensure that there are authentic samples that have not been tampered with in the collection plates. While the LAARS analysis instrument will be secured in an IAEA room within the facility, the LAARS-ES stand-alone sampler has yet to develop a tamper-indicating enclosure. Hence, LAARS needs this feature to be effective in an unattended mode. However, the LAARS analysis provides direct on-drum particle measurements, which sidesteps painstaking particle handling and fission track analysis. Since TRI-ACE and LAARS-ES can provide samples tied to location and date and could be analyzed without shipping and contamination, there is a real cost benefit for having samples taken in an unattended mode. The timeliness issue is also a key benefit with LAARS-ES, since this approach eliminates the many months delay before the verification results are known. The current safeguard measures could be better, especially when considering the timeliness for detection of undeclared HEU production is one month. The other key benefit in both systems is the suite of samples, time-dated so that any changes in enrichments could be tracked over time.

The LAARS-ES also has the capability of being used to detect undeclared activities when configured in the mobile (portable and backpack) mode. Inspectors could use LAARS-ES as a complementary access tool around the GCEP site.

4.5.2 DA Effectiveness

The present mode of collecting DA samples by the operator under IAEA inspector supervision, and shipping the samples off-site for analysis has several drawbacks. The storage of the samples on-site and shipment to a laboratory demands continuity-of-knowledge of the samples for over a year in some cases and the risk of losing an entire material balance period's samples with one glitch in a seal knot or an accidental breakage

of the seal by plant personnel. The timeliness of getting the samples analyzed within the one-year window is difficult because of the aforementioned shipments. Furthermore, international shipments of even gram quantities of UF₆ can be difficult for operators in some States. Therefore, if an inspector could collect DA sample at the microgram quantities and analyze them on-site within the ITV requirements, there could be a major time and cost advantage. The IAEA would have the results before leaving the site, and the issue of timeliness would be removed. If the DA sample-taking process is simpler than at present, then concerns about increased burden of larger numbers of DA samples in expanding GCEP facilities may be alleviated.

Future LAARS-DA deployment concepts could include an unattended, automatic DA sampling system. For example, a cartridge, containing many sampling substrates, could randomly collect DA samples during product withdrawal. During on-site inspections, the inspector could retrieve the cartridge and perform enrichment analysis using the on-site LAARS assay instrument. This deployment concept would require a higher level of tamper resistance/indicating measures. Also it would be more complicated than ES sampling, since it would require a DA sample link to a specific batch of material contained in a specific cylinder. Unique identification of the cylinder (Friend et al. 2009, Boyer et al. 2012) and cartridge sample substrate and authentication of the sampling process also would need to be developed.

5 REFERENCES

Anheier NC and BA Bushaw. 2009. "Isotope enrichment detection by laser ablation - dual tunable diode laser absorption spectrometry." In *50th Annual Meeting of the Institute of Nuclear Materials Management 2009: INMM 50th Annual Meeting* pp. 641-650, Curran Associates, Red Hook, New York.

Anheier NC and BA Bushaw. 2010. "Unattended monitoring of HEU production in gaseous centrifuge enrichment plants using automated aerosol collection and laser-based enrichment assay." In *51st Annual Meeting of the Institute of Nuclear Materials Management 2010 (INMM 51)*, Curran Associates, Inc., Red Hook, New York.

Anheier Jr. NC, JT Munley, and M. L. Alexander. 2011. "Design of an unattended environmental aerosol sampling and analysis system for gaseous centrifuge enrichment plants." In *52nd Annual Meeting of the Institute of Nuclear Materials Management 2011 (INMM 52)*, pp. 3984-3994, Curran Associates, Inc., Red Hook, New York.

Anheier NC, BD Cannon, HA Qiao, J Carter, BK McNamara, MJ O'Hara, JR Phillips, and MM. Curtis. 2012. "Onsite Gaseous Centrifuge Enrichment Plant UF₆ Cylinder Destructive Assay," In *53rd Annual Meeting of the Institute of Nuclear Materials Management 2012 (INMM 53)*, Curran Associates, Inc., Red Hook, New York.

Bench G, PG Grant, D Ueda, SS Cliff, KD Perry, and TA Cahill. 2002. "The Use of STIM and PESA to Measure Profiles of Aerosol Mass and Hydrogen Content Respectively, Across Mylar Rotating Drums Impactors Samples." *Aerosol Science and Technology*, 36(5):642-651.

Boyer BD and MT Swinhoe. 2009. "Defining the Needs for Non Destructive Assay of UF₆ Feed, Product, and Tails at Gas Centrifuge Enrichment Plants and Possible Next Steps." In *50th Annual Meeting of the Institute of Nuclear Materials Management 2009: INMM 50th Annual Meeting*, Curran Associates, Inc., Red Hook, New York.

Boyer BD and MT Swinhoe. 2010. "Analysis of the Effectiveness of Gas Centrifuge Enrichment Plants Advanced Safeguards." In *51st Annual Meeting of the Institute of Nuclear Materials Management 2010 (INMM 51)*, Curran Associates, Inc., Red Hook, New York.

Boyer B, JM Whitaker, JL White-Horton, and KR Durbin. 2012. "Next Generation Safeguards Initiative: Overview and Policy Context of UF₆ Cylinder Tracking Program." In *53rd Annual Meeting of the Institute of Nuclear Materials Management 2012 (INMM 53)*, Curran Associates, Inc., Red Hook, New York.

Bukharin O. 2004. "Understanding Russia's Uranium Enrichment Complex." *Science and Global Security*, 12(3):193-218.

Bush W, G af Ekenstam, J Janov, E Kuhn, and M. Ryjinski. 2001. "IAEA Experience with Environmental Sampling at Gas Centrifuge Enrichment Plants in the European Union." IAEA-SM-367/10/04. In *Symposium on International Safeguards, Verification and Nuclear Material Security Proceedings*, International Atomic Energy Agency, Vienna, Austria.

Bushaw BA and NC Anheier, Jr. 2009. "Isotope ratio analysis on micron-sized particles in complex matrices by Laser Ablation – Absorption Ratio Spectrometry." *Spectrochimica Acta Part B: Atomic Spectroscopy*, 64(11-12):1259-1265.

Cooley J, K Murakami, C Charlier, E Kuhn, M Ryjinski, W Bush, P Friend, and M. Eastell. 1999. "Experience With Environmental Swipe Sampling In A Newly Built Gas Centrifuge Enrichment Plant." In *Proceedings of the Institute of Nuclear Materials Management: 40th INMM Annual Meeting, Held Phoenix, Arizona, July 25-29, 1999*. Institute of Nuclear Materials Management.

Erpenbeck HH, BD Boyer, BM Fane, KD Ianakiev, KA Miller, and SL Ward. 2011. "Practical Issues Associated with Implementing an Advanced Safeguards Approach at Gas Centrifuge Enrichment Facilities." In *52nd Annual Meeting of the Institute of Nuclear Materials Management 2011 (INMM 52)*, Curran Associates, Inc., Red Hook, New York.

Everton C, R Leslie, S Bayer, and M East. 2011. "Transparency and other State-Specific Factors: Exploration of Ideas for Evolving the IAEA's System of State-Evaluations and Safeguards Implementation." *ESARDA Bulletin*, No. 46:93-97.

Fishbone LG, et al., *Field Test of Short-Notice Random Inspections for Inventory-Change Verification at a Low-Enriched-Uranium Fuel-Fabrication Plant*. STR-302/ISPO-371, Volumes 1-3, International Atomic Energy Agency, Vienna, Austria.

Friend P. 2008. "URENCO'S Views on International Safeguards Inspection." Presented at the 8th International Conference on Facility-Safeguards Interface, Portland, Oregon, March 30-April 4, 2008. Available at <http://web.mit.edu/stgs/pdfs/Friend%20-%20Urenco's%20Views%20on%20International%20Safeguards%20Inspection.pdf>.

Friend P, D Lockwood, and RD Hurt. 2009. "A Concept for a World-wide System of Identification of UF₆ Cylinders." In *Proceedings of the 31st ESARDA Annual Meeting, Vilnius, Lithuania, May 2009*. Gordon DM and JB Sanborn. 1984. *An Approach to IAEA Material-Balance Verification with Intermittent Inspections at the Portsmouth Gas Centrifuge Enrichment Plant*. BNL-51782, Brookhaven National Laboratory, Upton, New York.

Gordon DM, BD Boyer, JM Whitaker, and JM Younkin. 2004. *Evaluation of IAEA Safeguards at Medium-Sized Gas Centrifuge Uranium Enrichment Plants*. BNL/NCT-03-002, ORNL/ESP02-102, Brookhaven National Laboratory, Upton, New York.

Hase K and B Boyer. 2012. "Analysis of Probability of Detection of Plausible Diversion Scenarios at Gas Centrifuge Enrichment Plants Using Advanced Safeguards." In *53rd Annual Meeting of the Institute of Nuclear Materials Management 2012 (INMM 53)*, Curran Associates, Inc., Red Hook, New York.

IAEA – International Atomic Energy Agency. 2001. *IAEA Safeguards Glossary, 2001 Edition*, International Nuclear Verification Series No. 3. International Atomic Energy Agency, Vienna, Austria.

IAEA – International Atomic Energy Agency. 2005. *Environmental Sampling for Safeguards*. STR-348, IAEA SGCP-PSA, International Atomic Energy Agency, Vienna, Austria.

IAEA – International Atomic Energy Agency. 2013. “Factsheets and FAQs, IAEA Safeguards Overview: Comprehensive Safeguards Agreements and Additional Protocols.” Accessed February 25, 2013 at http://www.iaea.org/Publications/Factsheets/English/sg_overview.html.

Kemp RS. 2010. “Source Terms for Routine UF6 Emissions.” *Science and Global Security*, 18:119-125.

Laughter M, J McCowan, B McGinnis, J Morgan, JM Whitaker, and A Pederson. 2012. *Implementing Safeguards-by-Design at Gas Centrifuge Enrichment Plants*. ORNL/TM-2012/364, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Lundgren DA. 1967. “An Aerosol Sampler for Determination of Particulate Concentration as a Function of Size and Time.” *Journal of the Air Pollution Control Association*. 17(4):225-229.

Menzel JH. 1983. “Safeguards Approach for Gas Centrifuge Type Enrichment Plants.” *Journal of the Institute of Nuclear Materials Management*, 12(4).

Miller KA, HO Menlove, MT Swinhoe, and JB Marlow. 2010. “A New Technique for Uranium Cylinder Assay Using Passive Neutron Self-Interrogation.” In *Proceedings of the IAEA Safeguards Symposium, Vienna, Austria*.

Panasyuk A, A Vlasov, S Koshelev, T Shea, D Perricos, D Yang, and S. Chen. 2001. “Tripartite Enrichment Project: Safeguards at Enrichment Plants Equipped with Russian Centrifuges.” IAEA-SM-367/8/02. In *Symposium on International Safeguards, Verification and Nuclear Material Security Proceedings*. International Atomic Energy Agency, Vienna, Austria.

Sexton L, P Cable-Dunlap, D Radford, T Riley, M Wellons, J Degange, L Trowbridge, B Anderson, and R Poland. 2012. “Development of a Tamper Resistant/Indicating Aerosol Collection System for Environmental Sampling at Bulk Handling Facilities.” In *53rd Annual Meeting of the Institute of Nuclear Materials Management 2012 (INMM 53)*, Curran Associates, Inc., Red Hook, New York.

Trowbridge LD, D Simmons, A Harter, D Lee, H Jennings, P Szczygiel, DA Bostick, C Hexel, E McBay, A Wall, B Anderson, R Kapsimalis, and P Cable-Dunlap. 2011. *TRI-ACE Progress Report – 2011, Part 1*. ORNL/TM-2011/424, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Tsvetkov I, W Bush, R Fagerholm, D Hurt, M Jordan, and J Leicman. 2007. “Implementation of the IAEA’s Model Safeguards Approach for Gas Centrifuge Enrichment Plants,” In proceedings for the 29th ESARDA Annual Meeting, Aix en Provence, France, May 22–24, 2007.

United Kingdom Foreign & Commonwealth Treaty Section, “Agreement between the Governments of the United Kingdom of Great Britain and Northern Ireland, the Kingdom of the Netherlands, the Federal Republic of Germany and the French Republic regarding Collaboration in Centrifuge Technology”, Treaty Series No. 001 (2007) Cm 7047, Cardiff, U.K., July 12, 2005.

United Kingdom Foreign & Commonwealth Treaty Section, "Agreement between the Three Governments of the United Kingdom of Great Britain and Northern Ireland, the Federal Republic of Germany and the Kingdom of the Netherlands and the Government of the United States of America regarding the Establishment, Construction and Operation of a Uranium Enrichment Installation in the United States", Treaty Series No. 133 (2000) Cm 5009, 3 July 24, 1992, Washington, D.C.

Vanderpool RW, DA Lundgren, and PE Kerch. 1990. "Design and Calibration of an In-Stack Low-Pressure Impactor." *Aerosol Science and Technology*, 12(2):215-224.

Zhao K, M Penkin, C Norman, S Balsley, K Mayer, Peerani, C Pietri, S Tapodi, Y Tsutaki, M Boella, G Renha Jr., and E Kuhn. 2010. *International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials*. STR-368, International Atomic Energy Agency, Vienna, Austria.