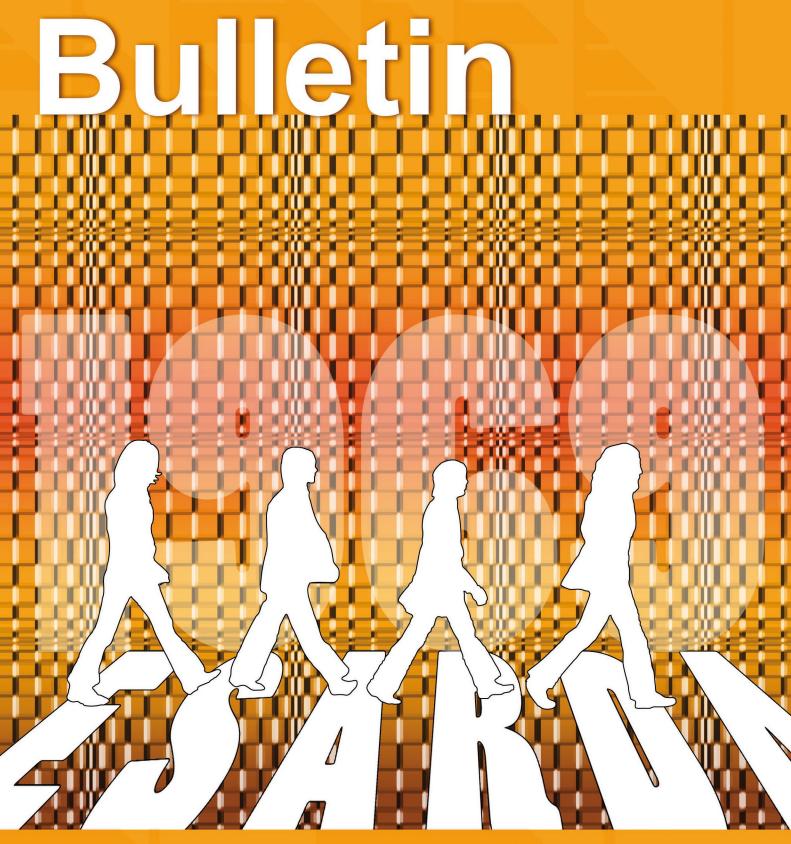
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Editorial

ESARDA is 40 and it does not look so...

L-V. Bril, F. Sevini Former and new ESARDA Secretary

ESARDA was founded in 1969 and, with its mature look and multi-disciplinary experience, is again looking to position itself to face the new challenges posed by evolving nuclear issues. A new group, *Reflection Group 2010*, will start work in the coming weeks under the chairmanship of Michel Richard.

As in the previous reviews of 1988, 1993 and 2000, the new group will assess the changes implemented following the recommendations of the *Reflection Group 2000*, and provide an opportunity to think ahead to how ESARDA might stimulate R&D in areas of concern that have emerged during the last decade.

Following the tragic events of 2001, and subsequent terrorist attacks, a European Union "Strategy against the proliferation of weapons of Mass Destruction" was formulated in 2003. More recently, DPRK's nuclear tests have been a demonstration of how nuclear proliferation continues in the face of international treaty, and how technology and knowledge gaps can be circumvented by illicit import practices.

There are signs of a *nuclear renaissance* that may extend to new regions, bringing a requirement for the development of new legal frameworks and infrastructure. This may generate an increased potential for nuclear proliferation: technology transfer increases the spread of common working standards for safety, security and safeguards, but also opens the possibility of clandestine replication of technology.

The announced increase in number of facilities and fissile materials will pose a heavy burden on safeguards and inspections, coinciding with the retirement of experienced staff. This calls for measures to attract a new generation of students to the subject of safeguards. In addition to traditional accountancy of fissile materials, new multi-disciplinary approaches are needed in order to enable information driven analysis with the aim to detect undeclared activities. Satellite imagery, environmental analysis, export and transfer control hence become important instruments in the fight against proliferation that require research and development.

2010 will also be the year of the NPT Review Conference, which will offer the chance to rethink this key legal instrument in the light of these new challenges and after the so-called "India deal". Last but not least, the renewed emphasis on nuclear disarmament and Fissile Material Cut-off also call for a review of the role of our Association.

It is also time to look back at a few of the achievements that occurred during the past years, mainly driven by the willingness of successive Presidents to implement changes. The ESARDA Secretariat has supported these changes as part of its duty.

Regarding Europe – almost the first word of ESARDA – following the European Union enlargement to 12 new Members, several European organisations have approached ESARDA to become new Parties. Today, some of them already have representatives in ESARDA. This development is still ongoing and is a proof of the dynamics ESARDA communicates for promoting co-operation.

In 2005, an administrative challenge was won: to create a new ESARDA contract with the objective to facilitate opening ESARDA to new Members, and to facilitate the establishment of new working groups. Since that time, four working groups have been established: Verification Technologies and Methodologies, Training and Knowledge Management (the idea of both groups was launched before 2005) and two audit groups covering nuclear material accountancy. When looking at new subjects and challenges, ESARDA will continue to evaluate whether dedicated working groups would deal with them more appropriately than the existing ones.

Another issue in which ESARDA has put a lot of effort is its communication strategy, through the Bulletin and the web-site. All the ESARDA publications, from the very beginning of ESARDA activities, were scanned and are available and searchable on line.

The first Bulletin was published in October 1981. Today, for the 40th anniversary, you are reading issue n 43. *Quel chemin parcouru !* Since 2006, there have been two issues per year. More recently there are two regular issues per year, complemented by special issues.

As part of its communication strategy, ESARDA has chosen to make a selection from the material published in the Bulletin. This was a wish of R. Schenkel, JRC Director General, who has always supported the Association. The Bulletin section on peer reviewed papers was hence born! Today, it has reached its cruise speed, thanks to the effort of the authors, reviewers and of the Editorial Committee. Complementarily, ESARDA has chosen to open widely its symposia every odd-numbered year, and to close its annual meeting every second year. For the latter, the participation is limited to working groups' contributors and invited experts.

Finally, and maybe the most transparent and important issue, the departing ESARDA Secretary would like to recall the dedication of Francesca, Chiara, Andrea, Monica and Elena for solving daily small and bigger problems: during eight years they have, one after the other, enriched ESARDA with their experience and their views; not forgetting the dedication of the ESARDA webmaster Jean-Claude.

Happy birthday ESARDA!

ESARDA News

Sweden towards Integrated Safeguards

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Abstract

This article is an extended version of a presentation that was given at the ESARDA symposium in Vilnius, Lithuania, 2009. In addition, the article will deal with the safeguards evolution in Sweden from the start of nuclear activities in Sweden until today. Furthermore, the article will reflect parts of the author's professional life, the last 30 years of which he has spent working in the field of safeguards at the Swedish authority.

Safeguards progressed in Sweden during the 1970s and 1980s, when new facilities and also new safeguards tools were introduced. Upon discovery of clandestine nuclear activities in Iraq, Sweden took part in the IAEA activities to strengthen safeguards under the '93+2 Programme', which, in 1997, led to the Additional Protocol.

After Sweden, together with the other member states of the European Union and the European Commission, signed the Additional Protocol in 1998, preparations have been ongoing for implementing Integrated Safeguards (IS).

In order to enable ratification and implementation of the Additional Protocol (AP), the Swedish legislation had to be amended, leading to the ratification in May 2000. The AP entered into force in the European Union on April 30, 2004.

Since then a long process has been going on to prepare for and implement IS in the EU states. For Sweden, the International Atomic Energy Agency (IAEA) drew the necessary conclusions to start implementation. There will be a combination of Short Notice Random Inspections and Unannounced Inspections. During 2008, discussions with the IAEA, the European Commission, the Swedish State Authority SSM and nuclear plant operators went on to pave the way towards IS. The most difficult issues to be dealt with were the LEU fuel fabrication plant, but also, for the Swedish State Authority, to make arrangements for its inspectors to be able to participate in the IAEA's Short Notice Random Inspections. This article will describe, how safeguards was organised in Sweden taking the Swedish fuel cycle into account, and how IS was implemented on all levels including the communication paths with the IAEA and the European Commission.

In 2008, a reorganisation of the Swedish safeguards authority took place. The first part of the article will provide information about this process.

1. A new Party to ESARDA – The Swedish Radiation Safety Authority

Before the main subject of this article is addressed, some information will be given about the structural changes of Swedish authorities dealing with nuclear and radiation matters.

In spring 2007, the Swedish government decided to merge the Swedish Nuclear Power Inspectorate, SKI, and the Swedish Radiation Protection Authority, SSI, into one new organisation, the Swedish Radiation Safety Authority, SSM. SSM has the collective responsibility for radiation protection and nuclear safety. SSM begun its operation on July 1st, 2008, with 240 employees; it is headed by a director-general appointed by the government.

SSM is divided into four departments having the following missions:

The Department for Nuclear Power Plant Safety

- supervision at reactor plants
- follow-up on incidents
- · perform safety tests
- review applications for increases in power.

The **Department of Radioactive Materials** (for the safe handling of radioactive materials in society)

- regulation and supervision of all nuclear facilities except power plants
- regulation and supervision of radioactive waste
- security

- rules and supervision concerning emission of radioactive materials
- supervision of transportation of radioactive materials
- ensuring ultimate storage of spent nuclear fuel including cost estimate.

The **Department of Radiation Protection** (for radiation protection, emergency preparedness and environmental monitoring)

- · maintaining the Authority's preparedness
- UV, solarium, radon, radiation from wireless technology and magnetic fields
- patient and personal radiation protection in healthcare
- supervision of all activities involving radiation resulting from sources other than nuclear power
- calibrating instruments within nursing and industry
- education activities.

The International Affairs Department

- support in coordinating radiation safety in Eastern Europe and Russia
- nuclear non proliferation issues.

There are also three support functions:

- Administration
- Director General Staff
- Communication.

Safeguards matters are handled within the International Affairs Department in the Section of Nuclear Non proliferation. This section adopted from SKI without changes in personnel and duties. In total, 9 persons are working in the section, all of them competent to perform safeguards inspections at the Swedish facilities. The section handles safeguards, export control, illicit trafficking and also the Support Programme to IAEA Safeguards (SWE-SP). The main projects within the SWE-SP are training of IAEA inspectors. Examples are spent fuel verification, fuel fabrication plant safeguards, use of satellite imagery and information collection from open sources.

SSM has a research budget of about 9 million Euros. The authority funds projects in basic research, carried out mainly by external companies and institutions such as consulting firms, universities and colleges. Some of the assignments are given to foreign research institutes and consulting firms. Only a small number of research projects are conducted within the authority. One reason for this is that there are a limited number of researchers available with competence within nuclear technology in Sweden, especially those who are independent of the industry.

The authority also funds professor services and postgraduate positions at some of Sweden's universities. This relates to professorships within radiation biology, radiation medicine and Man, Technology and Organization.

Part of the research programme is dedicated to nuclear non proliferation that is run by the Section of Nuclear Non proliferation. One outcome of the research is the development of the Digital Cerenkov Viewing Device (DCVD) which supported the SWE-SP and the Canadian Support Programme in the joint task to develop on instrument for verification of spent fuel.

The SSM contributions to ESARDA are funded from the research budget. SSM became a Party to ES-ARDA in autumn 2008, replacing SKI which had joined ESARDA in 1999. The main topics for SSM within ESARDA have been implementation of Integrated Safeguards and participation in the IS working group. SSM has also an increasing interest in the back-end of the nuclear fuel cycle. Previously, SKI participated in the currently inactive back-end of the fuel cycle working group. However, as for these new facilities licence applications will be submitted soon, the need for safeguards approaches is imminent. Therefore, SSM also participates in the C/S and NDA working groups for advice and assistance in issues related to the encapsulation plant and the geological repository.

SSM's main focus in the area of safeguards is now to get Integrated Safeguards running routinely and, together with IAEA and the EU-Commission, to develop safeguards approaches for the new facilities for the back-end; an encapsulation plant for spent fuel and a deep final geological repository. The licence applications for these facilities are expected in 2010 with the aim of the facilities to become operational by 2023.

What is new with regard to safeguards in the new authority? Non proliferation has a better position as being a section within the Department of International Affairs. This shows the importance of safeguards in an international context. The safeguards section co-operates with all the other departments within SSM and, now, a better overview is gained, as SSM supervises all those who handle radioactive material. This includes a better knowledge of the small holders. SSM also operates an analytical laboratory which can be a resource in connection with analytical sampling for safeguards purposes.

More information about SSM is available on the web site: <u>www.ssm.se</u>.

2. Sweden towards integrated safeguards – Introduction

Sweden, like many other states, started to implement its nuclear ambitions during the 1940s. In fact, only a couple of weeks after the Hiroshima bomb attack, a more structured research was initiated on the development of a Swedish option for use of nuclear, both civil and military. The idea was to use natural uranium, as there are quite vast resources within Sweden. For the civil part AB Atomenergi was created in 1947 by the state together with the nuclear industry.

Internationally, the progress in the development of the nuclear industry was made possible by the "Atoms for Peace Program" initiated by President Eisenhower in 1953. This made it possible to have trade with nuclear material and equipment. But the other side of the coin was that you also needed control of these activities which was the embryo of the Non proliferation Regime.

Operation of the first Swedish research reactor R1 started on July 13, 1954, in a rock cavern underneath the Royal Institute of Technology, KTH, in Stockholm. The uranium for this reactor was imported from France. The R1 was in operation until 1970, and the main use was neutron physics research. In the beginning of the 1950s, a research establishment was built 100 km south of Stockholm at Studsvik, where the research reactors R2 and R2-0 together with a couple of other small reactors were built.

This was the start of an intense nuclear development. Research projects to realize the Swedish line were launched, prospecting for uranium was initiated, a uranium mine was started in 1965, locations for possible reactor sites were identified, and a location for a reprocessing plant was selected. At the same time, the Swedish National Defence Research Establishment had ongoing research on the use of a nuclear option for military purposes. Both military and civil nuclear activities went along side by side during the 1950s and 1960s. The first commercial reactor was started in 1964 at Ågesta, a southern suburb of Stockholm, providing both electricity and heat to the suburb. The reactor was built in a rock cavern and was of the heavy water PWR type using natural uranium. The Ågesta reactor was in operation until June 1974.

More and more resistance arose against the military part of the nuclear programme. By the end of the 1960s, Sweden played an active role in the negotiation of the Non Proliferation Treaty, NPT, ratifying the NPT in 1970. This set an end to the military option of the Swedish nuclear programme. At the same time, Sweden abandoned its natural uranium programme, as the international market price of uranium had become so low that it was cheaper to buy enriched uranium from abroad than producing its own natural uranium. So, the uranium mine was shut down, the military research was stopped, and new types of light water reactors were constructed, the first one, a Swedish design of BWR in Oskarshamn, went into operation in 1972.

Sweden negotiated a bilateral agreement with the USA in 1956 resulting in supply of nuclear technology and material and, also, a US control of US-obligated material and equipment. A system of reporting and control was developed to fulfil the requirements, and US inspectors performed inspections in Sweden. In the beginning, this control was handled by AB Atomenergi, but, in 1956, was transferred to a new state authority, DfA. A national safeguards system was developed in the late 1960s.

In 1972, the first agreement with the IAEA was negotiated together with the US. It was a trilateral agreement which only covered US-obligated material. In 1975, a comprehensive safeguards agreement went into force according to the model agreement INFCIRC/153. At that time, Sweden had already developed and operated its own State System for Accountancy and Control. When the Swedish Nuclear Power Inspectorate, SKI, was created in 1974 replacing DfA, nuclear safeguards became one part of its responsibilities.

During the rest of the 1970s and 1980s there was an expansion in the use of nuclear energy, but one important event was the Harrisburg incident. The use of nuclear energy was questioned, and the Swedish government decided to hold a referendum in 1980 on the future of nuclear use in Sweden. The result was that a maximum of 12 nuclear reactors was to be built, including those already in operation, and eventually also a decision was taken by Parliament to phase out of nuclear energy by 2010. The last reactors began operation in 1985. Also a condition was set for using nuclear energy, and that was to present a method for taking care of the spent fuel. During the 1970s the option of reprocessing was chosen, and contracts were signed with both Sellafield and La Hague. But this changed, and the option became direct disposal in the Swedish bedrock. For this purpose, a common intermediate storage facility for spent fuel was built for all Swedish reactors, and the storage facility started its operation in 1985. Investigations were initiated to find a suitable location for a geological repository for the final storage.

3. The Strengthened Safeguards System, 93+2

Safeguards went on smoothly and no major disturbances occurred until the Iraq war in 1991 when the clandestine nuclear programme of Iraq was revealed. This caused activities among those involved in safeguards matters. At that time, the director of the safeguards division at the Swedish Nuclear Power Inspectorate, Mr Paul Ek, was the chair of the IAEA Director General's Standing Advisory Group on Safeguards Implementation (SAGSI). SAGSI initiated a lot of measures how to strengthen safeguards. This became the so-called '93+2 Programme' initiated in 1993 with the aim to be finalized within two years. Many states were involved in different field trials to test elements to strengthen safeguards, and so was Sweden.

4. Field trials in Sweden under 93+2

Sweden became engaged in testing four elements to strengthen safeguards: a so-called expanded declaration, increased and more timely information flow, unannounced inspections, and environmental sampling. In the following these four elements will be briefly described.

The Expanded Declaration: There was a proposal to evaluate the use of more detailed and timely information about the state's past, present, and future nuclear programme. As Sweden had had a two-line programme during the 1950s and 1960s, it was a good exercise to describe the Swedish situation. It was found that the description of the past activities was not easy to perform, though it was found of value to be able to explain what remained from the early activities. The Expanded Declaration eventually turned out to become the declaration format under the Additional Protocol after quite a few improvements.

Sweden also volunteered to test a timelier and also increased information flow. As the Swedish State System of Accounting for and Control of Nuclear Materials (SSAC) requires the operators to roughly daily report inventory changes to the Swedish authority, the authority always has available up-to-date information on the nuclear material in the State. The agreement with the IAEA for the field trial was to electronically submit weekly Inventory Change Reports (ICR). In addition, it was agreed, concerning the fuel fabrication plant, to submit, every Friday, information on the next week's planned production. This information together with the ICR information was then used by the IAEA to plan Unannounced Inspections.

An Unannounced Inspection scheme was also tested involving the power reactors, the fuel fabrication plant, and the research facility. The agreement was that the Agency inspectors would show up at the gate of the facility showing an inspection assignment to the guard. The operator should then immediately inform the Swedish authority which would send a state inspector to the facility. The IAEA would have access to the facility within 2 hours, and, if the state inspector still had not arrived, the operator would represent the state until the inspector arrived. In total, there were 5 or 6 Unannounced Inspections performed during a one-year trial.

The last element of strengthened safeguards to be tested in Sweden was the use of environmental sampling. This was done in the surroundings of three of the reactor sites, the research facility, and the fuel fabrication plant. Samples were taken both from soil and the sea.

5. The Additional Protocol and EU

The IAEA Board of Governors approved the proposal for an Additional Protocol (AP) to the Comprehensive Safeguards Agreements (CSA) at its meeting in May 1997. This happened, in fact, during the ESARDA Symposium in Montpellier, and Mr Murakami, IAEA-Director SGOC, had the honour to announce this to the participants of the symposium on the same day the decision was taken.

Now, the process started to prepare for signing and ratifying the AP. As Sweden had joined the European Union on January 1st, 1995, the Swedish situation had changed. Sweden joined the INFCIRC/193 CSA with the European Commission becoming the contact for the IAEA regarding the Swedish facilities. But the AP also covers areas not involving nuclear material. So, quite intense discussions were held within the EU concerning the implementation of the AP. Eventually, the decision was taken that the AP would enter into force in all EU-member states (including the two Nuclear Weapons States) at the same time. The AP was signed on September 27, 1998, with the ambition to have the AP ratified before the NPT Review Conference in 2000.

Intense discussions took place on the role of the different parties in implementing the obligations of the AP in EU. Given the fact that the AP identified responsibilities shared between the EU Commission

and the States, there was an agreement that those states, which so wished, could ask the EU Commission to perform all obligations for that state concerning the AP. This was achieved by submission of a so-called side-letter to the EU Commission. So, there arose a two-way solution in the EU, i.e., 'sideletter states' and 'non-side-letter states'.

In parallel, the old safeguards EU Regulation No. 3227/76 was subject to revision to reflect the new situation with the AP and, also, to be adjusted to the situation in the new millennium. That process went on during 2002, until the end of 2004, with a lot of involvement from the European Commission, the Member States and the Atomic Questions Group of the European Council. The new regulation, called Regulation (Euratom) No. 302/2005 on the application of Euratom safeguards, entered into force in March 2005.

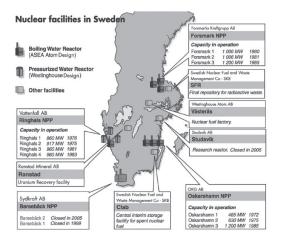
6. Swedish preparations between signature and entry into force of the AP

As Sweden had been involved in the '93+2 Programme' field trials, there were already some pieces in place, but one lesson learned was that one needed to know better about Sweden's past activities. So, several research projects were initiated to make a historical review of "nuclear Sweden". This initiative was taken to be able to answer possible questions from the IAEA about what happened with the old nuclear programme and facilities. Also, it was necessary to make a survey to be sure to get hold of all who were involved in production, import and export of nuclear-related equipment and material. The work to identify those who were involved in nuclear-related research was also a major task.

The Swedish Support Programme to IAEA safeguards had a task looking at the feasibility for the IAEA and also the States to use satellite imagery for checking site declarations. For this task two sites were chosen, i.e., the reactor site at Oskarshamn and the research complex at Studsvik. Satellite images were ordered and, at the same time, it was decided to order images for the other reactor sites in Sweden for the purpose of checking the completeness of the declarations received from the facility operators.

With regard to enabling ratification of and compliance with the AP, the existing legislation had to be analysed. As a result additional clauses were introduced on nuclear activities including an obligation to report on nuclear-related research and nuclearrelated equipment. A new law was required to enable access to facilities and activities not covered by the law on nuclear activities. Finally, it was necessary to inform the nuclear industry on the new obligations resulting from the AP. That was done through visits by the Authority to all who somehow had to submit information to the Authority to enable Sweden to fulfil the obligations of the AP. The installations that were regarded to become a site were asked to fill in a Site Declaration and to submit a draft declaration to the Authority. At this time, the Authority obtained from the IAEA the Protocol Reporter software, which was also made available to the safeguards personnel at the facilities, so that they could use this tool for preparing their declarations. In order to facilitate the process, representatives from both the IAEA and EU Commission jointly visited some of the sites together with the Swedish Authority. They discussed and agreed on the site boundaries and specified the scope of details that should be included in the information.

Sweden decided to be a 'non-side-letter state'. This means that Sweden is responsible for Articles 2a(i), 2a(iv), 2a(ix), 2a(x) and 2b(i). The EU Commission is responsible for Articles 2a(v), 2a(vi) and 2a(vii). For Articles 2a(iii) and 2a(viii) there is a shared responsibility, where Sweden prepares the declaration and submits it to the EU Commission which, after checking the content, forwards it to the IAEA. Regulation No. 302/2005 has a requirement to nominate a so-called "site responsible" for each site. Sweden decided to nominate its Authority as the site representative for all sites in Sweden, meaning that the operators report on changes of the site to the Authority; the information is compiled for all of Sweden before submission to the EU Commission and to the IAEA.



7. Entry into force of the AP and the route to Integrated Safeguards

Sweden ratified the AP in May 2000, but the entry into force did not take place until April 30, 2004. Once the AP entered into force, the installations which in previous years had sent their declarations, were asked to update the information to reflect the situation as of April 30, 2004. Eventually, there were only 8 sites that became candidates for the declaration. For those that were not included, an attachment to the declaration gave reasons for the exclusion from a Site Declaration. Examples were small installations for which exemptions were asked, old closed-down facilities without Design Information Questionnaires (DIQ) and Facility Attachments (FA). Some sites were declared with a smaller site area than discussed earlier, and for those the Authority described the buildings not included in the site in an attachment to the declaration. Finally, to complete the whole picture, all the research reports published on the nuclear history of Sweden were attached.

Sweden decided to use the reporting tool CAPE, developed by the European Commission, instead of the Agency's Protocol Reporter. The application of CAPE required converting the information received from the sites in Protocol Reporter-format to CAPEformat. The first declaration from Sweden, related to Article 2a(ixa), the export declaration, was submitted on August 5th, 2004, while the site declarations and the waste declaration were sent to the EU Commission at the end of September and the rest of the declarations directly to the IAEA on October 20, 2004. The most complicated declaration was related to the research projects, as it was difficult both to identify the possible actors for reporting research and, also, which projects were to be selected for the finial declaration. In total Sweden ended up with about 30 projects.

At this point in time, Sweden had completed the administrative parts and was waiting for the IAEA's response to the declaration.

Regarding the updates of the AP-declaration the Authority informed the facilities that, from 2005 on, an ordinary letter would be sufficient and that they were not obliged to use either the Protocol Reporter or CAPE. It was easier for the Authority to handle plain text and manually input the information into the CAPE-database, as it turned out that there was not a lot of information to be handled. At the same time, this led to a quality check of the submitted information.

The IAEA performed its first Complementary Access (CA) with 2 hours notice at the Studsvik research facility on March 16, 2005, in conjunction with the annual Physical Inventory Verification (PIV). The main issue discussed during this first CA was the Agency's use of a camera in support of visual observation. Sweden would not allow the Agency to use their own camera and offered instead a camera

supplied by the facility operator. After clearance by the facility security staff, the Agency inspector received copies of the pictures taken during the CA. The copies were stored on a CD-ROM. This became the agreed procedure for use of cameras. This CA was then followed by another four during 2005, one of them with 24 hours notice to the closed down Barsebäck site.

In July 2005, the IAEA sent out a letter asking for clarifications on the initial declaration. In fact, the letter was split into three parts, according to the responsibilities defined in the AP for the EU non-nuclear weapons states. In total, there were 50 questions. It was interesting to note that, when the AP-declaration was first submitted, Sweden attached information on its nuclear history but that did not include the uranium prospecting and mining activities performed during the 1950s and 1960s. Of course, there were questions about these activities and their consequences. Regarding the historical review, the lesson learned was to start from the earliest point where the state began its nuclear programme.

In 2006, Sweden received another IAEA letter asking for clarification, and that was linked to findings during one of the last CAs in 2005. This was all complementary information that IAEA searched for that included actions from Sweden after the first AP-declaration was submitted, five CAs and two clarification letters,. Then the IAEA was able to draw its Broader Conclusion for Sweden.

During 2007 and 2008 Sweden noted more activities both on the side of the IAEA and the EU Commission. The two organisations reactivated both their High Level Liaison Committee, HLLC, and their Low Level Liaison Committee, LLLC, and discussed how their cooperation had to be adapted to enable coping with the approaches under the Integrated Safeguards regime. In 2007 and 2008, Sweden and Finland together had joint meetings with the IAEA and EU Commission to discuss an Integrated Safeguards approach for Finland and Sweden. Finland reached an agreement on the approach early in fall 2008, and IS was introduced in Finland on October 15th, 2008.

For Sweden, the overall agreement on the implementation of IS was finalized late in fall 2008, but an agreement on the approach for the fuel fabrication plant was still pending. A joint meeting with the plant operator, IAEA, EU Commission and the new Swedish authority responsible for safeguards, the Swedish Radiation Safety Authority, SSM, was held in the beginning of November 2008. The discussions on the IS approach for the fuel fabrication plant started from the generic Partnership Approach on IS (PA-IS) document for LEU fuel fabrication plants that had been approved by the HLLC. After a full day of very constructive discussions with contributions from all sides an agreed PA-IS emerged for the Västerås fuel fabrication plant. The only remaining issue was to obtain a functioning mailbox system. This had to be postponed until the facility had finalized its ongoing upgrading of its safeguards accountancy system.

The Swedish impression was that the IAEA wanted to finalize the process of introducing IS in Sweden during 2008. Therefore, SSM visited the Studsvik research facility and the intermediate storage facility for spent fuel, CLAB, as these two facilities would be subject to Unannounced Inspections. During these visits procedures for granting IAEA access and for defining the contacts with SSM were discussed with both the safeguards and security staff.

Finally, there was a short meeting between representatives from the IAEA and SSM in Vienna in conjunction with another meeting early in December 2008 to confirm that all preparations for beginning IS were successfully completed on both sides. On December 19th, 2008, the IAEA sent a letter to the EU Commission, with a copy to SSM, confirming that IS would start in Sweden as of January 15th, 2009.

8. The Integrated Safeguards Approach for Sweden

For Sweden the approach eventually turned out to comprise the following:

For the 10 operating power reactors there is a SNRI regime with 48 hours notice and at least 2 inspections per year in total for all 10 reactors. In conjunction with the Physical Inventory Takings (PIT) there are two inspections, a pre-PIV and a post-PIV with surveillance during the period when the core is open.

For the fuel fabrication plant there is a SNRI regime with 24 hours notice and 48 hours retention time for the feed and produced products. There is also a mailbox system with daily information on the production and inventory. The EU Commission plans to have a maximum of four interim inspections with 24 hours notice, and the IAEA might appear unannounced to the operator during these inspections. A week-long PIV is planned as before.

For the Studsvik research facility and for CLAB there is an Unannounced Inspection regime with at least one inspection each. The inspectors shall be granted access within two hours. PIVs are performed as before.

For Locations-Outside-Facilities (LOF), Catch-All-Material-Balance-Areas (CAM), and other small installations one inspection within 4 to 6 years is planned in total for all installations.

The closed down Barsebäck reactors as well as Ranstad uranium recovery facility will have one PIV/DIV (Design Information Verification) each as before.

Complementary Accesses will be performed whenever the IAEA finds it necessary.

A consequence of the introduction of UI to CLAB and 48 hours notice SNRI for the reactors was that Sweden proposed to split the site containing both CLAB and the Oskarshamn reactors into two sites. The reason was that there is a right for the IAEA to ask for a 2 hours notice CA during an inspection, and that would mean that the reactors could be subject to CA when CLAB is inspected. This split is now accepted, and the update of the AP-declaration of this year reflects that situation, and there are now 9 sites in Sweden.

9. Administrative procedures at SSM for Integrated Safeguards

As soon as the IAEA letter confirming the introduction of IS in Sweden was received, SSM started to launch procedures to be able to participate in IAEA inspections. The receipt procedures of the inspection notifications were changed. A dedicated phone line was selected where all notifications, not only for UI, SNRI and CA, are received on a server that distributes the message to two mobile phones, to a dedicated email address and to a fax machine. This phone number was then communicated to the IAEA, the EU Commission, safeguards staff at Studsvik, CLAB and Västerås.

After Sweden joined the European Union in 1995, it has not been a legal obligation for the state to participate in the international inspections. This fact became obvious to the Swedish government, when IS were introduced in Sweden and it became clear that the EU Commission would not be able to participate in all inspections. So, the Government decided that Sweden should be represented by SSM at all IAEA inspections.

A rolling scheme was set up involving all 8 safeguards staff members in the non proliferation section. So, one inspector has one week at a time starting at noon on Fridays. This inspector has to be prepared to go for inspection immediately, when an advance inspection notice is received. One of the staff members has the responsibility to coordinate the activities, when a notice is received, that is to arrange for a rental car if needed, to communicate with the plant operator and to fax or email necessary information for the national inspector who has already left from his office. Studsvik and Västerås are possible to reach within the two hours limit, but for CLAB it will take about five hours. So, for CLAB the agreement with the facility is to let the IAEA inspector enter into the facility and, if needed, let the IAEA-inspectors be present to watch any process that has to be finished and then freeze the situation, until the SSM inspector arrives. IAEA can start with paper work as soon as that is possible.

The administrative procedures of SSM were tested almost immediately, as a 24 hours CA notice was received already 10 days after IS had begun in Sweden. That was for the Oskarshamn site with the three reactors and CLAB.

The first UI occurred on May 19, 2009, at the CLAB facility. The IAEA notice was received by SSM at 9.04 a.m., when the IAEA inspectors had notified SSM HQ that they were close to the facility. In fact, SSM got the fax, before the inspectors arrived at the gate. So, when SSM called the guardhouse, there were no inspectors there. But within moments they arrived at the building. This proved the proper functioning of the planned procedure. The inspectors got access to the facility within less then an hour. The SSM inspector on duty, who happened to be the author, was immediately on his way to drive the 350 km distance to the facility. The IAEA inspectors got the permission to enter into the process area, in order to "freeze the situation" by sealing equipment. So, no movements of material could take place undetected. When the SSM inspector arrived at 1.30 p.m., the decision was taken to perform measurements using the Spent Fuel Attribute Tester (SFAT) on the material known not to be verifiable by using the Cerenkov Viewing Device (CVD). The first day of inspection ended at 4.00 p.m., when the operator's working day ended. On the next day, the rest of the inventory was verified using the CVD, and the first UI in Sweden was successfully completed after lunch. The EU Commission did not take part in this inspection.

The first SNRI was performed at Ringhals 3 Nuclear Power Plant (NPP) on July 23rd, 2009, with a 48 hours advance notice. The SNRI activities are about the same as for the "old" interim inspections. Also this inspection conformed to the planned procedures, while also the EU Commission sent an inspector.

The second SNRI occurred at Forsmark 3 NPP on September 24th, 2009. When preparing for access to the facility SSM received another fax from the IAEA with a 24 hours CA notice, also for Forsmark. The SNRI was carried out without any problem; there had been a post-PIV only two weeks before. The subsequent CA was focused on waste handling and waste treatment. There was a visit to the underground storage for low-level and intermediate-level waste, and also the facilities for waste treatment at Forsmark 3 were checked. Finally, the mechanical workshop was looked at. At this CA, the IAEA had too long a list of buildings to visit, like for the preceding CA at Oskarshamn. Therefore, access to three buildings had to be cancelled. As long as it is clear for everyone that it is difficult to estimate the time needed for a CA, there would be no problem provided the buildings of highest priority are visited.

10. Conclusions

Implementing Integrated Safeguards (IS) has been a long and sometimes difficult process, as there are so many different parameters and actors to be taken into account. But it has also been very interesting and challenging for those within the Swedish Authority who have been involved. The co-operation between the different actors has improved a lot during the process and paved the way for a smooth implementation of IS. It is also important to mention that the ESARDA Working Group on Integrated Safeguards has played an important role for both Sweden and other states participating in the working group as well as for the EU Commission and the IAEA. For Sweden, it also provided the chance to document its nuclear history. More actors have been introduced to safeguards through involvement in different projects when looking for other tools to be used in safeguards, like the use of open source information including satellite imagery. But Sweden is just at the beginning of IS, and there is still a long way to go until there will be a solid safeguards system in place. It is noted that there still is a lot to be done, in order to have all parties act in line with the new regime. There is a need to train the inspectors, both at the IAEA and the EU Commission, how to perform the new IS inspections, and to develop guidelines for this. But it is equally important to develop instructions for the plant operators on how to act and prepare for being inspected. There are still states that need to adhere to the Additional Protocol, but states like Sweden with a functioning system in place can be good examples for those states which are still in their decision phase. The most important factor is to be clear and understandable when setting up rules and requirements and to inform, communicate and co-operate with each other.

6th ESARDA Course on Nuclear Safeguards and Non Proliferation

ESARDA Working Group on Training & Knowledge Management

1. Origin of the course

The knowledge retention problem in the nuclear field was acknowledged by the OECD in 2000. The United Nations study on disarmament and non proliferation education (2002) made detailed recommendations for urgently required improvements. ESARDA, the European Safeguards Research and Development Association reacted to these shortcomings with a strategy to tackle the problem and created a Working Group on Training and Knowledge Management (ESARDA WG TKM). The final objective of the ESARDA WG TKM is the setup of academic course modules to an internationally recognised reference standard.

This project is in line with the movement of establishing a European curriculum for Nuclear Engineering. Teaching in the Nuclear Safeguards field is indeed strongly influenced by national history so the objective of the course is to provide homogeneous material in Nuclear Safeguards and Non Proliferation matters at the European and international level.

2. Learning objectives

This compact course is open to masters degree students, in particular nuclear engineering students, but also to young professionals and International Relations/ law students. It aims at complementing nuclear engineering studies by including nuclear safeguards in the academic curriculum.

The basic aim of the course is to stimulate students' interests in safeguards. The course addresses aspects of the efforts to create a global nuclear non proliferation system and how this sytem works in practice: the Treaty on Non proliferation of Nuclear Weapons (NPT), safeguards technology, and export control. Also regional settings, such as Euratom Treaty, are presented and discussed. The course deals in particular with technical aspects and application of safeguards; i.e. how to implement the safeguards principles and methodology within the different nuclear facilities. Therefore the course will create an overview on inspections techniques, ranging



from neutron/ gamma detectors, to design information verification, to environmental sampling, etc.

3. Course content

Introduction: The evolution of the Non Proliferation Treaty-regime, safeguards, international control regimes in theory and practice, and present trends in the nuclear non proliferation efforts.

What is safeguarded: Definition of nuclear material that is subject to nuclear safeguards and related safeguards goals (significant quantity, timeliness and detection probabilities).

Where is it found: Description of the nuclear fuel cycle from mining to final repository, focusing on enrichment in the front-end and reprocessing in the back-end.

Which legal protection means exist: Overview on international and regional Non Proliferation Treaties and established Institutions and Organizations.

What is the methodology to verify: Nuclear material accountancy principles and statistics of auditing.

How are inspections performed: Overview on inspector tools and their use to verify the nuclear activities as declared under the safeguards agreements (Non Destructive Assay, Monitoring, Containment/ Surveillance); additional safeguards measures under the Additional Protocol (complementary access, satellite imagery, environmental sampling) and how they are applied in field (storage facility, process facility, enrichment facility, research institute, spent fuel transfer).

How to control Import/ Export: Guidelines of the Nuclear Suppliers Group, trigger list and dual-use list. Means to combat illicit trafficking, inclusive nuclear forensics.

What additional information offers: Collection of open source data and demonstration of some case studies (Iraq, 1993).

4. Practical organization

The course features a full five-days program with 1h lectures by experts in the field of nuclear safeguards, visits to five safeguards laboratories and some classroom exercises.

The course material, consisting of a complete set of presentations and literature will be provided to the participants. It is recommended that the students prepare themselves with the reading material on the website.

For this limited enrolment course early registration is recommended. A numerus clausus of 60 is introduced. Under the website <u>http://esarda2.jrc.it/internal_activities/WC-MC/Web-Courses/index.html</u> you find the registration form that has to be completely compiled and sent to <u>JRC-NUSAF-SECRRETARIAT@</u> <u>ec.europa.eu</u> before the deadline of 31 December 2009. University students can apply for accommodation free of charge, but only a limited number of places per university are available. Travel costs are not reimbursed by the JRC.

There is no course fee; lunches are offered free of charge.

All participants are encouraged to make an essay on a given topic selected from the list, which is handed out at the end of the course. Up to 2 best essays can be selected for being published in the ESARDA Bulletin or for being presented in the poster session at the next ESARDA Symposium.

Students can include this course, recognised by BNEN/ENEN for 3ECTS, in their academic curriculum. To be quoted for this course an additional Take-Home-Exam is foreseen.

Venue: JRC Ispra, Building 36, Amphitheatre

Schedule: From Monday, March 22nd, 2010 at 8:30 till Friday, March 26th, 2010 until 18:00

5. Pool of Course Lecturers

Y. Aregbe is responsible for analytical methods for nuclear material measurements at JRC Geel (IRMM)

J. Baute joined the IAEA in 1994 and became director of Iraq's Nuclear Verification Office. Presently he is director of the IAEA Safeguards Information Management Directorate.

R. Berndt leads the nuclear measurements at the Performance lab in the JRC with large experience in gamma spectrometry.

B. Burrows joined the British Nuclear Group in 1975 for nuclear material management and recently changed to the UK Nuclear Decommissioning Authority.

P. Daures worked as a nuclear engineer 10 yr at the CEA. He joined the JRC' Karlsruhe in 1994 to setup the OSL Lahague/ Sellafield, moved to Ispra as TACIS coordinator.

D. Dickman joined the Pacific Northwest National Laboratory in 1985, and is currently manager for Non proliferation and Global Threat Reduction Program.

P. Funk is since more than 10 years involved in French and International safeguards as leader of C/S lab at IRSN.

D. Grenèche is assistant director of Research and Innovation (formerly COGEMA: Companie Générale des Matières Nucleaires) of AREVA.

M. Hunt has been Nuclear Safeguards inspector of IAEA for the CIS, and is presently IAEA training coordinator.

O. Jankowitsch is head of the IAEA Office of External Relations and Policy Coordination, & Office of the IAEA Director General.

W. Janssens joined the EC in 1995 as nuclear inspector analyst for La Hague and Sellafield. He is presently head of the nuclear safeguards unit at IPSC JRC Ispra.

C. Jorant is director of Non Proliferation and International Institutions in the International and Marketing Department of COGEMA (AREVA).

T. Jonter is heading the Department of Economic History at the Stockholm University, leading educational programs on Nucl. Non proliferation at diff. univ. in former Soviet Union.

M. Kalinowski is director of the Carl-Friedrich von Weizsäcker Center for Science & Peace Research at the University of Hamburg and works for the Prep. Com. of the CNTBT organization. **G. Maenhout** joined in 2001 the nuclear safeguards unit at JRC Ispra and is part of the Belgian Nuclear Engineering teaching committee.

Q. Michel is Professor in European Studies and President of the Department of Political Science of Liège University.

P. Peerani leads the physical modeling (e.g. Monte Carlo) for nuclear measurements (NDA, solution monitoring) at JRC lspra with experience as analytical inspector.

L. Rockwood joined in 1985 the Office of Legal Affairs of the IAEA and is Section Head for Non Proliferation and Policy Making Organs.

P. Schwalbach joined the EC as EURATOM inspector in 1992 and is heading the logistic support for nuclear material verification.

M. Tarvainen is heading the Nuclear Trade Analysis Unit (NUTRAN) at the Department of Safeguards.

M. Wallenius works on destructive assay measurements and is responsible for nuclear forensics at JRC Karlsruhe (ITU).

Tribune and opinions

How to Become Involved in Nuclear Safeguards

B. Richter Jülich

Once, sitting together in Brussels - it may have been in January 2008 - Göran Dahlin, then President of ESARDA; Elina Martikka, then Vice-President; Louis-Victor Bril, then Secretary; and I chatted about this and that. Somehow, we reached the point where we recalled how each one of us had become involved in the strange field of international nuclear safeguards. We exchanged entertaining stories, and, finally, LVB stated that such stories might be motivating for young professional candidates to become interested in nuclear safeguards. At that time, the ESARDA course on safeguards had been fully established and was once more shortly to take place at Ispra. So he urged me, like he may have urged other "old fellows", to write "my story" for the Bulletin. For quite a while I hesitated to do so but, finally, I gave it a trial.

My getting involved in safeguards has undoubtedly quite a personal, and therefore non-representative, background, and I think it even evolved from my childhood, influenced by the circumstances that existed at the time. I will try to explain how it developed. I was born in 1944, in a small town which is now in Poland, but spent my first 14 years in Hamburg. In 1958 I moved to the Cologne-Bonn area, and finished my university studies in 1973 in Bonn. Then I worked for almost two years at the Weizmann Institute of Science in Israel, before, in 1976, I applied for a job at the Jülich Research Centre.

There is an old Spanish joke, saying that the important decisions a male human being makes in his life always result from a lack of something: He gets married for lack of experience; he gets divorced for lack of patience; and he gets married again for lack of memory. In my case, I believe it was more by accident that important perspectives evolved and decisions were made based on my experience and, by the way, I am still married to my first wife.

In 1945, after the end of World War II, the British Military Administration in Hamburg allowed my parents, who had evaded from Soviet-occupied East-Berlin, to stay in Hamburg and gave them jobs and residence. This gave me the opportunity, living in a British community, to become almost bilingual. Besides adopting the English tongue, I developed a very positive attitude towards people of a different nationality, as the British never gave me a feeling of inferiority because of my German nationality. This seemed to be amazing, after all that had happened during the Third Reich and World War II. But it showed their fairness in a situation of superiority, namely to give us a chance to do better in the future. Later, during the decade from 1957 to 1967, I had the opportunity to participate in student exchange programmes with England, and temporarily even thought of emigrating to the United Kingdom. The frequent visits opened my mind not only to the nuances of the English language but also to the thinking and approaches taken in another country. I wish I had had comparable opportunities in France because, as of 1958 after we had moved from Hamburg to the Rhineland, my second foreign language became French. Although France and the Federal Republic of Germany were founding states of the European Communities in 1957, my perception was that our relationship was different from the one with the United Kingdom and improving only slowly.

By chance, I had a very impressive and competent physics and mathematics teacher, who had spent many years in Bogotá, Colombia. It was under this influence that, after graduating at school, I decided to study science and to become a physicist. In the 1960's, nuclear physics was a booming science field in Germany, and Bonn University excelled in it. Yet, I found astronomy to be another interesting subject and spent some time as an intern at a radioastronomical observatory operated by the University of Bonn, where I realised that it would take me an astronomical amount of time to finish my studies. As it was in my interest to earn my living, marry, and raise children, I became an experimental nuclear physicist. At that time, I was still far from getting involved in nuclear technology, although this would have been an option for a nuclear physicist.

At this point, I should mention that Gotthard Stein, a former President (previously termed Chairman) of ESARDA, also studied physics at Bonn University, and it was there that we met during my first year. He was already in his third year, when we happened to have a temporal overlap in a laboratory where we took a practical course in analytical chemistry. Since he was more advanced than I, he was pleased to share his chemistry expertise with me. At a later stage, we happened to meet again in the nuclear physics institute, where we concluded our diploma and doctoral theses. On Thursdays we joined together on the university's sports grounds, where we played soccer with other students and senior scientists from our institute and never needed a referee: each participant was both player and referee, and it worked.

In order to make further progress in basic nuclear physics, and to lay a decision basis for my future professional life, I applied for a grant in Israel. Equally important for this application was the Holocaust. I wanted to meet former Germans who had survived and had become Israelis. My wife, our two children and I experienced an overwhelming fairness and acceptance, even on the part of survivors from the Holocaust, when we lived in Israel during 1974 and 1975. Today, we are still in contact with Israeli friends. In contrast, some US-American Jews, many of whom spent temporary terms in Israel, had difficulties to accept Germans. To the Israelis' surprise, there was nothing new they could tell me about the Holocaust, whereas it had been the general opinion that German students were not or not fully informed about the Third Reich and the Holocaust. On the other hand, an important experience I gained was the obvious need to survive in a situation of permanent threat and frequent wars (1948, 1956, 1967, 1973). In Germany, during the Cold War, I never felt equally threatened, as a nuclear war was looming which seemed so absurd that I could not spend a thought on it. In addition, this absurdity was a reason for me to postpone my service in the army until after finishing my university studies, which finally turned out to be a method to "circumvent" this duty. But, in Israel, it obviously made sense to be aware of a real threat and serve in the army, although this showed basically the high risk of any human existence. Yet, in Germany, the old generation suppressed these thoughts, and the young generation, for lack of experience, would not believe in this risk. Even today, Europeans seem to refuse to understand the situation in Israel, a state jeopardised by a number of states and groups which deny the United Nations decision of 1948 to establish the State of Israel. On the other hand, there is no question that also the Palestinians deserve a prospering civilian society in their own state but still fall short of an according approach. It is

only now, that some of their leaders have started to strive seriously for a political solution, whereas other leaders' intentions continue to be the military defeat and annihilation of Israel. It should not be ignored that one essential reason for this is the Palestinians' ever-lasting feeling of humiliation and inferiority. While this is not the place to propose an approach for solving the Israeli-Palestinian conflict, I want to make clear that the observed political problems touched me tremendously. I thought of becoming a diplomat, but waived these thoughts in view of my family. We concluded that a career in the diplomatic service would not be beneficial for our family life. However, a job in the "international business" might be quite desirable.

Back in Germany, I decided not to continue in basic research, but to do something with a more practical relevance. Bonn University gave me half a year to find a job, and I finally ended up at the national Jülich Nuclear Research Centre (in the meantime, the term Nuclear was dropped), which, in the 1970's, was in the process of diversifying its research activities away from exclusively nuclear engineering to nearly all fields of science. It was not yet meant to be the beginning of Germany's nuclear fading out policy, but rather indicated a stage of technical maturity in the nuclear field. The government continued to fund research on advanced reactor designs; spent fuel re-cycling; plant safety; and safe-guards, whereas it requested the nuclear industry to take the responsibility for commercialisation of power reactors.

Although I had been aware of the Treaty on the Non proliferation of Nuclear Weapons (NPT), I had not yet realised that it involved "international safeguards" executed by the International Atomic Energy Agency, a subsidiary of the United Nations Organisation. My return from Israel and start-up at Jülich in the mid-1970's almost coincided with the ratification of the NPT in Germany, but my initial responsibilities at Jülich were in science management until, after two years, I once again ran into Gotthard Stein. He had already entered into the nuclear field and his interest was in international safeguards. At that time, the German nuclear programme was largely growing, and this also required an increased involvement in safeguards research and development.

It was not only the ratification of the NPT and the entering into force of the Verification Agreement (based on the INFCIRC/153 Model Agreement) in the European Atomic Energy Community that triggered a lot of safeguards R&D activities in Germany, but also the presidency of Jimmy Carter in the United States of America (1976-1980). He immediately initiated the International Nuclear Fuel Cycle Evaluation (INFCE), which was perceived in Germany as an attempt at stopping commercial uranium enrichment and spent fuel reprocessing. Therefore, the German government recruited a lot of experts as consultants, among them Gotthard Stein. Furthermore, it seemed that it was not sufficient to have only one group of safeguards experts in Germany at the Karlsruhe Nuclear Research Centre, but to establish at least one more at the Jülich Nuclear Research Centre. So, it happened that Gotthard Stein raised my interest in nuclear safeguards with the perspective to contribute to the non proliferation regime and world peace. Without discrediting science management, I found this perspective really appealing and agreed to join the safeguards research group.

As a result of INFCE, the Hexapartite Safeguards Project on commercial gas centrifuge uranium enrichment plants was inaugurated for a duration of two years, and I became involved. It turned out to be very exciting, when I realised that a vast majority of US-American safeguards experts had a Jewish background, and I immediately remembered my experience with US-American Jews in Israel. Therefore, I was not surprised to run into people who had a basic concern about Germany acquiring nuclear weapons relevant technologies. In this situation, however, it helped a lot that I had lived in Israel. I was able to understand their concerns, and my US-American colleagues respected me, because sometimes I seemed to know more Hebrew and more about Judaism than they did. It became one of Got-thard Stein's achievements, when finally the Limited Frequency Unannounced Access was concluded for commercial gas centrifuge uranium enrichment plants, and Germany was not pushed to abandon this technology.

Ever since, I have had opportunities to participate in a great variety of safeguards research and development projects, ranging from the development of technical systems for containment-and-surveillance, as well as for non-destructive measurements, to plant specific safeguards such as developing safeguards approaches for geological repositories.

Even after thirty years of involvement in nuclear safeguards and safeguards R&D I must admit that it has never been boring, especially after the emergence of the Additional Protocol. For me, the appeal of nuclear safeguards lies in coordinating and accounting for the interests of many parties, i.e., the worldwide operating International Atomic Energy Agency (IAEA); the regional Euratom Safeguards Authority; the national government (in its obligation to cooperate with the IAEA under the NPT); the plant operators; the safeguards system developers; the universities and research laboratories; and, last but not least, the international safeguards community including the Member States R&D Programmes in Support of the IAEA. Within the German Support Programme, I was able to contribute to the develop-



International experts group on geological repository safeguards meeting at the Konrad Mine at Salzgitter/ Germany in 2004 (author: 1st row, left). Photo courtesy: DBE Technology GmbH.

ment of a number of safeguards equipment systems which the IAEA authorised for inspection use. This had only been possible where people from all involved parties were open, honest, and committed to the common goal of successfully achieving a development. In this respect, I should like to point out the importance of involving facility operators, in both the decision to start a development and in providing test beds for field testing under real facility conditions. A technical system has to be failsafe, yield conclusive results, and facilitate a reduction of both inspection effort and, if possible, facility operator's effort, and this has to be demonstrated and proven.

As safeguards is basically a cooperative approach of the world community, the acceptance of new developments on the part of the IAEA is highly dependent on the outcome of advisory and consultants meetings convened by the IAEA, where technical and political issues are discussed by groups of international experts and recommendations are given. This clearly shows the political dimension of safeguards which, as regards diplomacy, is a challenge for people with a scientific and technical background. In the earlier years of IAEA safeguards, i.e., after the implementation of the INFCIRC/153 agreement, there had been guite a competition between technical projects in different IAEA Member States. This led to an undue duplication of efforts and waste of resources, and put the IAEA in the delicate situation of having to use, maintain, and service a number of redundant technical systems provided by several Member States, where one system would have been sufficient and more cost effective. As a result of international consultancy, and in order to avoid further duplication, today's development efforts for the IAEA are widely shared between states: for instance in joint projects such as the recent development of the IAEA's next generation surveillance system, which is a joint US-German project.

The approach described above, for the IAEA safeguards, is also valid in the European Union (EU), with the advantage of a more homogeneous group of states that is less prone to political stress and competition than the IAEA. In order to coordinate safeguards R&D efforts, and make better use of limited resources in the EU Member States, the European Safeguards Research and Development Association was founded in 1969, shortly after the NPT had been opened for accession. ESARDA was to provide a permanent forum for experts of different scientific and technical disciplines. Basically, it is the working groups that make up ESARDA. I was grateful for the opportunity to be involved as a member and, later, as chairman of the Working Group on Containment and Surveillance, and also in the Steering Committee, Executive Board, and Editorial Committee. A working group functions very much like an orchestra: its success depends on its musical director, but also on the attendance, professionality, and motivation of its members. The chairperson of a working group has to rely on the input from and involvement of the meeting attendants. In the past, ESARDA parties had various interests which, not least, were nationally influenced. Therefore, it was partly a diplomatic task of the chairperson to address individual experts and raise their interest in joining the working group. Another as-pect was the widely spread perception on the part of party managements that ESARDA seemed to be more of a touristic club than a research and development association. The reason was that annual meetings, as well as working group meetings, were held at interesting places all over Europe, depending on which party volunteered to organise a meeting. However, the value of varying locations for conventions should not be underestimated as a means to improve mutual understanding regarding differences in culture and traditions in the different European regions and bring people closer together. "Together" was the European Commission's motto in 2007 when celebrating the 50th anniversary of the European Treaties. My personal curriculum vitae has led me to endorse the "touristic" component of ESARDA. With progressing European integration, the national component becomes more and more negligible, while we can already see signs of a European state level approach in nuclear safeguards.

By now, and thanks to the engagement of a great number of competent individuals, ESARDA has developed into a very successful association. The different working groups are highly respected by the delegating parties and by the European Commission. Their scientific output is used by recipient parties and provided to the public. ESARDA maintains a web site and issues a scientific journal, the ES-ARDA Bulletin. With its peer reviewed section, it has become attractive enough for many authors to publish their scientific results in the Bulletin. As a senior scientist and member of the Editorial Committee, I am grateful for having been able to contribute to this development. Last but not least, ESARDA has embarked upon another important mission: attracting and educating young professionals. During the last twenty years, there has been a decline of nuclear energy, with no new plants being built but with the necessity to cope with nuclear safeguards and safety even after abandoning nuclear energy. The "old" generation of nuclear experts, including safeguards experts, is about to retire. Therefore, it has been extremely important that ESARDA, like the Institute of Nuclear Materials Management in the United States of America, has taken the initiative to retain safeguards expertise and to introduce it into nuclear engineering studies at European universities. Moreover, it should be realised that nuclear safe-guards set a standard for treaty verification in general which, after the NPT, has influenced subsequent treaties, such as the Chemical Weapons Convention. It has become my generation's responsibility to transfer our knowledge to the next generation of nuclear experts by participating in teaching and by documenting our expertise, not only in papers but also in books and seminars, which we do. By now, I am optimistic that, in particular with the indication of a nuclear renaissance, it will be possible to raise the next generation of safeguards experts.

Book reviews

International Safeguards and Satellite Imagery Key Features of the Nuclear Fuel Cycle and Computer-based Analysis

Editors: B. Jasani, I. Niemeyer, S. Nussbaum, B. Richter and G. Stein Publisher: Springer. P200. 2008. ISBN: 978-3-540-79131-7

Book reviewed* by J. E. Cartwright

*A more detailed specific technical critique of the book is available from JECCartwright@aol.com

Introduction

With five contributing editors the book falls into three parts, and a well researched introductory section details the history and development of the Non Proliferation Treaty (NPT), Safeguards measures, agreements and means of verification including satellite based remote sensing. Satellite imaging systems and digital exploitation techniques are explained and case studies are used to demonstrate these capabilities. The book also sets out to determine any identifiable signatures or 'keys' related to nuclear facilities that could be used in the interpretation and verification of activities.

But close examination reveals that too many unexplained anomalies cast doubt on the efficacy of some of the digital technologies. No convincing set of 'keys' emerge and use of the demonstrated techniques in Safeguards applications, in their present state of development has to be questionable.

Leading Articles

Bhupendra Jasani explains orbital mechanics and compares current commercial satellite imaging platforms. Irmgard Niemeyer gives us a perspective of developing satellite technologies relevant to the NPT and considers the sensors, their technical limitations, controls and availability. Some of the terminology is unexplained and a technical glossary would have been helpful.

Part 1 – A Key for the Nuclear Fuel Cycle

Here Jasani proposes that a set of common features, identifiable signatures or 'keys' related to nuclear facilities can be determined for analytical use and to develop algorithms to automatically detect these facilities on a satellite image He uses a selection of German Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs) as examples

with a list of common features or indicators.- 'Keys'. If we consider just one: "PWRs are housed in 60m cylindrical containment buildings with a domed roof and that BWRs use rectangular containment buildings 35m x 45m. Thus it is possible to distinguish a PWR from a BWR and devise an algorithm to automatically detect them in a satellite image." Indeed there are common features in this selection but this is a false premise. Not all PWRs and BWRs will fit Mr Jasani's criteria, a criteria that cannot be globally applied. He also states "there are no extensive security perimeter fences at civil reactor complexes". This is incorrect - since 9/11, all civil reactor complexes have upgraded their perimeter and security measures! But it is his first 'key', based on a false premise that must surely undermine the legitimacy of any subsequent computer based results from algorithms.

On Research reactors he concludes that "too few examples were studied to be able to 'generalise' the 'keys". Yet he asserts (incorrectly) "that pool type reactors are housed within a rectangular building" This may often be the case but it is not always so and it is not a reliable 'key'. The quality and scale of some of the imagery used for illustrations is disappointingly poor and at the Bushehr NPP several features are misidentified and incorrectly annotated. The subsequent studies on 'keys' for Enrichment and Reprocessing plants lack depth and the Google Map images used are too small to see detail. At Capenhurst a unique opportunity to compare two centrifuge enrichment facilities on the same image is not taken up and the list of centrifuge enrichment 'keys' finally produced is neither accurate nor complete.

Only two Reprocessing plants are studied; Hanford (USA) and THORP (UK). Here, similarities in the selected sites are described but no attempt is made to consider other more complex facilities where the similarities would be much more difficult to define

Significantly perhaps, no definitive list of 'keys' emerge from this particular study which is shallow, sometimes vague, unconvincing and incomplete.

Part 2 – Computer-based Image Analysis

Sven Nussbaum introduces Object-based computer image analysis. He proposes that a generator hall at a nuclear power plant can be so classified by its size and rectangular shape, by a given distance from the reactor dome, and by small rectangular objects on the roof. Having set these parameters, feature recognition software can extract i.e. identify the generator hall. He demonstrates this for Power reactors, Research reactors and at the Bushehr NPP using the key features identified earlier by Jasani. But while his studies show the plausibility of the method, by using Jasani's 'keys' drawn from the questionable selection of sites in the previous chapter the same programming would not I fear, extract or recognise the 'generator hall' at numerous other NPPs worldwide where the real parameters vary so much from the model and, in one particular case, where turbine/generators are not housed in a building at all! Alas, there are more misclassification errors in the Bushehr study.

On Research reactors, Jasani earlier stated that more research was necessary to develop the 'keys'. Now Nussbaum moves on to use Jasani's untested criteria and redefines (contradicts) the criteria of "pool reactors being characterised by rectangular shaped rather than circular shaped, domed containment structures." Some wires are crossed here!

On the evidence presented, an experienced nuclear imagery analyst will see little cost effective value in such sophisticated techniques where the same key objects in the image are in an instant recognised immediately by the eye of the trained image interpreter.

Part 3 – Image Data and Safeguards

Nussbaum and Niemeyer describe how changes can be detected in images taken at different times using various methodologies; Combined Pixel and Object based change detection and Pre-processing of satellite data using geometric correction, atmospheric correction and Pan-sharpening techniques. Two studies demonstrate these techniques; the Arak Heavy Water Plant (HWPP) and the Natanz Uranium Enrichment Facility (UEF) in Iran. The Arak study uses images from 2004 and 2005 and demonstrates how newly constructed buildings and streets can be identified. But several other changes, noted but not explained, can often be attributable to causative factors not embraced by the algorithm such as differences in sun angle, reflectivity, radiance and more. So the technique is far from reliable. The Natanz study similarly uses imagery from 2003 and 2005. Here too, more anomalies and several classification errors persist where some features are not correctly defined and the failure is neither recognised nor explained. More anomalies arise where the algorithm has been unable to differentiate changed background from changed buildings. Elsewhere, further unexplained changes are in fact those of spectral characteristics rather than physical change.

Conclusions

Niemeyer herself concludes that much more work remains to be done with these new technologies. From the purist imagery analysts' point of view the science is inexact. There are still too many unexplained anomalies that cast doubt on the efficacy of some of the digitised object extraction and classification techniques. It follows then that any use in Safeguards applications has to be questionable. Identification 'keys' can have a part to play in imagery analysis of nuclear facilities but the authors' are less than convincing in their knowledge of nuclear facilities and have failed to provide a meaningful set of 'keys'. That said, 'keys' themselves are not a panacea. They are fallible not least because a wide range of nuclear facilities and installations do not conform to the model.

John E Cartwright had a long career as an imagery analyst, was an instructor at the UK Joint School of Photographic Interpretation (JSPI) and became a national IMINT specialist in nuclear proliferation studies. He held a staff appointment with the IAEA in the Satellite Imagery Analysis Unit (SIAU) where he developed a unique training programme and specialist material for imagery analysis of nuclear fuel cycle facilities and installations. He continues to provide specialist consultant advice and training to the IAEA, to the UK National Nuclear Laboratory (NNL) and the European Union Satellite Centre (EUSC) and has also published material with Jane's Intelligence Review.

Peer reviewed section

Unfolding Sample Parameters from Neutron and Gamma Multiplicities using Artificial Neural Networks

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Abstract

Expressions for neutron and gamma factorial moments have been known in the literature. The neutron factorial moments have served as the basis of constructing analytic expressions for the detection rates of singles, doubles and triples, which can be used to unfold sample parameters from the measured neutron multiplicity rates. The gamma factorial moments can also be extended into detection rates of multiplets, as well as the combined use of joint neutron and gamma multiplicities and the corresponding detection rates. Counting up to third order, there are nine auto- and cross factorial moments.

Adding the gamma counting to the neutrons introduces new unknowns, related to gamma generation, leakage, and detection. Despite of having more unknowns, the total number of independent measurable moments exceeds the number of unknowns. On the other hand, the structure of the additional equations is substantially more complicated than that of the neutron moments, hence the analytical inversion of the gamma moments alone is not possible.

We suggest therefore to invert the non-linear system of over-determined equations by using artificial neural networks (ANN), which can handle both the non-linearity and the redundancies in the measured quantities in an effective and accurate way. The use of ANN is successfully demonstrated on the unfolding of neutron multiplicity rates for the sample fission rate, the leakage multiplication and the ratio. The analysis is further extended to unfold also the gamma related parameters. The stability and robustness of the ANNs is further investigated to verify the applicability of the method. The ANN approach enables extraction of additional important information on the fissile sample compared to the application of the analytical method. **Keywords:** safeguards; neutron and gamma multiplicities; joint moments; material accounting and control; artificial neural networks.

1. Introduction

Neutron multiplicity detection rates, based on higher order factorial moments of the neutron counts from an unknown sample, can be used to determine sample parameters [1–3]. The factorial moments here refer to those of the total number of neutrons generated in the sample by one *initial source event* (spontaneous fission or (α, n) reaction). Due to internal multiplication through induced fission, the probability distribution of the total number of generated neutrons will deviate from that by the initial source event (mostly spontaneous fission), the deviation being a function of the sample mass (via the first collision probability of the initial neutrons). This property is transferred to the measured multiplicity rates, i.e. the singles, doubles and triples, and this is corroborated by the fact that in the latter the sample fission rate occurs explicitly. This gives a possibility to determine the sample mass.

Measurement of the first three multiplicity rates enables the recovery of three unknowns, which are usually taken as the sample leakage multiplication **M** (related to the first collision probability p), the ratio α of the intensity of single neutron production via (α , n) reactions to that by spontaneous fission, and the spontaneous fission rate, *F*, the latter being the most important parameter. This leaves the detector efficiency undetermined and it needs to be predetermined experimentally, or by using alternative approaches such as assuming the sample multiplication to be known and then the detector efficiency can be unfolded.

Recently it was suggested that in addition to neutron multiplicity counting, gamma multiplicities be also used [4–6]. The motivation for using gamma counting is manifold: higher gamma multiplicity per fission,

larger penetration through most of the strong neutron absorbers, and the relatively easy detection of gamma photons with organic scintillation detectors [7,8]. The goal is still the same, i.e. to determine the above factors, plus the further unknowns introduced, such as the gamma leakage multiplication, the ratio of single gamma to fission gamma intensity and gamma detector efficiency. These can though be handled since three neutron and three gamma multiplicities can be measured simultaneously, so one has still as many unknowns as measured quantities.

However, there exists the further possibility of using the joint (mixed) moments of the neutron and gamma counts, which supply further independent measured data to determine still the same number of unknowns. Accounting also for the joint moments up to third order, there are altogether nine factorial moments. Hence the problem becomes overdetermined.

At the same time, the searched parameters are contained in a highly non-linear way in the multiplicity expressions. This is already true for the gamma moments and multiplicity rates alone. To handle the non-linearity of the problem which prevents an analytical inversion of the gamma multiplicity rate formulae alone and in addition to make maximal use of the redundant information from the measurement when also the joint moments are used, the unfolding of the parameters has to be performed by leastsquare type unfolding methods. Actually, there is a conceptually simple non-parametric unfolding method for such a purpose, the artificial neural networks (ANNs), whose use will be demonstrated here.

In this paper we focus on showing the feasibility of the idea to use ANNs to unfold the sample parameters. Also we will look into how the ANNs are structured and applied to the problem in order to achieve optimum performance and accuracy.

2. Theory

2.1. Definitions

The following definitions and conventions will be used. Random variables and their moments referring to neutrons will be denoted by v, and those for gamma photons by μ . Variables referring to spontaneous fission will have a subscript *sf*, and those referring to induced fission a subscript *i*. For the factorial moments, there will always be a second index, giving the order of the moment. Hence, $v_{s/2}$ will stand for $\langle v(v-1) \rangle$ in case of spontaneous fission.

The factorial moments corresponding to the distribution of neutrons or gammas emitted in fission, whether induced or spontaneous, are nuclear constants and are known in advance. As is usual in such work, it is practical to include the (unknown) contribution from generation of single neutrons and photons, such as by (α , n)-processes for the neutrons, into the moments related to spontaneous fission. However, there is a need for introducing a similar correction for gamma photons, since they also can be produced either in bunches (in the spontaneous fission process) or as singular gamma photons, in the same (α , n)-reactions which lead to the emission of single neutrons. In addition, there is also the presence of a "background" type emission of single gamma photons from various processes, such as from radiative capture of neutrons.

To account for the presence of single neutron producing events in addition to spontaneous fission, one introduces the statistics of the total source events as a weighted average of the two processes [1,6]. Quantities belonging to such a generalized source event will be denoted by a subscript *s*. Hence, we will use

$$\nu_{s,n} = \frac{\nu_{sf,n}(1 + \alpha \delta_{1,n})}{1 + \alpha \nu_{sf,1}}, \quad \delta_{1,n} = \begin{cases} 1, \ n = 1 \\ 0, \ n \neq 1 \end{cases}$$
(1)

as source moments for neutrons. The factor $\boldsymbol{\alpha}$ is defined as

$$\alpha = \frac{Q_{\alpha}}{Q_f V_{sf,1}}$$

Here Q_f and Q_a are the intensities of spontaneous fission and (α, n) -processes, respectively. For gamma photons produced also in connection to (α, n) -reactions, the source distribution changes and leads to the following modified source moments:

$$\mu_{s,n} = \frac{\mu_{sf,n} + \delta_{1,n} \alpha v_{sf,1}}{1 + \alpha v_{sf,1}}$$
(2)

Here, $v_{sf,n}$ and $\mu_{sf,n}$ are the true moments of spontaneous fission (i.e. nuclear constants), whereas $v_{s,n}$ and $\mu_{s,n}$ are the ones corrected for the inclusion of production of neutrons and gammas by reactions other than fission. The moments relating to induced fission remain unchanged for neutrons and photons $(v_{in} \text{ and } \mu_{in} \text{ respectively})^{1}$.

2.2. Multiplicity detection rates

The measured quantities are the multiplicity rates. To convert the factorial moments of a single source

Following the notational traditions, no comma will be used to separate the subscript "i" from the moment order number "n".

event into detection rates of multiplicities, one has to account for the intensity of the source events and the detection efficiency. The principles of derivation of the factorial moments needed to formulate the multiplicity rates can be found in [1,4-6]. The effect of the finite measurement gate time in multiple coincidence measurements, quantified with the relative gate width factors as described in [10], will be omitted here. This is only for the sake of simplicity of notations, since the inclusion of the gate time factors does not represent any conceptual difficulty.

In the case of singles for neutrons, the following expression is derived:

$$S = F\varepsilon_n (1 + \alpha v_{sf,1}) \frac{\mathbf{M} v_{sf,1} (1 + \alpha)}{(1 + \alpha v_{sf,1})} =$$

$$= F\varepsilon_n \mathbf{M} v_{sf,1} (1 + \alpha).$$
(3)

Here, ε_n stands for the neutron detection efficiency. Note how the scaling factor $(1+\alpha v_{sf,1})$ between the spontaneous fission source $Q_f \equiv F$ and the total source intensity $Q_s \equiv Q_f + Q_a$ cancels out in the expression for the measurable singles². The neutron leakage multiplication **M** was also introduced and is defined as follows:

$$\mathbf{M} = \frac{1 - p}{1 - p \, V_{i1}} \tag{4}$$

where \boldsymbol{p} is the probability to undergo a reaction for the neutron.

In a similar way doubles and triples can be derived as:

$$D = \frac{F\varepsilon_n^2 \mathbf{M}^2}{2} \left[v_{sf,2} + \left(\frac{\mathbf{M} - 1}{v_{i1} - 1} \right) v_{sf,1} (1 + \alpha) v_{i2} \right]$$
(5)

$$T = \frac{F\varepsilon_n^3 \mathbf{M}^3}{6} \times$$

$$\times \left\{ v_{sf,3} + \left(\frac{\mathbf{M} - 1}{v_{i1} - 1}\right) [3v_{sf,2}v_{i2} + v_{sf,1}(1 + \alpha)v_{i3}] + 3\left(\frac{\mathbf{M} - 1}{v_{i1} - 1}\right)^2 v_{sf,1}(1 + \alpha)v_{i2}^2 \right\}$$
(6)

These are the quantities one measures in multiplicity counters. It is these expressions that serve as the basis for the different approaches to find the various unknown parameters, as described in [3]. Most commonly one assumes the neutron detector efficiency ε_n to be known, and solve the equations for *F*, **M**, and α . It is worth noting that Eqs (3), (5) and (6) are linear in *F* and α , but are highly non-linear in **M** (fifth order for the triples, Eq. (6)). It is a sheer coincidence that **M** can be obtained from the above as a solution of a third order equation. At any rate the complexity of these equations is on the borderline of the possibility of an analytic unfolding of the sample parameters.

In the case of photons, the moments are considerably more complicated due to the fact that they are produced in neutron processes, hence one has to account for both neutrons and photons. It is still possible to derive equations for the measurable quantities of singles, doubles and triples, in a manner similar to that of neutrons. In addition, when accounting for the effect of all source events, for photons one has to account for the possibility of a single photon source which is not connected to the neutron chain. An alpha decay that did not lead to a (α, n) reaction, and thus no neutron would be an example of this, but also other reactions producing single photons (of sufficient energy) uncorrelated to neutron emission would be of importance. This can be made in a way analogous to the accounting for the processes (α , n) for neutrons. Defining γ as the ratio between the single photon source strength, Q_{y} , and the total neutron source intensity Q_s , i.e. $\gamma = Q_{\gamma} / Q_{s}$, the gamma singles can be expressed as:

$$S_{\gamma} = \varepsilon_{\gamma} \left[\gamma F(1 + \alpha v_{sf,1}) + F(1 + \alpha v_{sf,1}) \times \left(7\right) \\ \times \left\{ \frac{\mu_{sf,1} + \alpha v_{sf,1}}{(1 + \alpha v_{sf,1})} + \frac{v_{sf,1}(1 + \alpha)}{(1 + \alpha v_{sf,1})} \mathbf{M}_{\gamma} \right\} \right] = F \varepsilon_{\gamma} \left[\gamma (1 + \alpha v_{sf,1}) + \mathbf{M}_{\gamma} \left\{ \mu_{sf,1} + v_{sf,1}(1 + 2\alpha) \right\} \right]$$

For doubles and triples the expressions grow longer:

$$D_{\gamma} = \frac{\varepsilon_{\gamma}^{2} F}{2} \left[\mu_{sf,2} + 2(\mu_{sf,1} + \alpha v_{sf,1}) \frac{v_{sf,1}(1+\alpha)}{(1+\alpha v_{sf,1})} \mathbf{M}_{\gamma} + v_{sf,2} \mathbf{M}_{\gamma}^{2} + v_{sf,1}(1+\alpha) g_{2} \right]$$
(8)

$$T_{\gamma} = \frac{\varepsilon_{\gamma}^{3} F}{6} \left[\mu_{sf,3} + 3\mu_{sf,2} \frac{v_{sf,1}(1+\alpha)}{(1+\alpha v_{sf,1})} \mathbf{M}_{\gamma} + \frac{(9)}{(1+\alpha v_{sf,1})} + 3(\mu_{sf,1} + \alpha v_{sf,1}) \left\{ \frac{v_{sf,2}}{(1+\alpha v_{sf,1})} \mathbf{M}_{\gamma}^{2} + \frac{v_{sf,1}(1+\alpha)}{(1+\alpha v_{sf,1})} g_{2} \right\} + v_{sf,3} \mathbf{M}_{\gamma}^{3} + 3v_{sf,2} g_{2} + v_{sf,1}(1+\alpha) g_{3} \right]$$

² In the multiplicity rates we shall use the better known notation *F* for the intensity of the spontaneous fissions, Q_f (also referred to as "spontaneous fission rate" in the literature).

with ε_{γ} being the detection efficiency of gamma photons and \mathbf{M}_{γ} the photon leakage multiplication per initial neutron. In (8) and (9), the notations g_2 and g_3 stand for the second and third factorial moments of the total number of gamma photons generated by one single neutron. These are not given here; they can be found in [4] and [6]. Hence the complexity of the above equations is larger than it looks at the first sight.

Likewise, expressions for the mixed moments, one double $(n\gamma)$ and two triples, $(nn\gamma)$ and $(n\gamma\gamma)$ can be formulated. Again, these are found in [6] and are not given here for brevity. It can be noted however that due to the mixed rates containing both neutron and gamma parameters such as the detection efficiencies, they are strong candidates for being successfully used to unfold many sample parameters.

3. ANN application

As mentioned previously, unlike for the neutron expressions, the complexity of the expressions for the gamma photons prevents the possibility of using analytical inversion of the photon multiplicity rate expressions. Hence we propose the use of artificial neural network (ANN) techniques for the unfolding of sample parameters from some or all of the measured multiplicity rates.

The use of ANNs can be tested on the familiar case of neutron multiplicities. This can therefore serve also as a first test. In addition, it offers the possibility to test one novel aspect of the ANN techniques. Namely, the analytical inversion of the neutron multiplicity rates is only possible as long as only three unknowns are attempted to be retrieved from the three multiplicity rates. This has the effect that one parameter, usually the neutron efficiency, needs to be known in advance. With ANN techniques, there is a larger flexibility, since ANNs can utilize the rich information in the non-linearity of the expressions to unfold more parameters than the number of expressions. Hence there is a chance that in addition to the usual three parameters, also the detector efficiency can be retrieved.

In this respect one can draw analogies between the above statement and the use of the Feynman-alpha method for determination of the reactivity. In the Feynman-alpha method, there is one single expression giving the dependence of the relative variance of the detector counts on the measurement time length. This expression contains both the searched prompt neutron decay constant alpha, but also the unknown detector efficiency. However, due to the non-linear dependence of the formula on the measurement time, both parameters can be determined by a curve fitting method.

3.1. Validating the ANN

Analytical unfolding of the parameters F (fission rate), M (leakage multiplication) and α (alpha ratio) for the known neutron detection efficiency is feasible from three neutron multiplicity rates. Therefore the neutron equations were used to verify the ANN application accuracy in the sample parameters unfolding. The first ANN calculation was performed to unfold the aforementioned three sample parameters and one value of the neutron detection efficiency from the neutron singles, doubles and triples rates. The analytical expressions were used, by sweeping with the parameters F, M and α over realistically possible values, to generate input patterns for the training of a simple feedforward backward propagation network with three inputs and four outputs. The training data set, consisting of the three neutron multiplicity rates for fissile sample parameters in a wide range corresponding approximately to Pu mass of 0.05 kg to 5 kg and for one value of the neutron detection efficiency of 0.5 (for an organic scintillation detector in energy range of fast neutrons), is shown in Fig. 1.

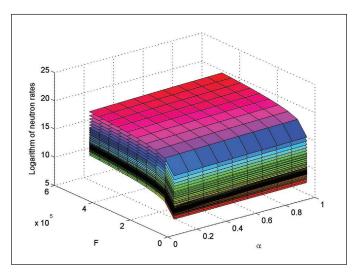


Figure 1: Training data used which are calculated for different values of *F*, p and α . The different surfaces correspond to different values of p while *F* and α are indicated on the x, y-axis. The z-axis is the logarithm of the three neutron rates for visual presentation of the training data.

About 20 % of the whole input set, consisting of up to tens of thousands of points, is used as the patterns for the ANN validation and testing. Preprocessing of the training data was carried out by normalizing the inputs and targets so that they have zero mean and a standard deviation equal to unity.

The network structure and complexity, the training algorithm and activation functions were determined by trial and error. We have used the multi-layered perceptron consisting of an input layer, an output layer and due to complexity of the problem, 2 hidden layers with 25 and 15 nodes, respectively. The number of the input and output nodes is defined by the problem itself, which means we have used the 3 input nodes for the single, doubles, and triples neutron rates and 4 nodes in an output layer for F, p, α and one value of the neutron efficiency . A network structure with "tansig" transfer function in both hidden layers and a linear transfer function in the output layer was used to unfold the four parameters mentioned before. The tan-Sigmoid transfer function looks as follows:

$$\tan sig(n) = \frac{2}{(1 + \exp(-2n))} - 1$$
 (10)

A schematic outline of the constructed neural network is shown in Fig. 2.

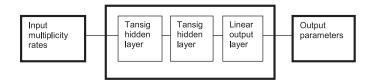


Figure 2: Schematic outline of the artificial neural network.

A few of the modified backpropagation (BP) algorithms were examined for the training of the network. The Levenberg-Marquard (LM) algorithm was found to be best suited for this problem since it is reasonably fast and provides the results with the highest accuracy compared to other algorithms available in the toolbox with the technical computing software MATLAB [11].

After 10000 epochs in the training, the relative errors on the whole neutron rate data set, including the training, validation and testing data, for all parameters (given in Table 1) were reduced to the value less than 2.1*10⁻³ %. To reach very high accuracies, longer training times for the ANN is beneficial. The data presented in this paper had training times in the order of ten hours. The values are related to the largest errors, while mean values of the errors for all parameters are much smaller. These results show that the parameters from the three input neutron rates can be unfolded with high accuracy by using the ANN.

parameters	F	α	р
Max. rel. error (%)	0.0001	0.0021	0.0001

Table 1: The maximum values of the relative errors(%) for the investigated parameters unfolded fromthe neutron rates.

However, the relative errors of the four above mentioned unfolded parameters for the case of two extreme values of the neutron detection efficiency (of 0.1 and 0.5 for an organic scintillation detector) are somewhat higher compared to the previous case with one value of the neutron efficiency, but still less than about 0.6 % for all parameters after 1000 epochs. The results are given in Table 2. Due to the overlapping of the input data to some extent for the neutron efficiency in the whole range, it was not possible to achieve convergence even with alternative structures of the network and larger numbers of the nodes in the hidden layers. However, it can still be demonstrated that by using either the neutron or mixed multiplicity rates one can extract good predictions for an underdetermined system (more unknowns than equations). Especially the mixed rates are suitable for this purpose due to their complicated form (1 double and 2 triples rates), where each variable occurs multiple times.

parameters	F	α	Р	E _n
Max. rel. error (%)	0.0265	0.5518	0.3233	0.4923

Table 2: The maximum values of the relative errors (%) for the investigated parameters from the neutron rates for 2 values (0.1 and 0.5) of the neutron detection efficiency after 1000 epochs.

3.2. Additional inputs

As mentioned previously, the complexity of the expressions for the gamma photons prevents the possibility of using analytical inversion of the multiplicity rate expressions, which was possible for the neutron expressions. The same is valid for the expression related to the mixed multiplicity rates.

In the case with 6 input rates (3 neutron and 3 gamma rates), the network structure remained the same but with 6 inputs and 6 outputs, as well as more nodes in the hidden layers (i.e. 30 and 20 nodes, respectively). For the same range of the input parameters, one value of the neutron efficiency of 0.5 and seven various values of the gamma efficiency in the range from 0.1 to 0.4 were used. Fission rate, being the most important parameter for evaluation of sample mass, was unfolded with a relative error less than about 0.02 %. Since the training procedure was going smoothly, smaller relative errors can be expected for more training iterations. The histograms of the relative errors for a few unfolded parameters are given in Fig. 3. Using the neutron and gamma multiplicity rates, more inputs are available, but at the same time the number of parameters increases similarly. Some increased errors are visible, but generally the ANN predictions are good.

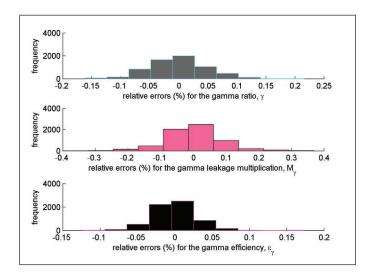


Figure 3: Histograms of the relative errors for a few unfolded parameters.

The complexity of the expressions for nine autoand cross factorial moments and corresponding multiplicity detection rates which prevents analytical solution is even more apparent, hence this case exemplifies even better the advantage of using the ANN approach. The use of 3 neutron, 3 gamma and 3 mixed multiplicity rates in the sample unfolding represents an overdetermined system with 9 measurable quantities exceeding the number of unknowns, i.e. sample parameters. We have demonstrated that the sample parameters such as fission rate, the probability of induced fission p, the alpha ratio, the gamma ratio, and the gamma detection efficiency can be unfolded with small relative errors from 9 input multiplicity rates. The maximum values of the relative errors (%) of the unfolded parameters are given in Table 3. The neural network was constructed with two hidden layers with 35 and 15 nodes, respectively, and tansig activation functions in both layers.

3.3. Investigation of the ANN accuracy with different outputs

We have investigated the influence of various combinations of the target parameters in the training of the neural network on the accuracy of the unfolded parameters for the case with three neutron input rates. The results of the analysis show that combination of the three parameters in the following arrangement [F (1+ α) p], where (1+ α) is a recurring expression in equations for S, D and T rates, compared to the other combinations of the target parameters, generates the smallest relative error for all three parameters in the ANN unfolding. This is illustrated in Fig. 4 for the alpha ratio. Hence, by selecting the output parameters in a suitable way, there is a possibility to increase the accuracy of the ANN, by essentially taking out part of the complexity of the problem from the neural network. The arrangement of variables investigated, were also tested when applying noise to the input data with a magnitude of 5% (the concept of "noise" is described below). Also in those cases the same [F (1+ α) p] setup provided the smallest variation in the output parameters.

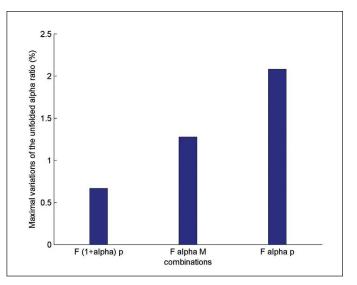


Figure 4: Influence of combination of the target parameters on the accuracy of alpha ratio.

4. Sensitivity analysis

So far, in all analysis, "clean" input data were used as inputs to the ANN. In other words, the multiplicity rates were calculated from the analytical expressions which are based on the exact theoretical expressions

Parameters	α	р	F	γ	\mathcal{E}_{γ}
Max. rel. error (%)	1.7500e+00	6.0842e-02	2.4592e-02	2.6908e-01	1.3126e-01

Table 3: The maximum values of the relative errors (%) for the investigated parameters from the network with 9 inputs (3 neutron, 3 gamma and 3 mixed rates) and 5 outputs.

for the moments. The results of a measurement, on the other hand, are not the exact values of the moments, rather their estimates based on a finite length measurement, and are inevitably inexact, and can even contain further inaccuracies due to background effects, measurement errors, etc.

Hence it can be interesting to investigate the robustness of the inversion procedure by simulating deviations from the exact multiplicity rates by modifying these latter before using them in the inversion algorithm. Since the ANN works in a non-parametric way, such an investigation can only be performed numerically. The method used here consists of adding a random number to each input data, taken from a Gaussian distribution with zero mean and a variance equal to a chosen percent of the variable in question. This process will be called "perturbing the input data" and the modification will be called a "noise". Thus, a "noise level of 5%" means adding a random number sampled from a Gaussian distribution with zero mean and a variance being 5% of the "clean" value which is modified by adding the noise.

4.1. Neutron sensitivity analysis

The effect of perturbation of the neutron rates, i.e. singles (S), doubles (D) and triples (T) with normally distributed random noise of different levels (1 %, 5 % and 10 % in magnitude) was investigated. The analysis was performed by application of a single-variable perturbation method, which means that each variable has been independently perturbed, while all other variables remained unchanged. Although the variables are not independent, useful information on the relative importance of the input multiplicity rates in the ANN approach can be obtained.

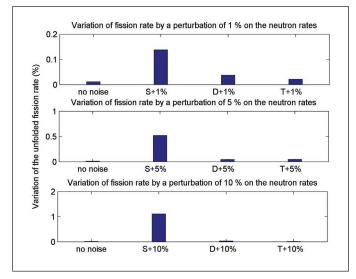


Figure 5: Influence of noise on different neutron rates, when unfolding the fission rate.

We observed that a perturbation of the singles neutron rates by different levels of noise has the largest effect on the relative errors for all three unfolded parameters, compared to the errors obtained after perturbing the double and triples rates with the same level of noise, see Fig. 5. The constructed neural network shows robustness to perturbation of the doubles and triples rates (which is a consequence of the specific arrangement of the training input data). Since the measured triples rates show the largest relative statistical uncertainty [12,13], the use of the ANN based unfolding can contribute to the evaluation of the searched parameters with small uncertainty, since the ANN approach shows the highest sensitivity to inaccuracies in the measured singles neutron rates which can be measured with the smallest relative statistical error. This is one of the promising characteristics of the ANN application in the parameter unfolding from the multiplicity rates.

4.2. Sensitivity analysis of gamma and mixed moments and of omission of inputs

We have applied a single variable perturbation model varying only one of the inputs (the full network with nine inputs and five outputs) by adding a random noise level of 5 % in magnitude, while holding all other variables constant to see how the perturbations affect the ANN unfolding process. In spite of dependence between inputs, within the context of the ANN approach it is useful to apply sensitivity analysis and to rank the inputs by their relative importance. The uniformly distributed perturbations (normal random noise) provide information on the reliability of the unfolded parameters produced by the ANN approach. The results of the neural network response with 9 inputs and 5 outputs to the input rate perturbations are presented in Fig. 6.

It can be seen that the largest variation for *F* is still less than 0.4 % when one of the inputs is perturbed by 5 % noise. The results obtained indicate that a higher relative importance can be assigned to the gamma (especially D_{γ}) and mixed multiplicity rates with respect to the neutron rates. The gamma inputs contain information on all five relevant outputs in contrast to the neutron inputs which contain values for only three output parameters and the mixed rates that contain the data on four outputs. For output parameters other than the spontaneous fission rate *F* the relative importance of the inputs might be different. This was though not investigated here.

An additional sensitivity analysis was performed with the network with 9 inputs and 5 outputs, based on eliminating one single input in each run (i.e. in

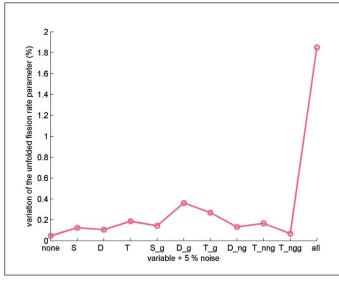


Figure 6: Maximal variations of the unfolded fission rate due to perturbation of the input rates.

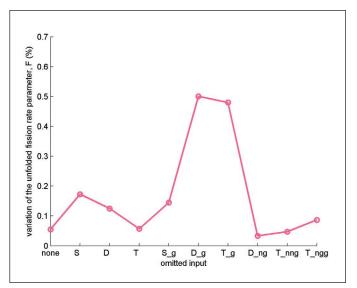


Figure 7: Maximal variations of fission rate after omitting one of the inputs.

each run we used 8 inputs). The maximum variations of the unfolded parameters after omitting one input are given in Table 4 and Fig. 7.

As Fig. 7 shows, once again we can observe that the importance of the gamma rates is higher than the others, due to all the parameters being part of those expressions. This type of analysis also serves a purpose to show which input parameter could be safely discarded if a measurement shows abnormal values on a certain rate.

5. Conclusions

The present paper shows that by taking all possible auto- and cross factorial moments of the neutron and gamma counts into account, one has nine expressions which are functions of six independent sample parameters and the two detector efficiencies.

It is demonstrated that these multiplicity rates can be inverted by non-linear non-parametric least squares methods, namely with the use of artificial neural networks (ANN), to which the above mentioned equations can be used to generate training data. Final validation and further development of the ANN is ongoing. The results are very promising and of good accuracy. When adding noise to the training data to simulate measurement uncertainties, the induced uncertainties for the ANN can be kept very low. In this work we have demonstrated the stability and robustness of the ANN unfolding technique. The training and performance of ANN's using all moments for both neutrons and photons is computationally more demanding, but still within a manageable range.

The uncertainties in the parameter unfolding by ANN techniques are the largest when there are un-

Omitted	Maximal variations (%)				
input	a	р	F	Ÿ	ε,
none	3.1641e+00	2.0865e-01	5.5202e-02	1.1875e+00	4.1813e-01
S	2.0972e+01	7.8709e-01	1.7281e-01	4.2750e+00	8.0476e-01
D	4.8590e+00	3.1847e-01	1.2484e-01	1.7010e+00	5.6626e-01
Т	6.5730e+00	6.1161e-01	5.6854e-02	1.6319e+00	6.5732e-01
S_g	1.4703e+01	5.2398e-01	1.4462e-01	3.3666e+00	2.0104e+00
D_g	1.2206e+01	7.9303e-01	5.0052e-01	5.1280e+00	5.0176e+00
T_g	9.1300e+00	2.4526e+00	4.7984e-01	1.8486e+01	2.1005e+01
D_ng	1.8643e+00	5.3483e-02	3.3181e-02	8.8273e-02	1.3911e-01
T_nng	2.2420e+00	8.7395e-02	4.7197e-02	1.6736e-01	1.9046e-01
T_ngg	3.5588e+00	8.3363e-02	8.6446e-02	3.6945e-01	3.5037e-01

Table 4: Maximum variations of the unfolded parameters by removal one input in each run. Legend: S, D, T -neutron rates, S_g, D_g, T_g - gamma rates, D_ng, T_nng, T_ngg - mixed rates.

certainties in the training data for singles, while in measurements the opposite behaviour is shown: the singles are measured with the highest accuracy, while doubles and triples have higher associated statistical uncertainties. Therefore, applying ANN to this problem has the strength of being least sensitive for the parameters whose experimental determination is the least accurate.

The ANN-based method was proven to be very versatile and can also be used to replace current analytical unfolding methods used for pure neutron measurements due to its smaller sensitivity to measurement inaccuracies. However, the greatest potential of the ANN approach lies with the use of joint neutron and photon multiplets, a problem which cannot be easily solved analytically even when using the point model formalism.

6. Acknowledgements

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COMPUCEA: A High-Performance Analysis Procedure for Timely On-site Uranium Accountancy Verification in LEU Fuel Fabrication Plants

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Abstract

COMPUCEA (<u>Com</u>bined <u>P</u>rocedure for <u>U</u>ranium <u>C</u>oncentration and <u>E</u>nrichment <u>A</u>ssay) is used for on-site analytical measurements in support of joint Euratom-IAEA inspections during physical inventory verification (PIV) campaigns in European Low-Enriched Uranium (LEU) fuel fabrication plants. The analytical technique involves the accurate determination of the uranium element content by energy-dispersive X-ray absorption edge spectrometry (L-edge densitometry) and of the ²³⁵U enrichment by gamma spectrometry with a LaBr₃(Ce) detector. For evaluation of the LaBr₃ spectra a modified version of the NaIGEM code is used, which has recently been adapted to handle the presence of reprocessed uranium.

This paper describes the technique, setup and calibration procedure of the instrument. Results from PIV campaigns in 2007 and 2008 are presented, which demonstrate the performance of the technique. First results obtained with a sandwich detector configuration for enhanced detection efficiency of the passive gamma spectrometry are discussed.

Keywords: uranium elemental analysis; uranium enrichment; X-ray absorption edge spectrometry; lanthanum bromide detector; NaIGEM analysis code.

1. Introduction

The <u>Combined Procedure for Uranium Concentra-</u> tion and <u>Enrichment Assay</u> (COMPUCEA) represents a testing method for the uranium element and ²³⁵U-enrichment assay routinely applied to the analysis of uranium product materials (uranium oxide powders and sintered uranium oxide pellets). The actual analyses on this type of sample materials are performed with mobile equipment in different European fuel fabrication plants for <u>Low-Enriched Ura-</u> nium (LEU) fuels during the nuclear material accountancy verification activities of international nuclear safeguards authorities (Euratom, IAEA). The analytical support provided on site by analysts from the ITU during the PIV campaigns involves the accurate determination of the uranium element content and of the ²³⁵U enrichment in verification samples selected by the Safeguards inspectors according to a defined sampling plan.

The missions made with COMPUCEA are unique because they represent, to our knowledge, the only example in the measurement practice of Safeguards, where high-accuracy analytical measurements for accountancy verification are directly performed in field with mobile analytical equipment. The benefits of this practice are obvious: analysis results are immediately available for a timely conclusion of the PIV, observed discrepancies can be investigated and (in most instances) solved directly on the spot, and sample shipment to remote Safeguards laboratories is avoided or at least significantly reduced.

1.1. Responsibilities

The know-how and the expertise for the dedicated analyses with COMPUCEA have been established at ITU, which is also involved in further development work to upgrade the measurement techniques. DG TREN, the IAEA and ITU therefore agreed that ITU takes over the responsibility for the in-field measurements with COMPUCEA during the PIV campaigns, and for the COMPUCEA technique in general. For ITU this means (i) to send qualified analysts in field (acting as analyst only and not as inspector), (ii) to prepare the equipment for the missions (including pre-calibration), (iii) to maintain and to upgrade the equipment, as currently done, and (iv) to validate new instrumentation and procedures. The equipment itself is procured and owned by DG TREN.

Every year, a joint meeting between DG TREN and ITU is held prior to the PIV campaigns for the planning of the respective missions. The dates for the PIVs, normally fixed by the plant operators, are mostly concentrated within a relatively short period in the months of July and August. The COMPUCEA equipment then travels between ITU and the plants located in Sweden, Belgium, France, Germany and Spain, or often also directly between the plants. The logistics for instrument transportation is handled by DG TREN in close cooperation with ITU. Occasionally, two PIV campaigns occur at different sites in the same week. ITU and DG TREN therefore keep 3 fully equipped COMPUCEA systems available, ready for use.

A typical PIV campaign in a LEU fuel fabrication plant is carried out within a period of 1 week. On average, ITU sends 2 analysts to each site to perform the in-field analyses during this week. For a timely and conclusive termination of the PIV, the analytical results have to be handed over to the inspectors by Thursday of the respective week at the latest. In order to keep this strict deadline, the ITU analysts sometimes arrive on site 2-3 days in advance for instrument preparation and calibration.

2. Analytical procedure

Although the basic measurements made with COM-PUCEA are of radiometric nature, the complete analysis procedure yet represents more a kind of combined chemistry-spectrometry analysis involving careful analytical steps (like quantitative sample dissolution, solution density measurements, quantitative aliquoting etc) than a mere spectrometry measurement. The general scheme of analysis followed in the measurements with COMPUCEA [1] is outlined in Figure 1.

It includes the following 3 main steps:

1. Sample preparation: The purpose of this first step is to transform the solid uranium samples (powders or pellets) into a nitric acid solution of approximately constant acidity (3 M) and uranium concentration level, and then to carefully characterise the obtained solution for its density and temperature. The nominal uranium concentration is set to be around 190 mgU/ml, which is close to the upper limit of the linearity range of the new L-edge densitometry measurement [1]. The analytical tools needed for this sample preparation step (hot plate, density measurement device, glass ware, pipettes etc) is brought on site as part of the COMPUCEA equipment, but the use of operator facilities (fumehood) is also required at this stage. This also holds for the later

disposal of the (non-problematic) liquid waste in the form of pure nitric acid uranium solutions.

- 2. <u>Radiometric measurements (L-Edge Densitometry and Gamma Spectrometry)</u>: Aliquots are taken from the sample solution and subjected, without any further treatment, to parallel L-edge densitometry and passive gamma counting measurements. The two radiometric measurements are described in more detail below.
- 3. Data evaluation: In the final step of data evaluation, the different pieces of information obtained from the sample preparation and from the two radiometric measurements are combined to evaluate the uranium weight fraction in the original sample and the ²³⁵U weight fraction in the uranium material. It should be noted that the two radiometric measurements are interdependent, i.e. each technique requires input from the other for final data evaluation: the L-edge densitometry measurement needs the knowledge of the enrichment for the calculation of the uranium atomic weight, and the gamma measurement needs as input the knowledge of the uranium concentration. The evaluation of the final uranium concentration and enrichment is therefore made in an iterative manner.

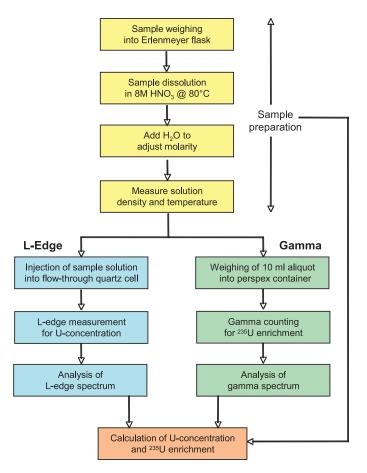


Figure 1: Scheme of analysis for COMPUCEA.

2.1. Uranium elemental assay

In the 2nd generation of the COMPUCEA equipment, a miniaturized 30 kV/100 μ A X-ray generator, and a Peltier-cooled, high-resolution 10 mm² x 0.5 mm Si drift detector are used to measure the energy-differential absorption at the L_{III}-shell absorption edge, which occurs for uranium at the energy of 17.17 keV. A representative measurement example together with a cross-sectional view of the equipment is shown in Fig. 2. In this setup, the sample cell consists of a fixed flow-through quartz cell with a path length of 2 mm, and a cell volume of 125 μ l.

The evaluation of the uranium concentration from the measured ratio of photon transmission across the L_{III} edge at 17.17 keV follows the proven analysis procedure adopted for K-edge densitometry with an X-ray continuum [2]. In this approach, the photon transmission as a function of energy, T(E), is measured relative to a blank spectrum from a nitric acid solution of representative molarity (3M), and then linearized in a representation lnln(1/T) vs lnE. Linear least-squares fits to the respective data on both sides of the absorption edge determine the photon transmission at energies slightly displaced from the absorption edge ('non-extrapolated fitting mode', $E_{+} = 17.60 \text{ keV}, E_{-} = 16.70 \text{ keV}$, or directly at the absorption edge energy ('extrapolated fitting mode'). Fitting intervals ranging from 15.50-16.70 keV, and from 17.60-18.80 keV were chosen for the evaluation of the transmission ratio across the L_{III} edge.

In the 'extrapolated fitting mode', where the photon transmissions are determined directly at the absorption edge energy, the uranium volume concentration ρ_U (in g/cm3) in the measured solution is obtained by the following relation:

$$\rho_U = CF_{AW} \cdot \left[\frac{\ln[T(E_-)/T(E_+)]}{|\Delta \mu_U| \cdot D} \right]$$
⁽¹⁾

Here, the quantity $|\Delta \mu_U|$ (in cm²/g) describes the difference in photon mass attenuation coefficients of uranium at the energies E₋ and E₊ and D (in cm) denotes the path length of the photon beam through the sample cell. The term CF_{AW} is a correction factor accounting for the atomic weight of the uranium under analysis. It is calculated from the known enrichment. The uranium concentration derived in the 'extrapolated fitting mode' from the transmission ratio directly at the L-edge energy is virtually insensitive to matrix effects.

However, the availability of two independent analysis results for the uranium concentration from the extrapolated and non-extrapolated fitting analysis represents a very useful diagnostic tool. A statistically significant difference observed between the two results will immediately point to any sort of deviation in the matrix composition of the measurement sample from the assumed 3M HNO₃ reference matrix. This knowledge is not of immediate relevance for the L-edge densitometry measurement, but of practical help for the parallel enrichment measurement, where any deviation in the matrix composition will have a direct influence on the gamma attenuation behaviour of the sample.

A practical example refers to the analysis of uranium samples containing a significant amount of gadolinium. In this case the difference observed between the uranium results from the non-extrapolated and extrapolated fitting analysis can be used for an estimate of the Gd content, provided the discordance between the two evaluated uranium results can be reasonably attributed to the presence

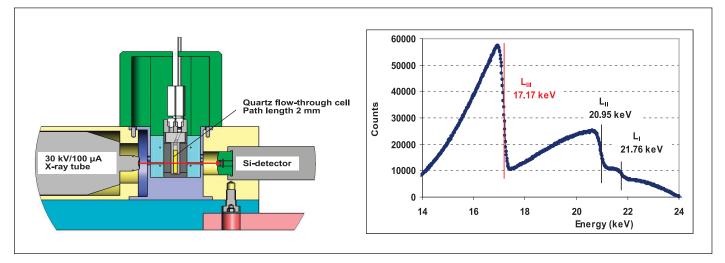


Figure 2: L-edge absorptiometry with an X-ray continuum used in COMPUCEA 2nd generation. Left: Measurement setup. Right: Measurement example.

of this additional element alone. The knowledge about the Gd content then allows calculating corresponding correction factors for the enrichment measurement.

2.2. ²³⁵U enrichment determination

The ²³⁵U enrichment measurement in COMPUCEA is based on the counting of the ²³⁵U 186 keV gammas of a defined amount of uranium in solution in a well-defined counting geometry. The new detector replacing the previous HPGe well detector is a standard-type 2" x 1" cerium-doped lanthanum bromide scintillation detector – LaBr₃(Ce). It offers the main advantage of being a detector operating at room-temperature, therefore eliminating the need for detector cooling with liquid nitrogen as required before. This practical advantage, particularly for infield applications, largely compensates for the drawback of an inferior energy resolution (FWHM @ 186 keV about 9 keV for the LaBr detector compared to a value of 1.3 keV obtained with the previous HPGe well detector). Fortunately, the relatively simple gamma spectrum of ²³⁵U allows accurate enrichment measurements also at this lower degree of energy resolution [3].

Since the recently developed $LaBr_3(Ce)$ scintillation detectors are not yet available in the form of welltype detectors, the configuration of sample counting was changed to a counting geometry with the sample located on top of the detector, as shown in Figure 3. As this measurement geometry is less efficient than counting in a well detector, the sample volume for the measurement with the LaBr₃(Ce) detector was increased from 2.5 ml (as used before in a HPGe well detector) to 10 ml.

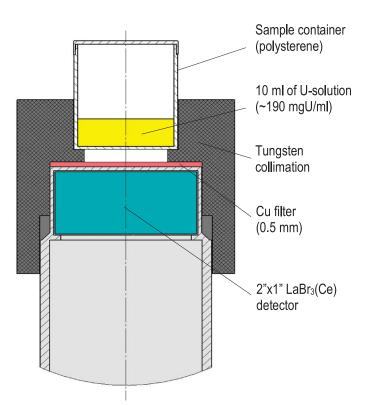


Figure 3: Configuration of the ²³⁵U enrichment measurement with a LaBr₃(Ce) scintillation detector.

The proper evaluation of the measured gamma spectrum for an accurate enrichment determination involves a two-step process: (1) analysis of the gamma spectrum itself for the extraction of the 185.7 keV net peak counts, and (2) the calculation of appropriate correction factors for the extracted peak counts accounting for the impact of variable sample parameters. In the first step, a modified analysis code based on the NaIGEM code previously developed for the analysis of uranium gamma spectra measured with NaI(TI) scintillation detectors [4] is used for the deconvolution of the gamma spectra

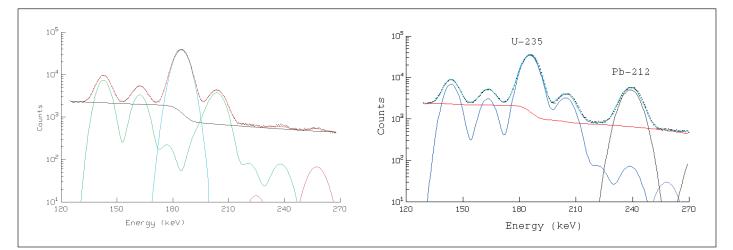


Figure 4: Examples of fitting graphs for LaBr₃ gamma spectra taken from low-enriched virgin uranium (left) and recycled uranium (right) samples.

measured with the new LaBr3 scintillation detector. Examples for fitting graphs are shown in Figure 4.

The software for spectrum analysis determines the net peak counts for the most prominent ²³⁵U gamma line at 185.7 keV with associated uncertainty. Additional information is provided on the quality of the response function fitting, and on the full width at half maximum value determined for the 185.7 keV line. The fitting code has recently been adapted to handle gamma spectra from recycled uranium materials which show an additional gamma ray at 238.6 keV originating from the ²³²U descendent ²¹²Pb (see example on the right side in Fig. 4). For those spectra, the analysis code also reports the peak area with associated uncertainty for the 238.6 keV line.

In the second step of the analysis, correction factors for the evaluated 185.7 keV net peak counts are calculated from known sample parameters (see also Section 3.3.). The major part of this calculation work is accomplished by means of a tailored Monte Carlo simulation of the gamma detection process. The simulation software is able to calculate, on a relative basis, for the modelled measurement configuration the detection rates for the 185.7 keV photons in dependence of crucial sample parameters. The relative detection rates calculated within a runtime of 100 s for the Monte Carlo calculation have a statistical precision of ca. 0.03%.

The sample parameters taken into account in the Monte Carlo calculation include:

- the uranium concentration,
- the gadolinium concentration (if this element has been detected),
- the solution density (representing also a measure for the HNO₃ molarity),
- the bottom thickness of the sample container, and
- the sample volume.

The relative detection rate delivered by the Monte Carlo simulation program is normalised to the detection rate calculated for a reference sample, yielding a final correction factor $CF(U,Gd,\rho,Bot,V)$ for the measured 185.7 keV net peak rate.

Another correction factor, *CF(Pa)*, accounting for a small contribution of interfering Pa gamma rays to the 185.7 keV line, is calculated separately. The ²³⁸U daughter products ²³⁴Pa and ^{234m}Pa, which normally are in secular equilibrium with ²³⁸U, emit weak gamma rays with energies at 186.15 and 184.7 keV, which are close to the main ²³⁵U gamma ray at 185.72 keV and cannot be resolved from the ²³⁵U gamma ray.

Their contribution to the observed peak intensity at 186 keV, though very small, should be not ignored in high-accuracy enrichment measurements. The numerical value for CF(Pa) calculates from the respective photon emission probabilities to:

$$CF(Pa) = 1 - \frac{(100 - enr)}{enr} \cdot 1.2223 \cdot 10^{-5}$$
 (2)

where *enr* denotes the ²³⁵U enrichment in wt%. The correction can be only applied when the enrichment is approximately known. The numerical factor in Eq. 2 represents the ratio of the emission rates (²³⁴Pa+^{234m}Pa)/²³⁵U.

3. COMPUCEA calibration

Both measurement techniques in COMPUCEA require an instrument calibration. The calibration approach has been revised and simplified in the sense that for each technique in principle only a *single calibration factor* needs to be determined. In order to arrive at this favourable situation, measurement and instrument properties considered as being relevant for measurement performance and calibration have been carefully studied during the instrument development stage.

Prior to the in-field measurements, all COMPUCEA systems are calibrated at ITU with a set of suitable reference solutions. With this pre-calibration, combined with the quantitatively known correction factors to be applied, calibration in field is reduced to the measurement of two calibration samples for a verification or re-normalisation of the basic calibration factors determined at ITU.

3.1. Reference materials

The reference materials available for calibration consist of a set of sintered UO₂ pellets with 3 different enrichment grades (0.72, 2.10 and 4.40 wt% ²³⁵U). The UO₂ pellets were previously taken from the production batches of a uranium fuel fabrication plant, and then subsequently characterized by primary analytical methods for the uranium element content and isotopic composition. The analytical measurements for material characterisation were independently carried out by ITU and IAEA-SAL.

The reference solutions required for instrument calibration, either in field or at ITU, are prepared from these reference pellets, following exactly the same procedures for sample preparation as applied for the normal measurement samples. From each reference material, a single reference solution is prepared, which is used both for the calibration of the L-edge densitometer and of the gamma spectrometer.

3.2. Calibration of the L-edge densitometer

For calibration with the pure uranium reference solutions the densitometry equation (1) is applied. The calibration factor to be determined from the measured transmission ratio $T(E_{-})/T(E_{+})$ is the value of $\Delta \mu_{U}$:

$$\left|\Delta\mu_{U}\right| = CF_{AW} \cdot \left[\frac{\ln\left[T(E_{-})/T(E_{+})\right]}{\rho_{U} \cdot D}\right]$$
(3)

The factor CF_{AW} accounts for the uranium atomic weight. For the calibration solutions, the quantities CF_{AW} and ρ_U are known reference values. The path length D of the measurement cell is also known (D = 0.2 cm), but specified from the manufacturer with a relative uncertainty of 0.5%. This uncertainty has no influence on the overall measurement uncertainty as the same cell is used for the calibration and all measurements. For the evaluation of $\Delta\mu$ U the nominal value D = 0.2 cm is used. Any deviation of the true path length from this nominal value is then accounted for in the determined value of $\Delta\mu_U$.

The high degree of measurement reproducibility is illustrated by results from calibration measurements performed with the same instrument at four different locations (in four different European countries). The L-edge calibration factors $\Delta\mu$ obtained from the four independent calibration exercises are listed in Table 1. The data prove an extraordinary high degree of reproducibility. The low (statistical) measurement uncertainty of the individual calibration factors is due to the large number of repeat measurements (typically of the order of 10) performed in each case. The counting time used for the calibration measurements is usually also somewhat longer (typically 5000 s).

Location	Date	Extrapolated fitting	
		Δμ (cm²/g)	1s uncertainty (%)
1	06/2007	60.7251	0.029
2	07/2007	60.7120	0.017
3	07/2007	60.7064	0.028
4	08/2007	60.7033	0.021
Mean		60.7117	
RSD (%)		0.0159	

Table 1: Reproducibility of calibration factors $\Delta \mu$ determined for a COMPUCEA system at four different locations.

3.3. Calibration of the gamma spectrometer

The main task of this calibration is to establish a proportionality or calibration factor, *K*, relating the amount of ²³⁵U in the measurement sample, $M(^{235}U)$, to the measured and evaluated net peak counts in the 186 keV line from ²³⁵U, P_{186} . For the calibration (and for the sample measurements later as well) the correction factors $CF(U, Gd, \rho, Bot, V)$ and CF(Pa) mentioned in Section 2.3. have to be taken into account, leading then to the following calibration expression:

$$M(^{235}U) = K \cdot P_{186} \cdot CF(U, Gd, \rho, Bot, V) \cdot CF(Pa)$$
⁽⁴⁾

The determination of the basic calibration factor, K, is made for a defined measurement condition, characterized by the following reference values:

Uranium concentration:	190.00 g/l
Gadolinium concentration:	0.00 g/l
HNO ₃ acidity:	3M
Sample volume:	10.00 ml
Bottom thickness of sample container:	1.10 mm

The corresponding measurement parameters are typically kept close to these reference values, both for the calibration and for the routine measurement samples (except for the Gd content in the routine samples, which is taken as found). The correction factor $CF(U,Gd,\rho,Bot,V)$ applied to the measured 186 keV peak count rate per g²³⁵U, then just represents the ratio of the detection rates calculated for the actual sample and for the above reference conditions. The applied corrections are typically far less than 1%. With the adopted calibration procedure the calibration for the enrichment measurement therefore simply reduces to the determination of a single calibration factor *K*.

Similar to the procedure for the L-edge densitometer performance, the high degree of measurement reproducibility can be observed from the results of the calibration measurements performed with the same detector at different locations. The calibration factors *K* (cps / mg ²³⁵U) obtained from the independent calibrations at the different sites are listed in Table 2. The data prove a very high degree of reproducibility. The good statistical precision of the individual calibration factors is due to the larger number of repeat measurements (typically of the order of 10) performed in each case. The counting time used for the calibration measurements is usually also somewhat longer (typically 5000 s).

	Location	Calibration factor (cps/mg U)	1s uncertainty (%)
Detector 1:			
	1	3.9172	0.15
	2	3.9125	0.14
	3	3.9133	0.14
	1	3.9172	0.14
	Mean	3.91505	
	RSD (%)	0.06	
Detector 2:			
	4	3.7906	0.15
	5	3.7834	0.145
	6	3.7745	0.14
	Mean	3.7828	
	RSD (%)	0.21	

Table 2: Reproducibility of calibration factors *K* for the different sites infield 2008.

4. COMPUCEA performance evaluation and validation

To evaluate the performance of the COMPUCEA 2nd generation equipment, the influence of relevant measurement parameters, such as working and linear range, matrix effects, counting precision, measurement reproducibility, gamma self-attenuation and counting geometry, was studied in detail [5]. With the identification and quantification of individual uncertainty components, it was then possible to present an estimate of the total uncertainty of the two analytical determinations made. This estimation also includes uncertainty components related to the sample preparation (sample weighing, dissolution and density measurement).

The estimations for the total measurement uncertainties of COMPUCEA 2nd generation are well within the International Target Values (ITV) for measurement uncertainties in the field of International Safeguards for nuclear materials [6], as listed in Table 3. The typical counting times with the 2nd generation of COMPUCEA are 3 x 2000s, however, even with a reduced measurement time of 1000 s, the ITV's are safely met.

Analysis	Total uncertainty (% rel. Std. Uncertainty)		
	2 nd COMPUCEA (3 x 2000 s)	2 nd COMPUCEA (1 x 1000 s)	ITV (1000 s)
U-concen- tration	0.13	0.20	0.25
²³⁵ U abundance	0.26 ^{a)}	0.40 ^{a)}	0.45

a) For a medium enrichment of 2 wt% $^{\rm 235}\text{U}.$

Table 3: Performance data for the 2nd generation ofCOMPUCEA.

For method validation, the measurement performance was then evaluated in three different ways by comparing the COMPUCEA results

- i. with results from parallel analyses made with a primary reference method,
- ii. with well-specified reference values for the quantity of interest, and
- iii. with data obtained in round robin tests

4.1. Validation of U-concentration measurements

During the in-field measurements made with the 2nd generation of COMPUCEA in 2007 and 2008, a total of 115 uranium samples were analysed at 4 different locations. For a subset (taken at 3 of the 4 locations), parallel samples were taken by the IAEA inspectors and sent for analysis with a qualified primary analytical method (potentiometric titration according to the method of Davies and Gray). The measurement uncertainties (1s) for the reference method were specified to be 0.05% for the random error, and 0.05% for the systematic error.

Figure 5 shows the results of the parallel analyses. The COMPUCEA results are in perfect agreement with the titration results (average difference of 0.033%). The combined random uncertainty of both methods (0.11% for COMPUCEA and 0.05% for titration) calculates to 0.12%. The observed standard deviation of 0.11% for the differences between the analysis results is in agreement with this value.

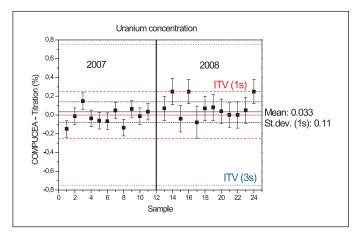


Figure 5: Plot of relative percentage differences between COMPUCEA L-edge in-field analytical results and the primary reference method titration.

The second approach for measurement performance evaluation bases on a comparison of the COMPUCEA analysis results with tightly specified reference values for the quantity of interest. This comparison applies to the uranium content in sintered UO_2 fuel pellets manufactured as reactor fuel. The very tight specifications for the oxygen-to-metal ratio (2.00±0.01) and for element impurities restrict the uranium content in this kind of nuclear materials to the very narrow range of 88.11-88.16 wt%. This margin holds for all pure uranium fuels produced around the world [7]. For the mean value of 88.135±0.025 wt% this uncertainty range means a relative uncertainty of ±0.028%.

Among the total number of 57 samples analysed in 2007 there existed 29 sintered UO_2 pellet samples with this kind of well-specified uranium content. The relative percentage differences between the COM-PUCEA analysis results and the specified reference value for this set of samples were calculated. The data show an average difference of -0.015 % with a relative standard deviation of 0.08 %. Both values are well within the ITV-values (0.015% for systematic, 0.2% for random effects) for the COMPUCEA measurement uncertainty.

A third key element for measurement performance evaluation, and according to the Laboratory Guide to Method Validation [8] the preferred way of validating methods, is the external control through interlaboratory trials. An interlaboratory round robin exercise pertinent to the validation of COMPUCEA is provided by the programme "EQRAIN" (from the French acronym for "Quality Assessment of Analysis Results in the Nuclear Industry") conducted by CEA-CETAMA. Under the EQRAIN programme highly concentrated uranyl nitrate solutions (> 200 gU/kg) of unknown concentration are distributed on a regular basis to interested parties for controlling their analytical methods for uranium analysis. COM-PUCEA is part of this programme for measurement control. The results from 4 round robin tests performed under EQRAIN 12 in 2008/2009 are summarized in Table 4. During each of the rounds, 2 or 3 sub-samples were analysed independently by COMPUCEA. The results obtained prove a high degree of reproducibility and, from a comparison with the reference values communicated after submission of the results, a high degree of accuracy with an average deviation as low as -0.023%.

4.2. Validation of ²³⁵U enrichment measurements

The validation of the enrichment measurements made with the new $LaBr_3$ detector setup bases on a comparison with results obtained with a recognized and validated measurement technique for isotope abundance measurements, i.e. Thermal Ionization Mass Spectrometry (TIMS). During the in-field COMPUCEA measurement campaigns in 2007 and 2008 a total of 16 parallel samples of low-enriched uranium were taken and sent for off-site analysis by TIMS.

The compared data are listed in Table 5. The overall mean difference for the whole set of data calculates to 0.12% with a standard deviation (1s) of 0.11%. If categorized according to the year of analysis, the compared data show a mean difference of 0.07 ± 0.10 % for the campaigns in 2007, and of 0.16 \pm 0.10% for the campaigns in 2008. For the underlying type of enrichment measurements made with a scintillation detector in the 2nd generation of COM-PUCEA, the results demonstrate a remarkable level of performance. Both the observed random and systematic errors are well within the estimated uncertainty of the gamma measurement of about 0.25% for enriched materials, and also well within the uncertainty levels set by the International Target Values.

	Reference values (g/kg)	COMPUCEA result (g/kg)	% relative difference (COMPUCEA-reference)
EQRAIN 12	210.81 ± 0.11	210.75 ± 0.15%	-0.028
1 st round		210.73 ± 0.15%	-0.038
EQRAIN 12	221.40 ± 0.11	221.38 ± 0.15%	-0.009
2 nd round		221.37 ± 0.15%	-0.014
		221.33 ± 0.15%	-0.032
EQRAIN 12	238.14 ± 0.11	238.09 ± 0.15%	-0.021
3 rd round		238.06 ± 0.15%	-0.034
		238.00 ± 0.15%	-0.059
EQRAIN 12	249.70 ± 0.25	249.79 ± 0.15%	0.036
4 th round		249.65 ± 0.15%	-0.022
		249.63 ± 0.15%	-0.029

Table 4: COMPUCEA results obtained under EQRAIN 12. The uncertainties stated are 1s.

Sampling facility	²³⁵ U content obtained by TIMS	Relative difference COMPUCEA-TIMS (%)	1s combined uncertainty (%)
2007:			
А	3.949	0.23	0.28
А	4.700	-0.05	0.28
А	4.287	-0.04	0.29
А	1.616	0.17	0.30
В	4.008	0.05	0.29
В	3.728	0.16	0.29
В	3.412	0.03	0.28
2008:			
В	3.691	0.27	0.21
В	3.699	0.03	0.21
А	4.682	0.01	0.24
А	3.960	0.17	0.26
А	4.422	0.11	0.30
С	4.002	0.32	0.25
С	4.006	0.20	0.25
С	4.003	0.15	0.25
С	3.999	0.18	0.25

Table 5: Comparison of COMPUCEA results and data obtained by TIMS for samples taken in parallel during PIVs in 2007 and 2008. The uncertainties stated are 1s combined uncertainty COMPUCEA and TIMS.

4.3. Upgrade for ²³⁵U enrichment determination: LaBr₃ sandwich detector

In order to further improve the measurement performance of the COMPUCEA equipment, an increase of sample throughput is of relevance during in-field operation, without loss of measurement performance. For ²³⁵U enrichment determination, a counting configuration using two LaBr₃ detectors in a sandwich-type arrangement, promises to significantly enhance the robustness of the counting configuration and the detection efficiency, allowing for shorter counting times and/or improved measurement precision. First experiments were performed using specifically designed sample containers, filled with 10, 12 and 13ml of sample solution. For data evaluation, the two spectra were evaluated separately and the number of counts determined for the 186 keV peaks summed up afterwards. For the 10, 12 and 13 ml samples, an increase of the overall counts of a factor of 2.5, 3 and 3.3, respectively, was obtained, compared to the counting configuration with one detector (and 10 ml sample volume). The reproducibility, both for several repetitions of measurements as well for a series of measurements where the sample container was taken out and put back into position, turned by 180°, was satisfactory,

a repetition of 5 samples led to a random uncertainty of < 0.15%.

The new detector configuration (with 13 ml sample volume) will be tested further during the next in-field campaigns, the automated evaluation of the 2 gamma spectra needs to be implemented in the COM-PUCEA Software package.

5. Conclusion

COMPUCEA is a compact and transportable system which allows high-accuracy uranium elemental assay and enrichment determination from solid uranium samples. The second generation Compucea avoids radioactive sample transport, does not need transport of radioactive sources and attains excellent accuracy with an easily portable system. It is routinely applied in physical inventory verification campaigns at European LEU fuel fabrications plants. The 2nd generation system with a compact L-edge densitometer and LaBr₃(Ce) scintillation detection has been evaluated and validated, the performance is well within the International Target Values. A sandwich detector configuration using two LaBr₃ detectors for ²³⁵U enrichment determination offers improved counting efficiency for higher sample throughput during in-field operation.

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Integrated Analysis of Satellite Imagery for Treaty Monitoring - The LIMES Experience

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Abstract

LIMES (Land/Sea Integrated Monitoring for European Security) is a FP6-funded project which aims at developing satellite-based services for a range of security-related applications such as maritime, land & border surveillance and emergency response. LIMES started in December 2006 and will run until early 2010. Most of the development work has now been concluded and was tested in a number of service demonstrations.

LIMES contains a workpackage focused on Treaty Monitoring, which has the objective to provide an integrated platform supporting the non proliferation image analyst in verifying treaty compliance. The main aspects addressed by the work package are:

- increased automation of the image processing workflow, in particular in the areas of objectbased change analysis, 3D information extraction and processing of radar imagery.
- improved information management using a GISbased platform capable of integrating information from multiple sources and time-frames, including satellite imagery, site models, open source information, reports, etc

The Treaty Monitoring workpackage carried out two service demonstrations in 2008 and 2009 using the nuclear site Olkiluoto (Finland), which hosts a nuclear facility under construction. The demonstration scenario was the monitoring of construction activities using different types of satellite imagery as well as Open Source information. The demonstration and platform validation was performed at the European Satellite Centre (EUSC) and the results were presented to a number of potential users including IAEA and DG-TREN. The paper presents the achievements of the Treaty Monitoring workpackage and in particular the results of the platform demonstrations.

Keywords: GMES; LIMES; treaty monitoring; earth observation; GIS; data processing and integration.

1. Introduction

GMES (Global Monitoring for Environment and Security) is a European initiative for the implementation of services assembling information received from Earth Observation (EO) satellites and ground based information in order to support environmental and security-related applications [1]. The services will be used by environmental agencies, local, regional, national and international authorities, civil protection organisations, etc. LIMES (Land/Sea Integrated Monitoring for European Security) is a FP6 Integrated Project funded by the EU under the security-dimension of GMES [2]. It aims at the development of pre-operational services supporting applications in the fields of i) maritime surveillance, ii) humanitarian relief and reconstruction and iii) land and infrastructure surveillance. LIMES started in December 2006 and the main development activities were finalised in late 2009. The project will be wrapped up with a number of dissemination activities in early 2010.

LIMES includes a workpackage on Treaty Monitoring which is targeted at the non proliferation image analyst who has the task of collecting, managing and analyzing satellite imagery – often in conjunction with data from other sources – and extracting non proliferation relevant information. The analyst generates a report on a country or location of interest and delivers it to the final user. Typically his tasks include:

- the verification of NPT declarations provided by a country, in particular of site declarations submitted as part of the Additional Protocol
- the generation of base-line analyses of nuclear sites
- the continuous monitoring of (construction) activities on nuclear sites.

Following recent developments in non proliferation the image analyst is faced with new and increased challenges, as for example the detection of clandestine nuclear activities and the assessment of an increasing amount of multi-type information. Although satellite imagery is already an important tool in nuclear non proliferation, current usage relies heavily on visual interpretation with little use of automated processing. Furthermore, current analysis tools usually provide an isolated view on satellite imagery with poor integration of collateral data, such as Open Source information, GIS data, internal databases, reports, etc [3, 4].

New satellites such as very high-resolution (VHR) optical and radar sensors further increase the number of possible applications in nuclear non proliferation and, therefore, also the amount of data to be processed. Hence, the Treaty Monitoring work-package aims at providing a platform supporting the image analyst in the forthcoming challenges. It includes the following components, thus contributing to the analyst's efficiency and effectiveness:

- Automatic change detection based on VHR optical data. User-friendly change detection and visualisation allows the analyst to easily assess changes with respect to their relevance for non proliferation. An integrated object-based image classification helps differentiating various types of changes (e.g. vegetation changes versus changes due to construction works).
- Extraction and analysis of 3D information from stereo satellite imagery, which can be used for several purposes including visual analysis, improved geometric correction of the satellite imagery and 3D change detection.
- Very High Resolution SAR (Synthetic Aperture Radar) imagery is becoming increasingly important for the monitoring of nuclear activities. However, visual analysis requires an highly experienced user. The platform incorporates a tool that automatically detects anomalies in SAR imagery by analyzing series of interferrograms taken at different instants in time.
- Integrated, multi-source analysis: The platform provides a single, map-based point-of-entry to

the information required for a specific analysis task, including all satellite imagery, derived products and required collateral and background information. It is designed to facilitate information sharing between analysts as well as to ensure long-term knowledge preservation.

The platform developed within the workpackage is a pre-operational prototype, which aims at demonstrating novel concepts to ease the task of the image analyst in a non proliferation context. The focus of the platform is to answer to the perceived need for integrating information from multiple sources, multiple time-frames and resolutions, for an efficient and thorough non proliferation analysis. This information integration concept should be obviously supported by tools adapted to the types of information to be integrated. The modular architecture chosen enables the integration of today's as well as tomorrow's supporting tools. By its nature, the integration platform is closely linked to the information infrastructure and policy of a given organisation. Therefore, it needs to be customized to an organization's internal procedures, workflow and security policy before being deployed in an operational context. Nevertheless, the concepts proposed by the workpackage remain valid independently of the chosen implementation.

Section 2 describes the different platform components and section 3 illustrates the results of the platform demonstration. Section 4 provides a summary and conclusion.

2. Platform Description

2.1. Change Detection and Analysis

Two software tools have been developed to assist the image analyst in change detection based on optical imagery: the stand-alone software *ChangeView* and a change detection plug-in for Definiens Developer. *ChangeView* automatically detects and visualizes changes in multi-spectral optical satellite imagery on the image pixel level, whereas the Definiens Developer Plug-in enables to perform object-based change detection.

Both tools are based on the Multivariate Alteration Detection (MAD) method [5, 6], which is a classical statistical transformation referred to as canonical correlation analysis to enhance the change information in the difference images. MAD was proven to be a very effective change detection method for optical images [7, 8].

The procedure is briefly described as follows: If the *N* multispectral bands of a scene acquired at time 1

and time 2 are represented by random vectors X and Y, which are assumed to be multivariate normally distributed, the difference D between the two images is calculated by $D = a^T X - b^T Y$. Analogously to the principal component transformation, the vectors a and b are sought subject to the condition that the variance of D is maximized and subject to the constraints that $var(a^T X) = var(b^T Y) = 1$. Determining the vectors a and b in this way is a standard statistical procedure which considers a generalized eigenvalue problem. For a given number of features N, the procedure returns N eigenvalues, N pairs of eigenvectors and N orthogonal (uncorrelated) difference images, referred to as to the MAD components.

The MAD components represent different categories of changes. Relevant changes of man-made structures will generally be uncorrelated with seasonal vegetation changes or statistic image noise, and thus be represented in different MAD components. Furthermore, the calculations of the MAD components are invariant under affine transformation of the original image data. As changes in the overall atmospheric conditions or in sensor calibrations are approximately equivalent to affine transformations of the pixel intensities, the method is insensitive to both of these effects, and thus requires no radiometric pre-processing of the original image data.

Pixel-Based Change Detection

ChangeView calls for two co-registered multi-spectral images as input without the need for any further user-interaction or parameter tuning. The MAD algorithm runs fast even on very large datasets and reliably produces the corresponding change map. The changes are colour coded according to their type, i.e. seasonal vegetation changes are coded differently from changes due to construction activities. Figure 1 displays the percentage of detected and real change depending on a user-selected threshold. By increasing the threshold (here based on the chi-square values), the detection rate (percentage of detected among all real changes) will decrease, whereas the percentage of real among detected changes will increase.

Some of the changes (e.g. seasonal vegetation changes) might, however, not be relevant to non proliferation. Therefore, a visualization tool supports the analyst in assessing the resulting change map: it provides an overview of the colour-coded change map thus highlighting potential areas of interest. When the user clicks on a particular point in the change map, the tool displays the original imagery of the corresponding area (at both instants of times) and the resulting change map in full resolution (see Figure 9). Thus, the change detection tool provides an additional layer of information, which supports the analyst in the visual interpretation of the satellite image and help to efficiently identify the changes which are of non proliferation relevance.

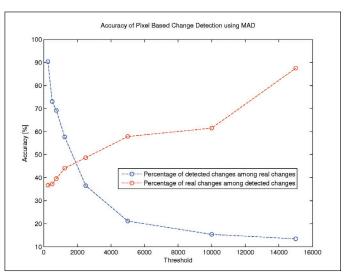


Figure 1: Accuracy assessment on the class "Changes".

Object-based change detection

Object-based image analysis is in a first approximation comparable to visual perception. An image interpreter recognizes, along with the colour of an image, also the shapes, textures and coherent regions present within it, and associates meaningful objects and their contextual relations. A similar goal is intended in object-based image analysis, although the complexity and effectiveness of human perception is of course far from being achieved. Object-based image analysis starts with the extraction of the image objects through segmentation, where the segments should ideally represent the real world objects.

Analysing satellite image data in an object-based way generally extends the possibilities to detect changes between two or more dates. In addition to the change pixel measures listed before, objectbased change detection techniques can also estimate the changes of the mean object, such as shape and size, assess the modified relations among neighbouring, sub- and super-objects and identify changes regarding the object class memberships. In this respect, the MAD transformation can also be applied in the image object domain, for instance by using the object features.

MAD transformation enhances different types of changes within the object level rather than classifying them. However, with a huge number of input features and thus output MAD components, visualization of relevant changes becomes difficult. Therefore, a clustering procedure based on the Fuzzy Maximum Likelihood Estimation (FMLE) was implemented in order to group the change types. The FMLE technique has the advantage of forming elongated clusters and clusters of widely varying memberships and was found to be suitable for classifying the change objects. Both MAD transformation and FMLE clustering were programmed and implemented as Definiens Developer plug-in using the Definiens Software Development Kit (SDK) [9].

A case study was carried out using the bitemporal Quickbird dataset acquired over Olkiluoto in June 2005 and July 2006, where surface changes are due to the construction of a new nuclear facility. The two images were pan-sharpened and co-registered. Object extraction was performed through Definiens Multiresolution Segmentation using all eight pan-sharpened image bands of the bitemporal data set. Changes between the two acquisition times were then calculated by a MAD transformation using 16 colour and Haralick texture features (based on the grey level co-occurrence matrix (GLCM)): mean colour value, the mean colour difference to neighbours, GLCM homogeneity, GLCM mean, each for the four given spectral bands.

Figure 2 shows the 16 MAD components as output from the change detection. Mid grey colour indicates no-change areas, whereas changes are represented by brighter and darker grey values up to white and black. For visualizing the relevant changes, two different approaches were applied: Display of changes detected by some thresholded MAD components (Figure 10) and FMLE clustering of all MADs (without figure). Among the 16 MAD components, MAD 12, 15 and 16 contain much less noise than the other components and were therefore selected for the display in Figure 10, differentiated into positive and negative values. MAD 12 indicates changes from bare soil to buildings, whereas MAD 16 shows changes from vegetation to bare soil. The clustering results for six classes are ambiguous: Except for the one class representing no change, no other thematic classes can be defined. Improvements are therefore needed for the post-processing.

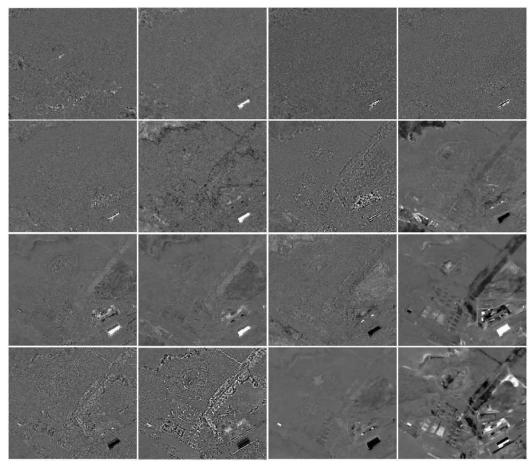


Figure 2: Results of the object-based change detection for Olkiluoto between June 2005 and July 2006 based on 16 object features: mean colour value, the mean colour difference to neighbours, GLCM homogeneity, GLCM mean, each for the four given spectral bands. The Figure displays MAD component 1 (upper left) to 16 (lower right).

Object-based Image Classification

The platform also incorporates object-based image classification to further analyse the change map and automatically identify non proliferation relevant changes. The classification is based on a rule set, which contains both geometric and radiometric rules for automatically identifying objects in the image [10]. The rule set is formulated by an experienced user; it is generally transferable from one image of a site to another (provided that the images are radiometrically normalized), however, it is usually not transferable to a different site. The speed for creating the rule set depends on the quality of the data and the experience of the user.

Figure 11 shows as an example the object-based classification of a Qickbird scene acquired over Olkiuoto in September 2007. The classes Road, Bright Buildings, Dark Buildings, Water, Shadow, vegetation and Bare Soil were extracted with an overall accuracy of 79% (95% significance level, based on 200 manually classified pixels).

Object-based analysis of satellite imagery is still in an early development phase and further research is required to make it more suitable for an operational context. However, if the reliability and user friendliness can be further improved, it has the potential to significantly improve the efficiency of the image analyst. Future work will focus on improved segmentation to extract man-made structures more accurately and to define the classification model in a more user-friendly, semi-automated manner.

2.2. 3D Information Extraction

Digital Surface Maps (DSMs) extracted from VHR stereo satellite imagery have a resolution and ac-

curacy considerably better than standard Digital Elevation Models (DEMs), e.g SRTM or Aster GDEM. They can be used for several purposes, e.g. i) for an improved geometric correction of the VHR satellite imagery during the image pre-processing phase, ii) for the creation of measurable 3D models and iii) for 3D-based change detection. The focus in Treaty Monitoring workpackage was on 3D change detection as described below.

Stereometric Processing Workflow

Standard techniques, which are normally used for the creation of DEMs from lower resolution imagery, often produce unsatisfactory results for the non proliferation application [11]. For example, manmade structures with sharp contours (such as nuclear facilities) are blurred in the resulting DSM. Also, depth continuities and occlusions as they appear in VHR imagery generate mis-matches thus producing erroneous results. The standard methods have been improved within the LIMES project and implemented in the RSG and Impact software package provided by Joanneum Research [12]. Figure 3 shows two versions of a DSM generated from a stereo pair over Olkiluoto. The left DSM results from applying a standard method; the right DSM is generated using an advanced algorithm. The red circles highlight the improved building contours: in the left image the building (one of the existing nuclear reactors) appears blurred; in the right image the contours are more accurate. The blue circles highlight an example of improved image matching: the left image contains errors induced by the depth discontinuities whereas the right image shows the correct results.



Figure 3: Digital Surface Maps (DSM) of the Olkiluoto site generated with the Joanneum RSG/Impact software from an Ikonos stereo pair (2008).

The RSG software implements the following stereometric processing workflow:

- Sensor modelling: The mathematical model of the sensor allows mapping a point from ground into the image and vice versa. Based on ground control measurements and/or tie-point measurements, sensor parameters can be optimized in a least squares manner.
- **Image matching:** The core procedure of the stereometric processing is the measurement of corresponding points in the stereo image pair, which is done by automated image correlation. The basic output is the geometric differences between the corresponding points which are known as *parallaxes* or also *disparity vectors*.
- **Point intersection:** the point intersection converts the 2D coordinates of the matching pixels into the 3D Cartesian coordinates of the corresponding ground point.
- Interpolation: The input to the final DSM generation procedure is given in ground coordinates being stored in raster file format (output of previous intersection). These ground coordinates represent irregularly distributed locations on the ground, for which the terrain height is known. Respective interpolation techniques are either a linear interpolation based on nearest neighbour re-sampling or an areal interpolation approach.

An estimate of the relative accuracy for the resulting DSM can be obtained by fitting a plane to a planar surface and analysing the fitting error. An analysis was made of the DSMs obtained from the two Olkiluoto stereo pairs acquired with the Ikonos and GeoEye-1 sensors, in 2008 and 2009 respectively. Three planar areas (parking lots) were selected in each of the DSMs and a plane was fitted to each of them. The resulting fitting errors (3 Sigma) are reported in Table 1. It illustrates that the values of the Gaussian noise are well within a range that makes the data suitable for non proliferation purposes. Other error types, for example originating from depth discontinuities and matching problems, are currently under investigations (see also Figure 13).

	Ikonos May 2009	GeoEye-1, May 2009
Area 1	171 cm	99 cm
Area 2	174 cm	81 cm
Area 3	150 cm	87 cm
Average	165 cm	89 cm

Table 1: Error values (3 Sigma) for fitting a plane to three parking lots in the DSMs obtained from an Ikonos and GeoEye-1 stereo pair, respectively.

3D Change Detection and Interpretation

The accuracy obtained from the 2008 and 2009 Olkiluoto stereo pairs allows using them for 3D change detection, which is done by calculating the height difference of the resulting DSMs [13]. It has the advantage that it detects only changes in the geometry of the site which is typically induced through construction activities. Irrelevant changes, e.g. due to seasonal changes are ignored. Besides the simple height difference also information about the change of the land cover is used. Here, the wellknown normalized difference vegetation index (NDVI) is calculated to separate vegetation from non-vegetation areas. Depending on a threshold for significant height changes different change classes can be identified as shown in Table 2.

2.3. SAR Processing

With the availability of the latest VHR sensors, SAR (Synthetic Aperture Radar) imagery has gained significant importance for non proliferation applications. The principle advantage of SAR imagery is its all-weather, all-time acquisition capability. Additionally, the potential to compute accurate DSMs and detect small ground movements are of interest. Future sensors, e.g. the TanDEM-X satellite scheduled

Case	Epoch 1	Epoch 2	$ \Delta H $	ΔH	Comment	Colour
1	Vegetation	Vegetation	< threshold		No change	
2	Non-Vegetation	Non-Vegetation	< threshold		No change	
3	Vegetation	Non-Vegetation	< threshold		New road, path, etc.	
4	Vegetation	Vegetation	> threshold	< 0	Clear-cut	
5	Vegetation	Non-Vegetation	> threshold	< 0	Clear-cut	
6	Non-Vegetation	Non-Vegetation	> threshold	> 0	New building	
7	Non-Vegetation	Non-Vegetation	> threshold	< 0	demolition	

Table 2: Classes of different changes detected in multi-temporal 3D information.

to be launched in 2010, will further increase the capabilities. However, SAR imagery does not come without its limitations. Some of the drawbacks with respect to optical imagery are listed hereafter:

- SAR imagery contains a high amount of **speckle noise**, i.e. neighbouring pixels of the same feature (e.g. agricultural field) will have different backscatter values. This makes both, visual interpretation and automated (pixel-based) analysis difficult.
- Due to the **acquisition geometry**, the perception of a SAR image is different compared to an optical system. Layover, foreshortening or stretching of the back slopes make the image difficult to interpret especially for man-made structure, e.g. buildings are often not rectangles but only one bright stripe or bright spots.
- SAR backscatter depends on scene acquisition parameters like look direction (ascending vs. descending), look angle and on physical parameters of the mapped surface such as soil moisture. Therefore, multi-temporal SAR analysis imposes tight constraints on these parameters.

The above issues render visual interpretation of SAR imagery less intuitive than optical imagery and usually an experienced analyst is required for the task.

Hence, the Treaty Monitoring platform includes an advanced, automated processing workflow, which addresses some of the problems and directly generates an anomaly map from a multi-temporal SAR series. It uses the CIAO software package, developed by CEA, for detecting anomalies by the means of analysing series of interferograms taken at different instants in time [14]. The analyst first collects

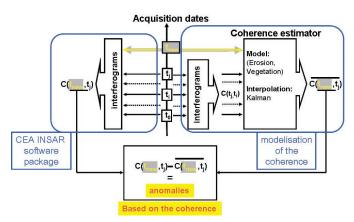


Figure 4: Principle of anomaly detection from coherence image.

the radar images for a given area (different radar satellites eventually) and then uses the software to generate the coherence images. Dark areas in the coherence image indicate changes between two images and the bright areas indicate possible infrastructure which did not change between the two image acquisitions. With this anomaly detection completed, the analyst can investigate suspicious areas further using another coherence image or optical images providing greater resolution. Figure 4 shows a schematic view of the anomaly detection and Figure 5 gives an example of the process applied to an industrial port area.

Because a coherence image is computed from two radar images acquired at different instants in time, it highlights changes (black areas) between those two acquisition dates. Two coherence images, one corresponding to changes between dates (t1-t2) and the other to dates between (t1-t3), can be used to generate a false-color composite highlighting

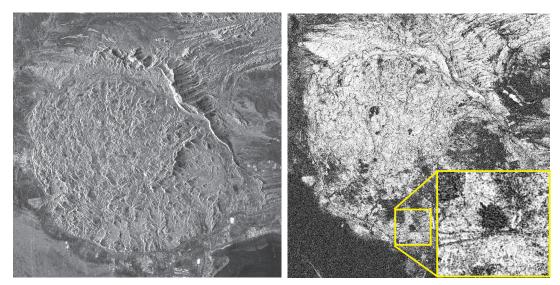


Figure 5: The left image shows a SAR image of an industrial port area. The right image shows the coherence map of the same area obtained from two SAR images. The yellow square highlights a detected anomaly.

changes due to the activities between dates (t2-t3). An example is given in Figure 6.

CIAO includes the possibility to blend radar and optical images, thus facilitating the interpretation of the radar image and the validation of anomalies highlighted in the coherence image. CEA is continuously improving CIAO with the objective to further automize the anomaly detection. The two main modules currently under development relate to Permanent Scatterers and atmospheric compensation. Numerous Permanent Scatterers appear in VHR X band images, especially in areas containing manmade infrastructures. Exploiting these features in the algorithm will further improve the suitability of CIAO for site monitoring applications. Atmospheric effects, on the other hand, interfere with the radar signal. Taking them into account computationally will improve the contrast of the resulting coherence images and therefore facilitate their interpretation.

2.4. Information Management and Integration

A core objective of the Treaty Monitoring workpackage is to provide an integrated platform to the non proliferation image analyst. In practice this means, that the analyst should have a central point of access, which allows to:

- retrieve, view and analyse all available (spatial and non-spatial) information for a given site, including satellite imagery, GIS information, external databases and collateral information
- access dedicated analysis software performing specialist tasks, such as change analysis, SAR and 3D processing tools as described above. Any results obtained from the tools (e.g. a resulting change maps) should be stored for later retrieval in the integrated platform.

The integration platform developed under LIMES is based on a standard three-tier architecture (data-

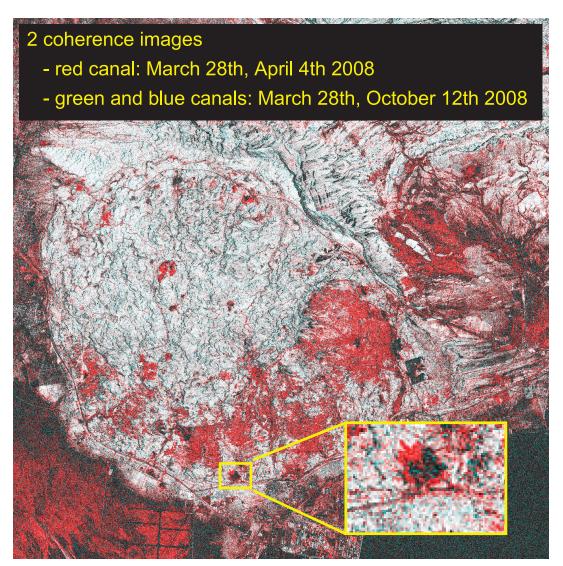


Figure 6: The image shows a false-colour composite of two coherence images. The yellow square shows an anomaly: the red part corresponds to a change that occurred between April 4th and October 12th 2008.

base, application server and web client) using common industry standards as illustrated in Figure 7. In order to support information of different types (both spatial and non-spatial), the platform incorporates three independent pillars each serving a particular purpose: a *geographic information system*, a *Wiki system* and a *document repository*. Each of the pillars is briefly described in the following paragraphs.

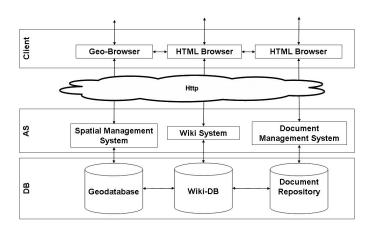


Figure 7: High-level architecture of the Treaty Monitoring integration platform. The system integrates a Geographic Information System (left), a Wiki (centre) and a document repository in order to support spatial and non-spatial information.

Geographic Information System

The Geographic Information System (GIS) provides an intuitive map-based interface to the user. It allows storing, retrieving and visualising spatial information. The focus is on user-friendliness and scalability, e.g. large (gigabyte) images are served as image pyramids for easy navigation. Each feature in the geo-database is context-sensitive, i.e. it can be selected from the user interface and cross-linked with other information, such as meta-information, collateral data and analysis results. Spatial information is stored in a central database and served via a web-application to the geo-browser on the client PC. It is designed to store different types of (preprocessed) satellite imagery, the results of manual or semi-automated interpretation and GIS-layers of general interest, which is then made available to the user for an all-source analysis.

Due to its wide-spread use and user-friendly interface, Google Earth was used as a client geo-browser during the platform demonstration. However, other geo-browsers might be adapted for the purpose, if this is required for security or other operational reasons.

Wiki System

The objective of the Wiki system is to capture unstructured, tacit information available in an organisation. For example, each feature in the geodatabase (e.g. a particular facility) can have a corresponding Wiki page containing relevant information or previous analysis. But the Wiki goes further and might contain supporting information, for example pages regarding relevant technologies, organisations, treaties, regulations, etc. Wikis are most known as Internet applications (e.g. Wikipedia), where the huge number of contributing users ensure reliable and exhaustive content. However, the potential of Wikis is also increasingly recognised in Intranet environments with a smaller number of users, e.g. in corporate Intranets or within intelligence communities [15, 16]. A prominent example is Intellipedia, which is an online system for collaborative data sharing used by the United States intelligence community [17, 18].

The Wiki system used for the Treaty Monitoring platform provides functionalities, which allow defining different classes of pages and for each class i) a set of relevant attributes and ii) forms for data input and visualisation. Thus, it combines the strengths of a Wiki (collaboration, ease-of-use and support for unstructured information) with those of a classical database application (querying and filtering based on attributes; consistency in information storing and visualisation). At this point, an important (and often difficult) task is the definition of an appropriate data model, i.e. of classes and attributes which describe the information relevant to an organisation. For the platform demonstration, the workpackage developed a data model which captures the information relevant to the image analyst, i.e. it describes countries, sites, nuclear facilities, satellite images, events, treaties and other relevant background information.

Document Repository

The document repository is designed as a central archive for all relevant documents collected from various sources. In particular, Open Source information is becoming increasingly important to trigger, guide and support imagery-based analysis. Many analysts collect thousands of documents from the Internet covering different types of information. Also restricted documents containing sensitive information might be required during the analysis work. Typically, each analyst has his personal approach to document storage and classification, thus making it difficult to re-use documents or share documents of common interest between a group of analysts. In order to overcome these limitations, the workpackage proposes the use of an off-the-shelf Content Management System (CMS), which is adapted to the needs of the project. A CMS provides i) a central repository for all documents, ii) advanced functionalities for data upload, search and retrieval, iii) access control and iv) the possibility to define a data model, i.e. sets of attributes (metadata) for different classes of documents. Using a common data model for the CMS and Wiki facilitates the information integration since the same query can be used to search the related information in both pillars.

The integration platform does not impose additional workload on the analyst for ingesting and maintaining information. Rather, it constitutes a centralised and standardised method to manage i) the information that is collected and validated during the routine work and ii) the knowledge generated during the analysis process. Therefore, the organisation's knowledge base will grow in time as a result of its core activity without creating a large overhead on human resources.

The implementation details of the integration platform depend on the constraints given by a specific organisation, e.g.:

- *IT infrastructure:* the platform needs to be compatible with the existing IT infrastructure and reuse existing IT skills and technologies.
- *IT policy:* the implementation needs to comply with the organisation's IT policy, which might favour commercial off-the-shelf products, Open Source implementations or proprietary developments.
- Procedures, workflow and corporate culture: each organisation has existing procedures and workflows for information collection, analysis and management. The concepts proposed by LIMES might imply a change of procedures in some areas, e.g. with respect to information sharing and storage. In other areas, the workflow is imposed by external constraints - such as the need for stringent information validation and review - and therefore needs to be reflected in the integration platform.
- Information Security is of paramount importance in the non proliferation context. However, each organisation has its own security and access policy. An operational integration platform needs to follow the existing rules and implementations precisely.
- Size, mandate and budget of an organisation: A small organisation with a single group of analysts

has different needs regarding information integration than a large organisation with different groups of analyst and inspectors. Likewise, the size of the organisation will determine the budget available for implementing, maintaining and customising the integration platform.

For those reasons, the workpackage did not intent to implement a finished product as a solution for a specific organisation. Instead, the objective was to devise a high-level architecture and implement a prototype that addresses the above issues and at the same time allows demonstrating and evaluating the implementation-independent concepts for an integrated information analysis.

3. Platform Demonstration

Two platform demonstrations were carried out in July 2008 and October 2009. The objective was to present the developments to interested stakeholders and obtain feedback from potential users. For the purpose of the demonstration, a typical scenario was defined consisting of a initial base-line analysis and the continuous monitoring of a nuclear site. The Finish NPP Olkiluoto was selected as test site for the demonstration scenario.

The reason for selecting Olkiluoto was that i) it hosts two operational nuclear reactors with a third reactor currently under construction, ii) it was possible to obtain ground-truth information and validate the outputs of the analysis and iii) the site authorities (STUK) and operator (TVO) were very collaborative in supporting LIMES activities. Archived satellite imagery reaching back to 2002 as well as new imagery between 2007 and 2009 was purchased, thus simulating a continuous monitoring of the site over seven years. The data used for the demonstration include

- VHR optical satellite imagery (2002, 2005, 2006 and 2007)
- VHR optical stereo imagery (2007, 2008, 2009)
- SAR imagery (image series from May to September 2009)
- Open Source information including documents, maps, images and videos.
- Ground information, such as GPS data

Demonstration Results

Figure 8 shows snapshots of four multi-spectral VHR images acquired over Olkiluoto. They include the annotations resulting from the standard, visual interpretation carried out by an image analyst.

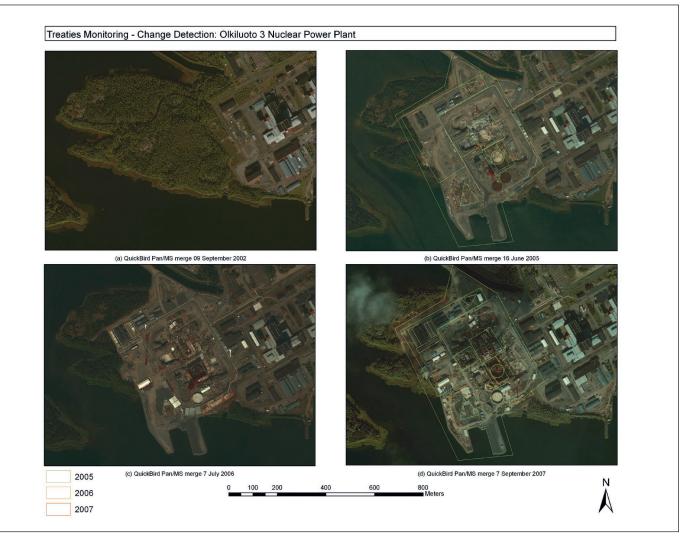


Figure 8: The four images show the site of the EPR reactor before the construction (2002, top-left) and at different instances during the construction (2005, 2006 and 2007).

Figure 9 shows a screenshot of the TUBAF change visualization tool. The left window displays an overview of the original Olkiluoto images taken in 2005 (upper left) and 2006 (upper right) as well as the resulting change map as colour coded MAD components (lower left) and absolute change intensity (lower right). The right window displays the same information in full resolution.

Figure 10 and Figure 11 show results of the objectbased change analysis and image classification, respectively. Figure 12 illustrates the results of the 3D change detection.

Figure 13 is a snapshot of the JRC 3D Reconstructor showing 3D information derived from the 2009 Geo-Eye-1 stereo pair. Changes relative to the 2008 Ikonos stereo pair above a selected threshold of 5m are highlighted with a given colour code. The main construction works at the Olkiluoto 3 reactor clearly show up (bottom left). Additionally, a number of other new buildings distributed over the site are easily detected, as well as a deforested area at the top-right of the image. The 3D change detection not only highlights the changed areas, but also provides a quantitative measure through the colour-coded height difference. There are only few false positive, which mainly relate to forest areas and artifacts stemming from depth discontinuities, e.g. at the existing reactors Olkiluto 1 and 2. The image illustrates that the 3D change detection provides a robust result: with the selected threshold, all changes above 5m are reliably detected. When decreasing the threshold, also smaller changes will be identified; however, the noise level will increase. Further improved accuracy is expected with new sensors and advanced processing workflows.

Figure 14 shows different types of information integrated in a single environment: the client application is visualising the spatial and non-spatial information served by the integration platform. The spatial information (multi-temporal vector or raster information) is selected from the hierachical tree on the left. For each feature, related non-spatial information can be loaded into the html browser on the right.

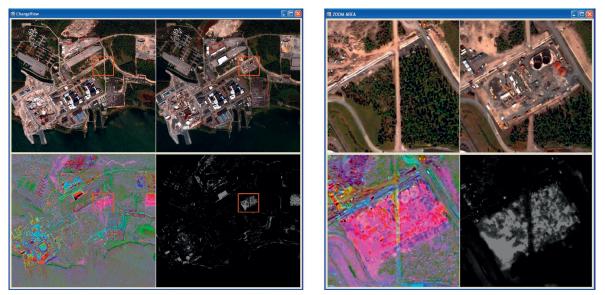


Figure 9: Snapshot of the TUBAF change visualisation tool.



Figure 10: Results of the object-based change detection for Olkiluoto between June 2005 (left) and July 2006 (right) based on 16 objects, given by the MADs 12, 13 and 16 (right).

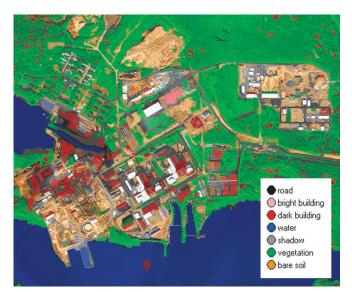
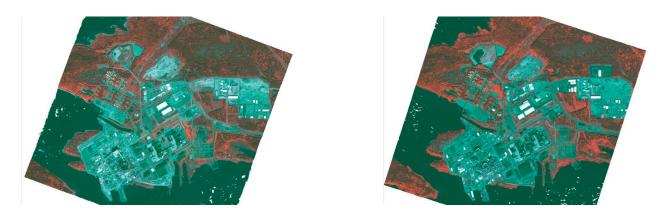


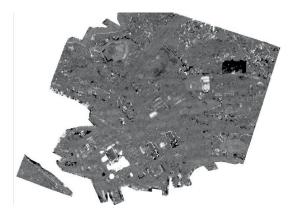
Figure 11: Results of the object-based classification of the Quickbird scene acquired over Olkiluoto in September 2007.



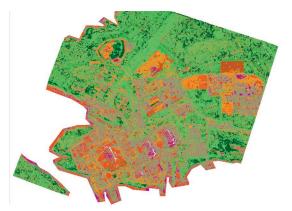
Digital Surface Map (DSM) of the Olkiluoto site from an Ikonos stereo pair acquired in 2008 (left) and a Geo-Eye-1 stereo pair acquired in 2009 (right)



Rectified false colour composite of the Olkiluoto site (R: Near infrared, G: Red, B:Green) of the left lkonos stereo partner acquired in 2008 (left) and the left GeoEye-1 stereo partner acquired in 2009 (right)



Difference height model of the Olkiluoto site (Geo-Eye-1 minus Ikonos). White indicates a positive height change, black a negative one



Classified height differences as a first output of the 3D change detection. The colour coding is described in Table 2.

Figure 12: Input and output of the 3D change detection of the Olkiluoto site generated with the Joanneum RSG/Impact software.

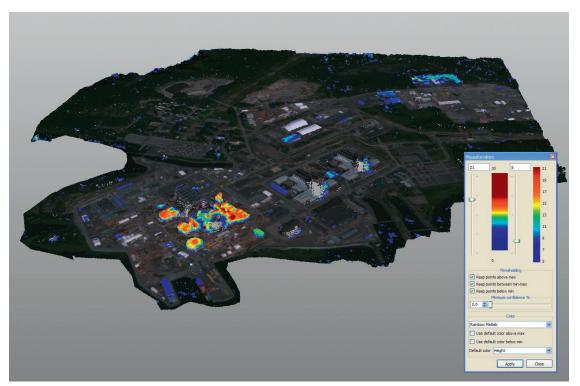


Figure 13: Snapshot of the JRC 3D Reconstructor showing 3D Information extracted from the 2009 Geo-Eye-1 stereo image. All changes with respect to the 2008 stereo image that are above 5m are colour coded as shown in the dialog box.

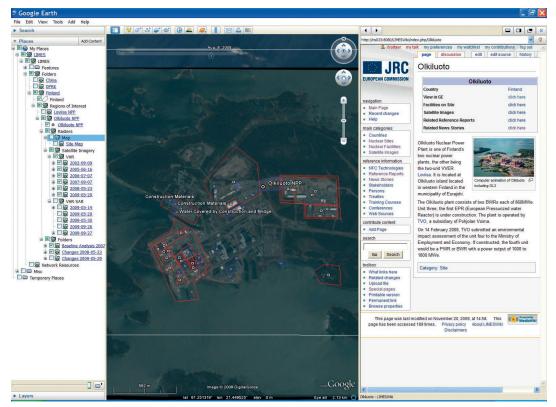


Figure 14: Snapshot of a geo-browser (Google Earth in this case).

Demonstration Feedback

For the service demonstration, the platform was deployed at the European Satellite Centre and evaluated using the Olkiluoto monitoring scenario. The results of the testing were presented to and discussed with other stakeholders in the nuclear Nuclear Safeguards community including IAEA and DG-TREN. Table 3 summarizes the evaluation of the platform and its components from a user point of view.

Pixel-based change deter	ction using optical VHR imagery
Added value	The tool highlights changes in bi-temporal optical image pairs. An interactive, user-friendly interface allows the user to identify relevant changes more quickly and consistently. The change map provides an additional layer of information, which supports the image analyst in the visual interpretation of the image.
Limitations	The automated process does not distinguish between functional and irrelevant (e.g. seasonal) changes.
Development status	A prototype is available and ready to use.
Operational impact	The processing overhead is very small: the input imagery is the same as used for the visual interpretation (an orthorectified and co-registered image pair). The change detection itself runs within a few minutes.
Object-based change and	alysis using optical VHR imagery
Added value	It detects and analyses changes in multi-temporal optical images. Changes are classified according to their functional relevance, e.g. new construction of buildings, roads, etc. Irrelevant changes are suppressed.
Limitations	The classification is based on a rule-set, which needs to be adapted for each site. Also radio- metric variances between multi-temporal images of one site might cause false alarms.
Development status	A prototype has been developed as plug-in to the Definiens Developer suite. Object-based satellite image analysis is in an early stage of development and further research is required to improve the workflow and the reliability of the resulting change map.
Operational impact	The tool can increase the effectiveness of the analyst in finding all functional changes. How- ever, it requires expertise and time to set-up the rule set needed for each site.
3D information extraction	I from stereo VHR imagery
Added value	VHR 3D information can be used for several purposes: to improve the geometric correction of raw satellite images, to measure distances, heights and volumes, for visualisations and for 3D change detection.
Limitations	In order to be used for monitoring nuclear facilities, the DSM requires an accuracy which can only be achieved from the highest resolution satellites currently available, e.g. Quickbird, Geo-Eye-1 or Worldview. The process is sensitive to image artefacts such shadows and satura-tion.
Development status	A functional software package is available for the extraction of 3D information. Developments continue in order to improve the quality of the DSM as well as the processing workflow.
Operational impact	A VHR DSM provides valuable input for site analysis and monitoring, in particular for complex sites, which are regularly monitored. However, the generation of a DSM requires time and expertise. Stereo imagery of the required quality can have considerable cost implications.
3D change detection	
Added value	3D change detection is insensitive to seasonal changes and radiometric influences and there- fore draws the attention of the non proliferation analyst directly to the relevant changes such as new constructions.
Limitations	The DSM accuracy that can be achieved with today's satellites is on the limit of what is re- quired for site monitoring. Sufficient noise level and detection rate can only be achieved from few stereo sensors.
Development status	A software package is available. Developments are on-going in order to improve the accuracy and reliability of the resulting change map
Operational impact	3D change detection can provide additional input and increase the effectiveness of site moni- toring, in particular for complex sites that are being monitored regularly. However, it requires multi-temporal VHR DSMs, which can have a considerable impact on costs and resources as input.

Automated anomaly dete	ction using multi-temporal SAR data
Added value	The general value of SAR imagery is its all-weather, all-time acquisition capability. With the recent VHR sensors, SAR became even more important for non proliferation purposes. The software proposed by the Treaty Monitoring workpackage provides multi-temporal anomaly detection reducing the complex visual interpretation that is usually related to SAR analysis.
Limitations	The artefacts of a SAR image such as speckle noise, complex SAR geometry and backscatter make the visual interpretation of SAR imagery more difficult than the interpretation of optical VHR imagery. The anomaly detection demonstrated in the project addresses some of these issues, an experienced user to interpret the result anomaly image.
Development status	The SAR analysis tools are implemented in the CIAO software package.
Operational impact	CIAO supports the analyst in identifying anomalies in both medium resolution and VHR SAR imagery and can therefore enhance the effectiveness of site monitoring and wide-area screening. The processing overhead is small.
Information Integration	
Added value	The focus of the platform is to answer to the perceived need for integrating information from multiple sources, multiple time-frames and resolutions, for an efficient and thorough non pro- liferation analysis.
Limitations	The integration platform is closely linked to the existing information infrastructure and policy of a given organisation and therefore any solution needs to be adapted to the specific needs.
Development status	The integration platform is a fully functional prototype implementation, mainly based on avail- able off-the-shelf products. To be used in an operational context, it typically would need to be customised to an organization's internal procedures, workflow and security policy.
Operational impact	The integration platform proposes novel and standardised methods to manage and store the information within an organisation, thus increasing the efficiency and effectiveness of the single analyst and the organisation as a whole. Some change in procedures and the related training are required.

Table 3: The table summarises evaluation of the different platform components.

4. Summary and Conclusion

Satellite Imagery plays an important role in nuclear non proliferation. One of the major challenges faced by the image analyst is the monitoring of nuclear sites with respect to new constructions and their operational status, i.e. the detection and analysis of relevant functional changes in multi-temporal images. Currently, operational work relies to a large extend on optical VHR imagery, which is visually interpreted using standard analytical tools. VHR SAR imagery is gaining importance due to its all-weather, all-time capabilities. However, the analysis of this complex imagery is often a challenging task.

The image analyst is faced with an increasing number of customer requests on one hand and increasing quality and quantity of available input data on the other. The Treaty Monitoring workpackage proposes a number of tools addressing these challenges. The focus of the platform is to answer to the perceived need for integrating information from multiple sources, multiple time-frames and resolutions, for an efficient and thorough non proliferation analysis. The integration platform is designed as a collaborative environment; therefore it not only integrates information of different types and sources, but also allows sharing information between different analysts and groups within the organization. Furthermore, it supports knowledge preservation, which is often an important issue in environments with high staff turn-over. The implementation details of the integration platform depend on constraints given by a specific organisation, e.g. existing IT infrastructure, workflows, security policies and corporate culture. Therefore, the workpackage did not intent to implement a finished product as a solution for a specific organisation. Instead, the objective was to devise a high-level architecture and implement a prototype that addresses these issues and at the same time allows demonstrating and evaluating the implementation-independent concepts for an integrated information analysis.

This information integration concept is supported by tools adapted to the types of information to be integrated. The tools process raw satellite imagery to extract higher-level information (i.e. change maps, anomalies and 3D information), contributing to the analyst's efficiency and enhancing the consistency of the resulting product.

The platform was demonstrated in two user workshops to a number of interested stakeholders including DG-TREN and IAEA. The general feedback was that the project addressed important issues for the non proliferation image analyst, regarding both supporting analysis tools and information integration. Clearly, none of the components can replace the analyst in interpreting the images from a non proliferation point-of-view. However, they put a range of additional tools at his disposal, which can facilitate the analysis by highlighting changes potentially of interest and providing quantitative measurements which are not readily available from the images.

Some of the components are already in a development phase where they might be incorporated into the operational workflow; further user testing will be carried out for that purpose. Other components require additional research and development. The project partners will continue to collaborate to this extend in EU-funded (e.g. G-MOSAIC) and other projects.

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Working Groups activities

Report on the Workshop on Measurements of Impurities in Uranium

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2. National Nuclear Laboratory, UK

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4. European Commission-Joint Research Centre, Institute for Reference Materials and Measurements, Geel, Belgium

1. Introduction

The ESARDA Working Group on Standards and Techniques for Destructive Analysis (WG DA) organised a Workshop on 16 and 17 March 2009 at the Joint Research Centre Institute for Transuranium Elements (ITU) on MEASUREMENTS OF IM-PURITIES IN URANIUM SAMPLES in order to exchange views and information on the needs, the applicable measurement techniques, the required measurement quality results and statistical evaluation techniques. The workshop attracted some 30 specialists in impurity measurements in support of safeguards, non proliferation and illicit trafficking from Europe, the US, Australia and Asia. Following a series of presentations, the participants formed two working groups, to debate future requirements and make recommendations for further work.

The workshop was hosted by the Institute for Transuranium Elements (ITU) in Karlsruhe. The director of ITU, Prof. Th. Fanghänel, opened the meeting and welcomed the participants, underlining the importance of impurity measurements in different areas and highlighting the prominent role the ESARDA working group on destructive analysis (WG DA) is taking in this specialized area. The chairman of the WG DA, Klaus Mayer, outlined the workshop objectives and introduced the working methodology.

The institutions participating to this workshop are listed in Table 1.

2. Objectives of the workshop

The workshop objectives as were recalled to the participants at the beginning of the meeting:

1. Formulate **needs and requirements** related to chemical impurities in uranium of Safeguards Authorities, fuel manufacturers and nuclear forensics laboratories.

- 2. Identify **measurement challenges** that should be addressed. Such challenges include, but are not limited to: quantification, limit of detection, calibration, standards and reference material, quality control, sample preparation.
- Identify evaluation challenges such as statistical tools, reference data, comparison samples, data interpretation and attribution of parameters to source material or to process types.

Moreover, the workshop aimed at formulating observations reflecting the current status of measurement capabilities and use of impurity patterns for evaluation purposes in different area. Finally, the workshop served for expressing recommendations for further research work, for the identification of characteristic parameters, for evaluating the need for establishing performance goals and for considering the benefits of statistical data treatment techniques.

3. Workshop structure

Two invited papers opened the technical part of the workshop and set the scenery. In the subsequently presented technical papers, experience gained on various laboratories was exchanged. During the discussions that followed the individual presentations a number of questions were phrased that were addressed in more detail during the breakout sessions. The main discussions were then held in two working groups (breakout session), where the observations and conclusions were formulated. Working Group 1 addressed "Measurement Quality, Measurement Techniques and Protocols" and was chaired by S. Boulyga (IAEA). The second Working Group was chaired by J. Tushingham (NNL) and focused on "Data Evaluation and Identification of Characteristic Patterns or Parameters".

Institution	Country
Australian Nuclear Science and Technology Organization	Australia
University of Natural Resources and Applied Life Sciences BOKU-VIENNA	Austria
Joint Institute for Power and Nuclear Research	Belarus
Belgian Nuclear Research Centre SCK·CEN	Belgium
Directorate General for Transport and Energy, EC DG TREN	European Commission
Joint Research Centre-Institute for Institute for Transuranium Elements - EC-JRC-ITU	European Commission
Joint Research Centre-Institute for Reference Materials and Measurements - EC-JRC-IRMM	European Commission
Commissariat à l'Énergie Atomique - CEA Marcoule	France
AREVA NP GmbH	Germany
Institute of Isotopes, Hungarian Academy of Science	Hungary
Japan Atomic Energy Agency	Japan
Semenov Institute of Chemical Physics, Russian Academy of Sciences	Russian Federation
Korea Atomic Energy Research Institute - KAERI	South Korea
Paul Scherrer Institut - PSI	Switzerland
URENCO	The Netherlands
AWE Aldermaston	United Kingdom
Sellafield Ltd.	United Kingdom
National Nuclear Laboratory	United Kingdom
International Atomic Energy Agency - IAEA	United Nations
International Atomic Energy Agency - Safeguards Analytical Laboratory – IAEA-SAL	United Nations
National Institute of Standards	United States of America
US Department of Energy, Lawrence Livermore National Laboratory - LLNL	United States of America

Table 1: List of participating institutions.

4. Plenary session

In two lectures by invited speakers from international safeguards and nuclear forensics, the current benefits and limitations in using chemical impurities in uranium samples were addressed. In further papers, specific subjects such as measurement problems, quality control or data evaluation were discussed.

Mr Ryzhinskiy (IAEA) provided an overview of the IAEA's needs for impurity analysis, and a more detailed consideration of some of the issues affecting data evaluation. Under the Additional Protocol, the IAEA has responsibility to verify the completeness of a State's declaration. Export and Import controls provide the IAEA with the means to determine the origin of material. A significant increase in uranium deposit processing is expected to require a similar increase in the workload in verification of material origin. Such verification cannot rely on traditional measurements of uranium concentration and enrichment alone, therefore chemical impurities are considered a potentially useful additional source of information. Measurement data, however, are insufficient to draw a safeguards conclusion without:

Reference data; and

• Standardised evaluation procedures for multiple data sets

For uranium, trace element characteristics are considered the most important tool for defining the origin and process undergone by nuclear material. It should be noted, however, that the behaviour of impurities during processes such as conversion is not fully understood. The IAEA has therefore issued a Task Proposal, seeking support in establishing the behaviour of impurities during the conversion process, and this has (so far) been accepted by four Member State Support Programmes.

In establishing the origin of uranium materials there are a number of considerations:

 Sampling – Source uranium ore concentrate (UOC) must be characterised: ideally with analysis of all available lots. Fewer samples are required from intermediate products, because these are used soon after production. The IAEA does not propose to sample scrap but, for other materials, samples should be taken from the bulk material, at different depths: not just the surface layer. The main problem is sample inhomogeneity, which may follow through the conversion process. Therefore a minimum sample size of some 20g is recommended, to be contained in hard polyethylene bottles.

- Analysis ICP-MS is the most promising technique, but any technique used must be validated with appropriate quality assurance/quality control. Currently, SAL uses two external laboratories, and is hoping soon to add its own in-house capability. The speaker considered that laboratories must have comparable performance criteria, and emphasis was placed on the need for consistency in limits of detection. There was considered to be a major issue over the variation of performance in detection limits and how this could be accounted for during the data evaluation process. It was proposed that procedures for determining limits of detection should be evaluated, establishing appropriate values and performing internal and inter-laboratory exercises.
- Data reporting Associated with the issue of limits of detection was a desire on the part of data analysts to receive "uncensored" data, e.g. negative values, with appropriate comments on closeness to detection limits.
- Data evaluation Groups of samples should first be identified that were statistically different, before comparing an unknown sample with each group. Results from different groups should be compared, to see if they came from the same process. Multivariate statistical analysis was recommended, e.g. cluster analysis or principal component analysis, with a confidence level determined as to whether a particular sample belonged to a group. Comparing different materials, e.g. ammonium diuranate (ADU) and UOC, cannot be done statistically, but will require understanding of the process. Hence the Agency's request for support from the Member States.

Mr Ryzhinskiy concluded that network laboratories involved in impurity measurements should work to similar limits of detection, and that these should be monitored regularly. A standard procedure should be established for data evaluation.

Safeguards and illicit trafficking share many of the same requirements in terms of measurement capability, as M. Wallenius pointed out in her presentation (JRC-ITU). One of the differences, however, is the fact that the laboratory may take on a greater role in comparison and evaluation of data in nuclear forensics application. Often, it proved to be beneficial if laboratory (measurement experts) and authority (evaluators) work closely together in data interpretation. Impurities measured within seized uranium samples were considered to originate from three potential sources:

- The source material;
- Intentional (process) additions, such as burnable neutron poisons (e.g. gadolinium, erbium), or alloying components (e.g. aluminium);
- Accidental additions, resulting from cross contamination.

A major challenge was identifying which impurity came from which source, and how the process would affect impurities. ITU has embarked upon a horizontal study (comparing the same intermediate product of the nuclear fuel cycle for different suppliers around the world), measuring ca. 100 different UOC samples.

Two case studies were presented, the first concerning whether five samples came from the same batch of material. Whilst there were similarities, results showed that there were also differences – possibly resulting from variations in sampling procedure (sample size, container material etc). This emphasised the importance of a knowledge and understanding of how to take appropriate samples, and recommendations were required on how to sample and to maintain a detailed record of the sampling history.

A second case study involved pellets found in a garden in Germany, which were found to be higher purity than those available from some fuel manufacturers. Fuel manufacturers had supplied their specifications for impurities, providing important information in ruling out their facilities as being the origin of the material. The high purity pointed at a production process which involved an additional purification step, thus helping identify the origin of the pellets.

Ms Wallenius concluded that the current horizontal study should be complemented by a vertical study, i.e. investigating the behaviour of impurities during different stages of material processing.

Mr Boulyga (IAEA-SAL) reported on the ICP-MS laboratory at the Safeguards Analytical Laboratory (SAL), which had been completely renovated, with clean sample preparation areas to avoid sample contamination. Microwave dissolution was employed, with an internal standard (rhodium) added to blanks, standards and samples, and standards matrix-matched with uranium (checked for trace impurities).

An Element-2 Sector Field ICP-MS (Thermo Corporation) was utilised, set up in high mass resolution and resulting in low interference from oxide ions.

The disadvantage of this arrangement was lower stability in mass calibration and lower sensitivity. At high mass number, interference was inevitable and had to be corrected for. External calibration, using matrix-matched standards, worked well for many elements at low concentration. Sample dissolution procedures are being tested whilst measurement and quality control procedures are under development. To validate the technique's performance, certified materials close to "real" samples are required. However, such materials are currently not available, suggesting a need for the preparation of standards and interlaboratory measurement and certification.

The role of minor isotopes and transuranics in determining origin was considered, for example ²³⁰Th/²³⁸U which should be <10⁻¹⁰ in freshly-separated uranium. The measurement of such small ratios may suffer from instrumental effects such as peak tailing and detector settling time.

Mr. Ramon (LLNL) compared the performance of a Thermo Elemental ICPMS "out of the box" and following adjustment and fine-tuning in the hands of experienced operators. With respect to matrix effects, the instrument could be optimised for greater sensitivity by tuning on the matrix of the sample. However, this resulted in instrumental effects, like reduced stability of the signal. A combination of matrix tuning and stability optimisation provided a five-fold increase in sensitivity without compromising stability. It was also observed that removal of the uranium matrix was often to be preferred over sample dilution, for measurement of those elements that might otherwise be added during dilution.

Mr Ramon noted a number of other techniques that still played a role in analysis, depending upon the elemental signatures under investigation. For major elements that contributed to the stoichiometry, such as nitrogen in ADU, XRF (X-ray fluorescence) and XRD (X-ray diffraction) were used. For those trace elements that were relatively high in abundance, XRF was particularly useful due to its nondestructive character. ICP-OES and atomic absorption were considered to be "historic", largely replaced by ICP-MS, but still could play a valuable role in the measurement of impurities in more concentrated solutions. ICP-MS was considered to be very versatile and sensitive, capable of measurement of virtually all elements that formed positive ions. However, a combination of the above techniques was often used.

Mr Varga (JRC-ITU) provided further information on ITU's work to identify characteristic parameters in uranium ore concentrates. To this end, some 50 metallic impurities are measured as well as isotope ratios in some of the trace elements (lead, strontium, neodymium). The work is being extended to cover also measurement of organics and crystal structures.

He reported on the use of the Rare Earth Element pattern in UOC samples. Rare earth elements tended to be less (or equally) affected by the processing of the uranium ore; thus they are good candidates as parameters for origin determination. Using a group extraction with TRU resin, followed by measurement by ICP-SFMS (Thermo Element 2), provided a relatively easy means to compare and assist in determining the origin of ca. 50% of UOC samples examined.

Lead and strontium isotopes proved particularly useful in classifying UOCs, because the lead composition was dependent upon the age of uranium, whilst strontium varied considerably with location. Sr.Spec resin was used to separate Sr and Pb in small bed volumes (200µl), followed by MC-ICPMS. High variability in lead isotopic composition was found between mines, but also within mines with a resulting overlap in values from different mines. Critical assessment of the data was required in terms of lead concentration and possible processing. Strontium displayed lower within-mine variation, although there was still overlap between mines. Geological samples were used as reference materials for this work.

M van Wijnkoop (Urenco) provided a report on chemical impurities from the point of view of an industrial laboratory, focusing on production control and quality control of the products. Conversion to UF6 removes non-volatile impurities, and enrichment will then enrich any remaining impurities, whilst pool-feeding is utilised at enrichment plants, mixing stock. As a consequence, it is not possible to trace the origin of material through enrichment.

ASTM-C787 and C996 specifications are used by Urenco for UF₆ feed and product, respectively. In addition, isotopic assay of UF₆ tails is undertaken together with analysis for some impurities in the feed that would otherwise cause problems (e.g. organics by FTIR, GC-MS or mass spectrometry). However, typically, the supplier's own impurity analysis data is relied upon. In the product, boron, silicon and technetium content are determined.

Analysis techniques utilised by Urenco include titrations and gas mass spectrometry, the latter providing precision off-line of better than 0.01%. Gas phase ICP-MS has also been developed for direct measurement of UF6. Unfortunately, the instrument manufacturer will not support future development of this application.

Minor isotope concentrations can yield information on the history of uranium:

- ²³⁴U in feed mine of origin;
- ²³⁴U in product down-blending or re-enriched tails; and
- ²³²U, ²³⁶U in the feed: burnup, and in the product: reprocessed uranium

Trace elements in the feed may give information on the source, but this has not been tested because Urenco's analytical requirements do not extend beyond meeting specifications. Organics can be traced, for example, to a particular pump oil used within a particular plant. But analytical methods require parts-per-billion sensitivity and are not easy to apply.

L. Keegan (ANSTO) reported on work recently completed on the measurement of anions in ITU's UOC archive samples, focussing on sulphate, phosphate and halide concentrations. A Metrohm 861 advanced compact ion chromatogram was used to undertake sample measurements, including a series of kinetic leach tests. Results were still to be assessed, to see if the data could be used to detect sample origin.

The study was only semi-quantitative, and this raised the question of how to combine semi-quantitative data. The intention of the current study was to assess anion and cation data in combination, and it was considered that additional data, when considered in combination, would assist with classification.

The Japanese Atomic Energy Authority's capabilities for fission-track TIMS, SIMS and bulk analysis were described by M. Masaaka (JAEA). XRF is used for swipe screening (as at SAL) whilst Total Reflection XRF is used to analyse for impurities on SIMS planchets prior to measurement. SEM is also used, to measure specifically for fluorine. Although impurities in particles are measured, they are not currently reported to the IAEA.

C. Puxley (AWE) presented the influence of interstitial impurities – carbon, hydrogen, nitrogen and sulphur – on the physical and mechanical properties of uranium, and the importance of measuring them. The levels of interstitial elements can give an indication of how the uranium was produced: for example, that high interstitial carbon content was indicative of preparation in a graphite mould. Combustion techniques, combined with infrared detection, were used for carbon and sulphur. For oxygen/nitrogen and oxygen/hydrogen ratios, the sample is melted in a flow of helium (for O/N) or nitrogen (for O/H). Oxygen reacts with the graphite sample crucible at 2000°C to produce carbon dioxide; nitrogen or hydrogen are analysed in the gas stream by thermal conductivity meter. The techniques are calibrated using a blank crucible and carbon steel standards, providing a 1-10 ppm detection limit and relative precision of 10%. AWE had collaborated with a single overseas laboratory in sample exchange and interlaboratory comparison.

CETAMA's activities in the area of impurity measurements were presented by A Hanssens. Interlaboratory comparison exercises within EQRAIN were undertaken for the measurement of trace elements, typically using a matrix of 2% nitric acid solution. Results were used in part to determine detection limits.

Three types of impurity uranium reference material are available from CETAMA:

- Metal chips;
- Uranium oxides; and
- Uranium compounds.

The uranium oxides were prepared by addition of solutions of impurities to the dry oxide and thorough mixing prior to measurement. Some large variations were identified between the expected and measured values of impurities in these materials. For metal chips, impurities were added to the molten metal – again with confirmatory analysis.

CETAMA is prepared to consider organizing interlaboratory exercises, but needed to consider both industrial and safeguards requirements: ore, yellow cake, oxides, and nitrate. The range of requirements had been discussed within CETAMA's uranium analysis working group, but arrangements were not yet complete.

The application of chemometric methods for evaluation of trace element analysis was subject of a talk given by O. Rondionova (Institute of Chemical Physics, Moscow). Work in the classification of uraniumcontaining samples was described, using:

- A multivariate approach;
- The presence of hidden correlations that reflect physical phenomena;
- The concept that data comprised useful information and noise.

Principal component analysis and soft independent modelling of class analogy was demonstrated and

a standard procedure to convert data into a useable form was described. Explorative analysis could be used to show differences and similarities, whilst quantitative classification enabled the classification of materials with uncertainties. Uncertainty ranges within data could be treated, although this raised concerns of the accuracy of the declared uncertainty values. It was noted that it was particularly difficult to assign uncertainties in ICP-MS because the uncertainty varied with matrix and concentration.

5. Working Group Conclusions

The questions and issues raised during the presentations were addressed in more detail in two working groups. The first one dealt with topics related to the actual measurements, while the second group focused on data evaluation and interpretation. The observations and recommendations of the working groups are given hereafter.

5.1. Measurement Quality, Measurement Techniques and Protocols

The Group noted that impurity analysis in uranium material may include:

- Minor and trace metal impurities
- Organic impurities
- Impurity analysis in uranium particles
- Isotopic signatures of heavy and light elements contained as impurities in uranium

Recognizing the relevance of impurity measurements for nuclear safeguards, non proliferation, nuclear forensics and other applications, the Group discussed best practice in sampling, measurement and sample disposal. It was agreed that participating laboratories would contribute their current practices for review and incorporation into documents to describe laboratory best practice. Standard procedures may be produced at a later date, to reflect best practice.

Detection limits and uncertainties were also considered. Even though uncertainty estimates may not currently be used in data evaluation, it is necessary to report these for incorporation into future databases and evaluation procedures. The Group recommended unification of the way in which detection limits are calculated and reported. To assist in establishing requirements for detection limits and uncertainty, current state of practice should be assessed through review of quality control charts. To this end, control samples and laboratory or procedural blanks should be measured and reported with each sample batch.

Two round-robin exercises were proposed as external quality control measures. The first would aim to assess measurement procedures through analysis of a uranyl nitrate solution containing certified amounts of a set of well defined impurities. The second, involving the analysis of solid samples with a uranium matrix (e.g. uranium oxide) would enable laboratories additionally to test their dissolution procedure and enable comparative measurements of trace elements and minor isotopes. CETAMA offered to organize such intercomparisons and expressed preparedness to take into account suggestions (on trace elements and their respective concentrations) as expressed by laboratories.

The availability of appropriate reference materials was recognised as being of critical importance. The Group recommended the production of a certified "Trace Elements in Yellowcake" standard, with certification on the basis of measurement of the original material (i.e. without spiking). Impurities certified should include rare earth elements, transition metals, calcium, sodium and potassium. The certification of those elements difficult to analyse was particularly important, to enable the reference material to be utilised in testing the performance of analysis procedures. Additional reference materials proposed (in decreasing order of priority) were:

- Trace elements in uranium dioxide;
- Isotopic reference materials (Pb, S, Nd) in uranium-containing matrices; and
- Oxygen isotopic standard in oxidic uranium matrix.

Sampling procedures were considered: both in-field and in the laboratory, with agreement that the latter would be incorporated into best practice documents. For in-field sampling, analysts should be aware of practical aspects and pitfalls for trace impurity analysis, and evaluate sampling procedures and make recommendations for improvements on a case-by-case basis.

The Group concluded with the following recommendations:

- The ESARDA DA working group should coordinate the preparation of best practice documents for impurity measurements in uranium, based upon submissions from participating laboratories;
- Uncertainties and detection limits must be reported, with a unified approach to the expression of detection limits;

- Control samples and blanks must be measured with each sample batch and reported;
- CETAMA and IRMM should give consideration to an interlaboratory comparison exercise involving measurement of uranyl nitrate and uranium oxide samples; and
- CETAMA and IRMM should give consideration to production of a certified reference material for trace elements in yellowcake, with other reference materials to be considered for future production.

5.2. Data Evaluation and Identification of Characteristic Patterns or Parameters

The second Working Group considered the two main purposes of data evaluation in the measurement of impurities in uranium:

- The identification of key parameters for utilisation in further research and development, for build a detailed knowledge base of the behaviour of impurities through process and for gaining improved understanding of the practicalities and limitations in characterising the types and origins of material through impurity measurements; and
- Enabling attribution of a specific sample to an existing data set and comparing different data sets.

Statistical tools play an important role in data evaluation, enabling quantitative comparison between samples. However, combination with scientific information is essential in order to enable a proper assessment. In particular, knowledge of the process and chemistry of the impurities is essential, and should be used to determine the optimum means of presenting data for statistical analysis. An example given was that of the behaviour of zirconium and hafnium, both exhibiting similar chemistry during the conversion process. As a consequence, whilst examination of the zirconium and hafnium concentrations found in materials may provide limited information and enable a crude match to be made between different samples, consideration of the zirconium/hafnium ratio should provide information not of the process but of the origin of the material through comparison of the zirconium/hafnium ratios from known sources.

Studies were suggested to establish the optimum sets of parameters indicative of:

 Geographic and geological origin (for example, the pattern of concentrations of rare earth elements); and • Production processes and facilities (for example, anions or organic contaminants).

This would require input both from analytical laboratories, through measurement of samples and evaluation of results, and, equally important, also from industry. Support from industry was required not just from the viewpoint of provision of sample material, but also through access to expertise and detailed knowledge of the process. Historical data from process operators was also considered to be of value, although the value may be limited by the nature of data required to meet plant specifications.

It was emphasised that the benefit to industry of their support, particularly in consideration of illicit trafficking studies, was to enable their facilities to be discounted as the source of seized material, or to assist in identifying the point beyond their plant control at which material was diverted.

The experience of scientific and industrial communities was also considered in terms of more qualitative assessment of samples. For example, the colour and visual appearance of certain materials may give an indication of their nature and help in identifying the origin. Such non-numerical information is difficult to incorporate into a statistical evaluation, but the Group considered that such information, which was relatively easy to gather, should be fed back into the evaluation. In part, this would be a question of ensuring that safeguards inspectors and other authorities made sufficient and appropriate observations, and that these were transmitted back to the data analysts.

Parameters should be prioritised, with the most important ones evaluated first. However, the order of priority might vary with material type and purity, and care would therefore be required to ensure that correct conclusions were drawn. For example, in the case of high-purity materials, different impurities and different ratios might require consideration in order to avoid either classification of all materials into a single data set or incorrect attribution due to comparison of impurity values close to or below the detection limit.

The treatment of detection limits could not be resolved during the Working Group. To the analytical chemist, the detection limit is a measurement value for a specific sample on a specific instrument. (For example, where sample material is more plentiful, a lower detection limit would be expected or, in the case of instrument instability, a higher detection limit may result.) In contrast, data analysts need to be able to compare results from different laboratories (i.e. different instruments at different times) and this comparison would be aided by common performance criteria. Similarly, an appreciation of uncertainties is required within the evaluation process, in order to assess the significance of a measured value and the weight that might be given to a result within the evaluation.

Further work would be necessary, to consider the requirements for performance standards in impurity analysis and how these might impact upon the evaluation process. If common detection limits were set, then these might result in a loss of available information from laboratories capable of exceeding the detection limit, if "less than" figures were quoted instead of a measured value. Conversely, too low a detection limit may be unattainable for some laboratories and either result in their exclusion from certain measurement exercises, or introduce cost or timeliness issues in cases where a higher detection limit may have been sufficient. Ultimately, it may simply be a case of ensuring that the instrument used and its performance are recorded by the data analysts, to enable any subsequent evaluation of the significance of results.

This highlighted one of the fundamental differences between impurity measurements in support of safeguards and illicit trafficking. For the former, the IAEA is the custodian of information: results and their evaluation are treated as safeguards confidential and, to a great extent, the laboratory has to work "blind" in supplying a wealth of data without necessarily appreciating the significance or otherwise of parameters. In contrast, in support of illicit trafficking seizures, the laboratory has the opportunity to work more closely with the authorities, applying a more systematic approach to the analysis and minimizing redundant measurements through its own ongoing evaluation of data. In this case, experience in the more qualitative aspects of attribution of a material (e.g. colour) can take on a more prominent role within the laboratory. Overall, subject to constraints of confidentiality, it was considered that greater interaction between the laboratory and authority would both reduce the analytical effort required and assist in the timely and accurate attribution of materials.

The majority of the working group discussions focused on the need to involve appropriate experts (chemists, physicists, fuel cycle and industrial plant experts) in data evaluation. Turning to the pure statistical analysis itself, a number of tools were available, had been employed by various laboratories and authorities in data evaluation, and results had been presented for different materials. The working group considered that these statistical tools should be compared, using the same dataset, with particular consideration of how best to incorporate uncertainties and limits of detection.

The working group concluded with the following draft recommendations:

- The ESARDA DA working group should explore opportunities to work with industry and share expertise, data and samples appropriate to measurement of impurities in uranium;
- Further research should be undertaken in data pre-processing, evaluating the optimum parameters for measurement and their treatment;
- Appropriate scientific and fuel cycle experts should be identified and involved in the evaluation of data obtained from impurity measurements;
- Non-numerical data should be included in the evaluation procedure;
- Further consideration should be given to the need for performance standards (International Target Values?) in impurity measurements to support safeguards;
- A comparison should be undertaken of different statistical tools using the same data set.

Acknowledgement

The workshop organizers and the authors of the present paper would like explicitly to thank S. Balsley (IAEA-SAL) for initiating the workshop and to acknow-ledge his contributions in setting up the agenda.

International Workshop on Gamma Spectrometry Analysis Codes for U and Pu Isotopics: Workshop Results and Next Steps

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Abstract

In November 2008, the Institute of Nuclear Materials Management (INMM) and the European Safeguards Research and Development Association (ESARDA) co-hosted the International Workshop on Gamma Spectrometry Analysis Codes for U and Pu Isotopics at the Oak Ridge National Laboratory (ORNL). This workshop was conducted in response to needs expressed by the international safeguards community to understand better the capabilities and limitations of the codes; to ensure these codes are sustained; and to ensure updates or revisions are performed in a controlled manner.

The workshop was attended by approximately 100 participants. The participants included code developers, code suppliers, safeguards specialists, domestic and international inspectors, process operators, regulators, and programme sponsors from various government agencies. The workshop provided a unique opportunity for code developers, commercial distributors and end users to interact in a hands-on laboratory environment to develop solutions for programmatic and technical issues associated with the various codes. The workshop also provided an international forum for discussing development of an internationally accepted standard test method. This paper discusses the organisation of the workshop, its goals and objectives and feedback received from the participants. The paper also describes the significance of the working group's contribution to improving codes that are commonly used during inspections to verify that nuclear facilities are compliant with treaty obligations that ensure nuclear fuel cycle facilities are used for peaceful purposes.

1. Background

In November 2005, the international workshop "Gamma Evaluation Codes for Plutonium and Ura-

nium Isotope Abundance Measurements by High-Resolution Gamma Spectrometry: Current Status and Future Challenges" was held in Karlsruhe, Germany. Some of the main issues discussed during the November 2005 meeting were related to concerns of the inspectorate authorities (IAEA, European Atomic Energy Community and ABACC) about the standardisation and sustainability of gamma evaluation codes. Clear guidelines were identified for future challenges (including technical developments). As a follow-up, the IAEA issued a roadmap for future developments of gamma codes, followed by a request for support for this project to several Member State Support Programmes (MSSPs) to the IAEA, namely the United States, the European Community and France. In response to this IAEA request, the International Working Group (IMWG) on Gamma Spectrometry Techniques for U and Pu Isotopics was formed. [1]

In parallel with the JRC activities described above, a different initiative was carried out under the framework of the United States–DOE/ABACC bilateral safeguards cooperation agreement with the establishment of the regional Isotopic Measurements Working Group (IMWG). The focus of this working group was to evaluate codes used to estimate uranium isotopics because of the applicability to ABACC inspections. [3]

Based on these results, those involved in the development, control and use of these gamma evaluation codes began considering expanding the participation in the project. The expanded effort broadened the scope of the working group to include plutonium isotopic measurements and established a working group that would respond to the needs expressed by the IAEA and other national and international inspectorates. Because this initiative already included many of the American subject matter experts, it was logical to look at ESARDA, and in particular to the Non-destructive Assay (NDA) working group, as the catalyst for the establishment of this dedicated working group. The proposal to launch an International Working Group on Gamma Spectrometry Techniques (IWG-GST) for U and Pu Isotopics was accepted in a meeting organised in Aix-en-Provence, France, on May 21, 2007.

The scope of the newly formed working group is to provide a forum for exchange of information, technical developments, and validation and testing of gamma-spectroscopy techniques used to determine the isotopic composition of uranium and plutonium samples. Specifically, but not exclusively, the IWG-GST will address issues related to gamma evaluation codes, such as their applicability, capability and limitations, standardisation, sustainability, and version control. The IWG-GST is open to all international participants that are interested in issues related to gamma spectrometry analysis codes. Terms of Reference have been drafted that define the working group's mission and objectives and provide guidance for participation. The workshop described below was one of the initial activities recommended and coordinated by this newly developed working group. [4]

2. International Workshop on Gamma Spectrometry Analysis Codes for U and Pu Isotopics

Under the purview of this newly formed working group, from 3–7 November 2008, the INMM CRC in conjunction with the MC&A Division and ESARDA co-hosted the International Workshop on Gamma Spectrometry Analysis Codes for U and Pu Isotopics at ORNL. The workshop was sponsored by the DOE/NNSA International Nuclear Safeguards and Engagement Program (INSEP) that is coordinated



within the Office of Global Security Engagement and Cooperation (NA-242).

The workshop had several objectives. The first objective was to allow the end users an opportunity to interact with many of the original code developers and current commercial distributors in a hands-on laboratory environment to understand better the application and limitations of the various codes. The second objective was to provide an opportunity for workshop participants to develop cooperatively solutions for programmatic and technical issues associated with each code's capabilities, limitations, applicability, sustainability and version control. The third objective was to provide a unique opportunity for code developers and commercial distributors to interact with the user community to understand better the challenges that the users face when utilising the codes in a laboratory or field environment. Finally, the ultimate goal of the workshop was to discuss and exchange ideas on software quality assurance issues and promote development of an internationally accepted standard test method to validate, sustain and administratively control distribution of current and newly developed codes. To achieve this goal, code developers, code suppliers, safeguards specialists, domestic and international inspectors, process operators, regulators, equipment developers, material control and accounting personnel, and programme sponsors from various government agencies were encouraged and invited to participate in the workshop. Appendix A provides information on the organisations and technical disciplines that participated in the event. The workshop was attended by 101 participants, 30% of whom represented the following countries (the rest being US): Argentina, Austria, Brazil, France, Germany, Hungary, Italy and the United Kingdom.

The workshop was one week in length. It included a key note speech, invited presentations, code and equipment demonstrations, hands-on exercises that included measurements of special nuclear materials, group discussions, and tours. Complete workshop materials and presentations are also available at the INMM and ESARDA websites [5,6].

The key note speech by Dr. Rolf Arlt provided a detailed overview of the importance that gamma spectrometry codes played in safeguards verification activities. The remainder of the first day was dedicated to presentations from various code developers. Each of the presentations discussed the history of the codes, drivers for development of the codes, theory of operation, techniques used for validating and verifying results generated by each code, and information on the codes' applications and limita-



tions. Table 1 provides a summary of the codes that were covered during the workshop.

FRAM	NaIGEM	WinUF6
MGA	MGA++	U235HI
MGAU	WinU235	IGA

Table 1: Gamma Spectrometry Codes Demonstrat-ed During the Workshop.

The morning sessions of the second and third day focused on presentations from end users. Technical personnel from the IAEA, national and international research institutions and the nuclear fuel cycle provided insight to issues commonly encountered when the codes are used in a laboratory or field measurement environment. The end users provided information on challenges that are commonly faced for their specific application. These presentations provided code developers with an opportunity to understand end user issues.

The workshop also included dedicated time in the Oak Ridge Safeguards Laboratory for each individual to participate in measurement exercises. The measurement stations were designed to provide each participant with an opportunity to spend some time conducting measurements on special nuclear materials with the original code developers and current commercial distributors. Each of the groups also actively participated in analysis sessions for each of the codes. These sessions were set up in a conference room that was separate from the measurement stations. The code developers lectured the participants on the theory of operation for the codes.

A session of the workshop was also dedicated to discussing issues related to software quality assurance. The participants openly debated the need for version control, software validation measures and the need for developing a standard test method for such codes. The results of the panel discussion were noted in a presentation that each of the groups gave during the final afternoon of the workshop.

After all workshop activities were completed, the International Working Group on Gamma Spectrometry Techniques held a meeting. The purpose of the meeting was to introduce workshop participants to the newly formed working group and to discuss the Terms of Reference that were developed by the working group co-chairs. The meeting also included an overview of a website that is being supported by the ESARDA NDA Working Group. Mr. Paolo Peerani, the IWG-GST co-chair, provided an overview of the IWG-GST website. The website is designed to provide a forum for discussion on issues related to gamma spectrometry analysis codes. It was suggested that the website be used to provide information on a standard test method, application guide and version updates for the various codes. The website may also be used to access a standard set of spectra that can be used to test and validate gamma spectrometry analysis codes.

3. Workshop Results and Next Steps

Both the workshop organisers and participants agreed that the International Workshop on Gamma Spectrometry Analysis Codes for U and Pu Isotopics was a great success and achieved all of its goals and objectives. One of the primary objectives of the workshop was to provide a forum for interaction between end users and code developers. Because of the workshop, code developers have a better understanding of end user needs and end users have a better understanding of the capabilities and limitations for each of the codes. The workshop provided a great opportunity for the representatives of the international safeguards community to work with and learn from the various subject matter experts. This interaction enhances the ability of safeguards inspectors to meet safeguards objectives from a domestic and international safeguards perspective by improving the data quality objectives for these codes. In addition, the format of the workshop facilitated an open dialogue between the code developers, end users and commercial distributors on the programmatic and technical issues associated with each code's capabilities, limitations, applicability, sustainability, and version control.

The workshop also provided a forum for discussion and debate on software quality assurance issues and development of an internationally accepted standard test method to validate, sustain and administratively control distribution of current and newly developed codes. It was concluded that there is a need for a standard test method; however, it should be implemented with constraints (for example, restrictions a certification could impose on the use of codes, etc.). Therefore, it is necessary to weigh all of the costs and benefits associated with developing a standard test method. The general consensus was that developing a standard test method for these codes is a complex matter that requires a rigorous, structured approach.

Finally, the workshop facilitated the goals and objectives of the International Working Group on Gamma Spectrometry Techniques by raising the awareness of challenges faced by end users of these codes. The IWG-GST will continue to support development of an internationally accepted standard test method and best practice guide that promotes a common understanding of the capabilities and limitation for these codes. The IWG-GST will provide a mechanism for open discussion through the ESARDA NDA Working Group website. The IWG-GST has taken the recommendations of the workshop under advisement and is actively working to accumulate a standard set of spectra that is accepted by the international community to test and validate these codes. The IWG-GST is also working

to develop a concise best practice for each of the codes. The IWG-GST is also working with code developers to provide information on the most commonly used versions for the individual codes.

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Regional and International Inspectorates				
Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC)	EURATOM	International Atomic Energy Agency (IAEA		
Government Regulatory Agencies				
French Institute of Radioprotection and Nuclear Security (IRSN)	Argentine Nuclear Regulatory Authority (ARN)	National Nuclear Energy Commission of Brazil (CNEN)		
French Atomic Energy Commission (CEA) - Saclay	U. S. Defense Nuclear Facilities Safety Board	U. S. Department of Energy National Nuclear Security Administration		
	Research Institutions			
European Commission - Joint Research Centre Institute for the Protection and Safety of the Citizen	Institute of Isotopes (Hungary)	European Commission Institute for Transuranium Elements		
Oak Ridge National Laboratory	Los Alamos National Laboratory	Lawrence Livermore National Laboratory		
Brookhaven National Laboratory	Sandia National Laboratory	Pacific Northwest National Laboratory		
Fraunhofer- INT	Savannah River Site	Y-12 National Security Complex		
Nuclear Ins	strumentation Developers and Commerc	cial Vendors		
Areva Federal Services	Mirion Technologies	NorthWest Nuclear, LLC.		
Canberra Industries	NucSafe, Inc.	ORTEC/Ametek		
Sampson Professional Services	ICX Radiation, Inc.	Amtek UK		
GBS Elektronik GmbH				
Nuclear I	Fuel Cycle Facilities and Government Co	ontractors		
B&W Pantex	Applied Research Associates, Inc.	Energy Solutions		
S. Cohen & Associates, Inc.	WTI/Fairfield	USEC Inc.		
Westinghouse Safety Management Systems				

Appendix A

Workshop on Environmental Monitoring

M. B. Kalinowski

Carl Friedrich von Weizsäcker Center for Science and Peace Research, University of Hamburg

In 1997, the Additional Protocol opened the doors for nuclear inspection capabilities to become more effective and powerful. Satellite images have since been applied with increasing success to the remote detection and analysis of unreported nuclear material production plants. Environmental sampling has, so far, mainly be applied within inspected facilities whilst the potential of stand-off and remote detection has not yet been brought to bear.

Progress in safeguards methodologies based on environmental sampling is not only urgently needed with regard to implementing the Additional Protocol related to NPT safeguards. It would at the same time address verification issues for a Fissile Materials Cutoff Treaty (FMCT).

To address this need, and to support the IAEA Novel Technologies Programme in developing novel techniques and instruments, the independent Group of Scientific Experts (iGSE) dedicated to the development of new approaches for the detection of unreported nuclear-weapons-materials production was established in 2006. This topic was discussed earlier in the framework of the ESARDA working group on Verification Technologies and Methodologies. It was discussed again in this framework at a joint iGSE-ESARDA workshop, as part of the 30th ESARDA Annual Meeting on 28-29 May 2008 in Luxembourg.

The workshop was co-chaired by the chairman of the ESARDA WG VTM, Gotthard Stein (Research Centre Jülich), and the chairman of iGSE, Martin Kalinowski (Carl Friedrich von Weizsäcker Centre for Science and Peace Research, University of Hamburg).

The first presentation was given by Bhupendra Jasani (Department of War Studies, King's College, London), providing an overview of the status and progress of satellite imagery for international verification. Whilst reactors can readily be identified, reprocessing facilities have no unique optical signature. The thermal emissions of uranium enrichment plants based on diffusion might be used as an indicator, but the newly built plants that make use of centrifuge technology cannot be detected because of their low thermal signature. Remote environmental sampling is under investigation as a complementary approach to satellite imagery, to obtain indications of unreported nuclear material production at a distance. Amelie Hubert (Direction des Application Militaire, Commissariat à l'Energie Atomique) introduced the state-of-the art of environmental sample analyses technologies. The focus of experience lies in particle analysis from swipe samples.

For verification of the Comprehensive Nuclear-Test-Ban Treaty (CTBT), remote and stand-off technologies are already being implemented in order to detect tiny traces of anthropogenic radioactivity in the environment. Mika Nikkinen (Provisional Technical Secretariat of the Preparatory Committee Comprehensive Test Ban Treaty Organization, Vienna) introduced the International Scientific Studies (ISS) activities that are reviewing the status of the build-up of these instruments. The goal of the ISS is to assess the verification capability of the CTBT. Much of these developments and experiences can be transferred to the needs of the IAEA. Two other presentations report the two major approaches taken for CTBT verification. One is continuous verification, based on the International Monitoring System (IMS) with 80 radionuclide stations distributed over the globe. The second is on-site inspections (OSI), to be conducted in the field in the case of a suspected nuclear test. Wolfango Plastino (University degli Studi Roma Tre) explained the environmental sampling activities as well as airborne and carborne radioactive monitoring procedures performed during the OSI exercise in Kazakhstan 2006. Franca Padoani (Bologna Research Center ENEA) and Robert Werzi (CTBTO PrepCom) reported on experience from operating the IMS radionuclide stations that is of interest for remote environmental sampling for nuclear safeguards.

More generally, Vitaly Fedchenko (Stockholm International Peace Research Institute) considered the possible synergies with other verification frameworks that environmental monitoring techniques utilised by the IAEA would have.

Julian Whichello (IAEA) gave an update on the IAEA Novel Technologies Programme and Andrew Monteith (IAEA) described the exercise on gathering indicators and signatures conducted under the lead of Cynthia Annese (IAEA).

From extensive studies of environmental sampling for safeguards in the 1990s, the experts agree that krypton-85 has the best features to be suitable for the detection of plutonium separation from irradiated nuclear fuel at a distance. However, establishing a network of stations in a wide-area-environmental sampling (WAES) approach has been assessed to be too expensive. New findings have led the IAEA to anticipate that krypton-85 sampling might prove to be effective and cost efficient.

The specific krypton-85 source term associated with the production of one significant quantity of plutonium depends on the applied production scheme. Paul Stanoszek (Carl Friedrich von Weizsäcker Centre for Science and Peace Research, University of Hamburg) has studied these schemes systematically and presented the krypton-85 activity per kilogram of plutonium. As a result, the lowest specific activity released per kg plutonium is caused by the irradiation of depleted uranium. A related topic was covered in the presentation of Paolo Peerani (Joint Research Centre of the EU Commission, Ispra) on simulation results achieved in collaboration with Jochen Delbeke and Greet Janssens-Maenhout. The isotopic analysis of fission noble gases and other radionuclides allow for the characterisation of reprocessing fuel by stack measurements, as well as by remote environmental sampling.

The workshop was rounded up by a discussion on common future activities regarding remote environ-

mental sampling for nuclear safeguards. A followup meeting took place on 2-4 November 2009 in Vienna. It marked the end of the first phase of the iGSE network and summarized the current state-ofthe-art.

The most promising environmental signature for unreported nuclear weapons material production that can still be detected at a distance is krypton-85 resulting from plutonium separation. New studies undertaken under the German Support Programme for the IAEA at the University of Hamburg established that krypton-85 releases from a small facility producing just one significant quantity (8 kg) per year can still be detected against the background variations at distance of 500 km with a probability of about 50%. However, it is still under investigation as to what extent a detection can be associated with a confined geographic area as a possible source location.

For detection of unreported production of highly enriched uranium, the task is even more challenging, because the signatures are weaker. In fact, they appear to be typically too weak to be even detected with a stand-off system in close vicinity of the release point. In this inspection scenario, the IAEA would apply a mobile system that could sense the off-gases of industrial facilities from outside their fence.

Materials of the iGSE meeting at Vienna in November 2009, as well as of the joint ESARDA-iGSE workshop in Luxembourg in May 2008, can be accessed at the iGSE website (www.igse.net).

Fingerprinting of Nuclear Material for Nuclear Forensics

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Abstract

Unknown nuclear material may originate from several sources. Nuclear forensics allows by using fingerprinting and comparison with reference data to determine the origin, the intended use, the last legal owner and the smuggling route. These information are essential in the cause of theft or diversion as measures of safeguards can be implemented to prevent future thefts.

Certain measurable parameters can point to a specific material and provide therefore a 'fingerprint' of the unknown material. Comparing the measured parameters with reference material give clues to the origin and the last legal owner.

Characteristic parameters and possible information they contain are presented.

Keywords: nuclear forensics; uranium; plutonium.

1. Introduction: What is nuclear forensics?

As the former Soviet Union disintegrated in the early 1990s a new phenomenon was discovered – 'nuclear smuggling'. The first cases were reported in 1991 in Switzerland and in Italy. New questions had to be answered: intended use of the unknown nuclear material, its origin, the last legal owner and the smuggling route. Techniques to answer these questions were known from nuclear safeguards and material science. Combining these analytic methods was the starting point of nuclear forensics.

Information that is obtained by nuclear forensic analysis can be divided into two groups: endogenic and exogenic information.

Endogenic information is meant as self-explaining information as age, intended use and mode of production, for which only model-calculation might be required for data interpretation.

Exogenic information is meant as information by comparison. To interpretate exogenic information which include geolocation and production data comparison with reference data and reference information is necessary. Great efforts are made to set up databases containing such reference data.

Case development in nuclear forensics follows a deductive way (Fig. 1). Taking the available results into account a hypothesis or a set of hypotheses are built. Databases and other experts serve for the knowledge to build the hypothesis. Further analysis has to be made to prove the presence of signatures according to the hypothesis. If the signatures are absent in the sample a new hypothesis has to be developed.

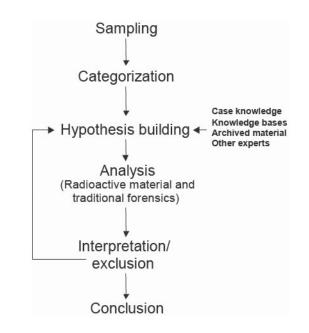


Figure 1: The deductive way in the nuclear forensic process [1].

2. The "nuclear fingerprint" method

To characterize an unknown nuclear material a set of items are measured to establish the so called nuclear fingerprint. Information that form the nuclear fingerprint are:

- Macroscopic parameters (e.g. pellet diameter, height)
- Isotopic composition

• Elemental composition (such as elements and impurities)

In cases of a mixture of components, a powder or if it may be possible that substances of different chemicals or isotopic composition have been added, it is necessary to investigate the microstructurual fingerprint which consists of following items:

- Particle morphology
- Particle size and size distribution
- Grain size and size distribution
- Porosity size distribution and density
- Dislocation density and character
- Precipitation of other phases.

2.1. Isotopic patterns of U and Pu

The following information can be obtained by investigating the isotopic patterns:

- The presence of small amounts of U-236 will indicate a contamination with recycled uranium and hence point at reprocessing activities.
- The isotopic composition of plutonium is a useful indicator of the reactor type in which the material was produced.
- Uranium oxide can be found in different forms, e.g. UO₂ or U₃O₈, which give information on various points of origin in the uranium fuel cycle.

Plutonium is generated as by-product in nuclear reactors when ²³⁸U absorbs a neutron creating ²³⁹U, which β -decays into ²³⁹Np and finally to ²³⁹Pu. Also heavier isotopes of Plutonium are produced by further neutron captures. Therefore the isotopic composition may give answers which reactor type was used to produce the unknown nuclear material.

The isotopic composition of Plutonium provides information to indicate the type of the reactor:

- The higher the initial ²³⁵U enrichment of the fuel is, the higher is the ²³⁸Pu abundance due to multiple neutron capture on ²³⁷Np.
- The neutron energy spectrum influences the Pu isotopic composition (the softer the spectrum, the higher is the ²⁴²Pu/²⁴⁰Pu ratio).

The measured isotopic patterns can be compared with model calculations using computer codes (e.g. ORIGEN or SCALE for the RBMK and the VVER). It was demonstrated that the main reactor types can be separated clearly from each other (Fig. 2).

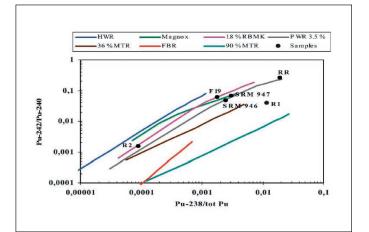


Figure 2: Calculated isotopic composition of Plutonium for various reactor types (continuous line) and measured isotopic composition (points) [2].

2.2. Age determination

The age is defined as the time elapsed since the last chemical processing took place (e.g. production, reprocessing or purification). The age of the material may serve as exclusion parameter in the search for the production or reprocessing plant.

Since radioactive isotopes decay at a rate determined by the initial amount and the half-life of the isotope, the relative amounts of decay products (daughters) in comparison to the parent isotope can be used as chronometer.

The age of the material is short in comparison to the half-life of the observed nuclides. Using the table of nuclides many parent/daughter pairs can be found. The optimal nuclide ratios for Uranium are:

- ²³⁴U/²³⁰Th
- ²³⁵U/²³¹Pa

and for Plutonium:

- ²³⁸Pu/²³⁴U
- ²³⁹Pu/²³⁵U
- ²⁴⁰Pu/²³⁶U
- ²⁴¹Pu/²⁴¹Am.

The radioactive decay of each of these nuclides is unique, therefore measuring the parent/daughter ratio allows to calculate the time elapsed since the last chemical separation. If the material has not been fully separated, the chronometer hasn't been set properly to zero and so misleading information about the age is gained. To avoid this systematic error it is recommended to measure various parent/ daughter ratios.

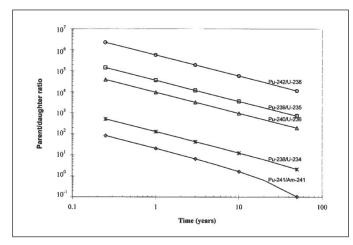


Figure 3: The parent/daughter mass ratios of aged plutonium material [3].

2.3. Metallic impurities

Nuclear material contains metallic impuritites at varying concentrations. In the starting materials metallic impurities are accompanying elements, during processing to the intermediate and final product the impurities are drastically reduced. A reduction factor of 10³ in the impuritity level is possible after processing. Impurity patterns within the processing remain for the most mines the same.

Otherwise metallic impurities may enter in the nuclear material at the different processing stages. The systematic behind this is not well understood up to now, but a full theory must contain that the concentration of the impurities are a function of exposure time to the container material or storage tank as they are leached from the surface of the walls.

Nowadays in sample analysis the ratio of elements of similar chemical behaviour are examined because the ratio will vary only within narrow limits.

2.4. Stable Isotopes

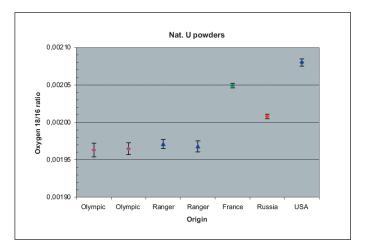
Measuring the stable isotopes is a established technique in geolocation. Two substances which are chemically identical have different stable isotope compositions if either their origin and/or their history are not the same. In nuclear forensics the oxygen and the lead isotope ratio measurements are applied.

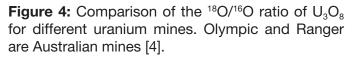
2.4.1. Oxygen

Natural oxygen exists in three stable isotopes: ^{16}O (99.762%), ^{17}O (0.038%) and ^{18}O (0.200%). Temperature, latitude and distance to the sea cause variations up to 5% in the $^{18}O/^{16}O$ ratio.

Water is used as a common solvent in uranium processing. During the processing isotopic ex-

change takes place and the final U-oxide product carries the signature of the ¹⁸O/¹⁶O ratio of the used water. Measuring the oxygen isotope ratio provides information about the geographical region where the material was processed. Figure 4 shows the correlation between the geographic location of the production site of uranium oxid examples and the variation in the (¹⁸O)/(¹⁶O) ratio.





2.4.2. Lead

From the four stable lead isotopes one is primordial (natural) ²⁰⁴Pb and the other three are endproducts of radioactive decay series: ²³⁸U \rightarrow ²⁰⁶Pb, ²³⁵U \rightarrow ²⁰⁷Pb, ²³²Th \rightarrow ²⁰⁸Pb. Therefore the stable lead isotope composition gives information on the initial U/Th ratio in the mine and on the age of the ore. Due to the fact that the variations in the composition for different mines are significant, investigating the stable lead isotopes can locate the origin mine.

2.5. Anionic impurities

Uranium crude ore undergoes different chemical processes before it becomes uranium ore concentrate (yellow cake). Since uranium is mined from ores with different mineralogical natures (acidic, alkaline and phosphatic ores) different chemical leaching processes are used. In the subsequent processing also different chemical processes are used for the precipitating and dissolving the uranium.

Different acids are used for the different processes and may leave anionic impurities (Cl⁻, F⁻, Br⁻, NO₂⁻, NO₃⁻, SO₄²⁻ and PO₄³⁻). These anions may be an indicator for the process used. In practice anion ratios are used because the leaching rate in the mines can change and absolute anion concentrations differ more than the ratios.

It has been shown that anionic impurities can distinguish between different mines (Fig. 5), but there are also differences between different sampling campaigns in the same mine (Ranger-old vs. Rangerfresh and Beverly-old vs. Beverly-fresh in Fig. 5).

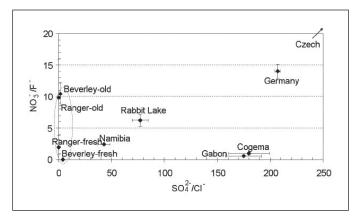


Figure 5: Anion ratios in different uranium ore concentrate samples [5].

2.6. Limitations of these techniques

2.6.1. Cross contamination

There are two problems which may cause cross contamination by investigating the stable lead isotopes. First, natural lead (²⁰⁴Pb) is omnipresent – so special care has to be taken performing the chemical separation – second, lead is often used as shielding material.

Presently the methods of the analysis are continuously improved. If the methods are sensitive enough to detect even metallic impurities from the spatula used to collect the evidence, new problems for the analysts are created.

2.6.2. Reprocessing and enrichment

Presence of ²³⁶U can point to reprocessing activities. It has also been shown that in natural uranium variations in ²³⁶U and in ²³⁴U abundances occur. Better measurement methods have to be developed for ²³⁶U abundance levels close to natural abundance.

If nuclear material was reprocessed for non-peaceful purposes the identification of this material is very challenging. Additional information is necessary to calculate predictive signatures for enriched uranium produced from reprocessed uranium [8]:

- The ²³²U content does not only depend on the operation mode of the production reactor but also on non-reactor-related history (i.e. length of storage periods before and after irradiation).
- HEU may be enriched in a single cascade, but it is also possible to use several interconnected smaller cascades.
- The identification of the enrichment process (gaseous diffusion vs. gas centrifuge) due to small differences in the concentrations of the trace uranium isotopes (²³²U, ²³⁴U and ²³⁶U) is very challenging.

2.6.3. Blending

A special challenge to determine origin of nuclear material arises if it is blended. In former times Russia blended the spent VVER fuel with spent fuel of propulsion reactors, after reprocessing this nuclear material is suitable for RBMK reactors [9].

Therefore the nuclear material measured value of the ²³⁸Pu/tot.Pu vs. ²⁴²Pu/²⁴⁰Pu value will not fit the simulated values (Specimen R1 in Fig.2).

Sometimes however cross-contamination has occurred before collection to disguise the origin. Transmission electron microscopy (TEM) is used to determine the grain-size distribution and can therefore indicate that a powder consists of different particle types.

2.6.4. Computer codes

Estimating the uncertainty of all quantities used in the calculations (i.e. cross sections) and determining how these uncertainties propagate through the entire simulation is called uncertainty analysis.

Uncertainty analysis is not implemented in the ORI-GEN2 and SCALE code.

An interesting overview how to determine the uncertainty in such computer codes is given in [10].

3. Overview of analytical techniques used in nuclear forensics

Due to the tremendous fingerprint diversity and the requirement of high accuracies of measurement many different analytic techniques have to be applied in nuclear forensics. Figure 6 gives a short overview of information gained from Uranium and Plutonium samples and which analytic techniques are required to obtain this information. A short overview of most widely used techniques is given in [2].

3.1. Including traditional forensic methods

"Wherever he steps, whatever he touches, whatever he leaves, even unconsciously, will serve as a silent witness against him. Not only his fingerprints or his footprints, but his hair, the fibers from his clothes, the glass he breaks, the tool mark he leaves, the paint he scratches, the blood or semen he deposits or collects. All of these and more, bear mute witness against him. This is evidence that does not forget. It is not confused by the excitement of the moment. It is not absent because human witnesses are. It is factual evidence. Physical evidence cannot be wrong, it cannot perjure itself, it cannot be wholly absent. Only human failure to find it, study and understand it, can diminish its value."

Dr. Edmond Locard

This is the famous Locards Exchange Principle that states that every contact leaves a trace. Traditional forensic methods establish relations between locations, events and individuals - entirely other information than the nuclear forensic methods. In summary forensic methods provide information adherent to the material while nuclear forensic methods provide information inherent to special nuclear material [7].

A detailed description of collecting both nuclear and traditional forensic evidence is given in the Nuclear Forensics Support [1]. During the collection of evidence and analysing the evidence in a laboratory special attention has to be taken on sufficient protection against the radiation.

To fulfil good radiological safety practice the Institute for Transuranium Elements developed jointly with the German Federal Criminal Police a so called glove-box, where contaminated evidence can be visually inspected, photographed and fingerprints and DNA samples can be taken (Fig. 7).



Figure 7: Glove-box for handling of special nuclear material [7].

4. Data interpretation

Measured exogenic information (such as ¹⁸O/¹⁶O ratio, Pb isotopic composition, impurities and microstructure) needs comparison with reference data from known samples.

4.1. Supporting information

It is important to have access to reference data and to keep information, that may vary with time (e.g. as seen in 2.5. level of impurities), up to date. The reference information can be gained from Nuclear Materials Database, Open Literature, ITDB, IAEA Research Reactor Database, other databases and comparison samples.

The Nuclear Materials Database was established in collaboration between the ITU in Karlsruhe with the A. A. Bochvar Institute in Moscow. The database contains information on fuels for commercial reactors as collected from open literature and from bilateral agreements with fuel suppliers.

Parameter	Information	Analytical technique
Appearance	Material type (e.g. powder, pellet)	Optical microscopy
Dimensions (pellet)	Reactor type	Database
U, Pu content	Chemical composition	Titration, HKED, IDMS
Isotopic composition	Enrichment \Rightarrow intended use; reactor type	HRGS, TIMS, ICP-MS, SIMS
Impurities	Production process; geolocation	ICP-MS, GDMS
Age	Production date	AS, TIMS, ICP-MS
¹⁸ O/ ¹⁶ O ratio	Geolocation	TIMS, SIMS
Surface roughness	Production plant	Profilometry
Microstructure	Production process	SEM, TEM

HKED, hydrid K-edge densitometry; IDMS, isotope dilution mass spectrometry; HRGS, high-resolution gamma spectrometry; TIMS, thermal ionisation mass spectrometry; ICP-MS, inductively coupled plasma mass spectrometry; SIMS, secondary ion mass spectrometry; GDMS, glow discharge mass spectrometry; AS, alpha spectrometry; SEM, scanning electron microscopy; TEM, transmission electron microscopy.

Figure 6: Information that can be obtained from nuclear (U, Pu) material and used analytic techniques [6].

Certain data have limited accessibility (commercially sensitive data such as chemical impurities) or are not shared (detailed information on weapons grade material).

4.2. Exclusion Principle

In order to avoid a full characterization each time when unknown nuclear material is found, the exclusion principle is applied. The exclusion principle works as follows:

- 1. The first measured information (e.g. pellet dimensions and isotopic composition) are compared with the database entries.
- The non-matching records are rejected as they could not be manufacturer of the unknown nuclear material.
 Evit condition: A single record is left. This is the

Exit condition: A single record is left. This is the best case.

 The remaining records, which match the measurements, are compared to each other in order to identify parameters which could distinguish between these records.

Exit condition: No further parameters can be investigated.

- 4. These parameters are measured next.
- The measured data are compared with the reduced database entries. Go back to 2).

5. Conclusion

Combining various analytic techniques can give information about the origin, the intended use, the last legal owner and the smuggling-route of unknown nuclear material. But this is only possible if enough reference datas are available and accessible.

Therefore it is necessary to strengthen the international cooperation and the cooperation with the fuel manufactors. Since the beginning of nuclear forensics in the early 1990s more and more parameters proved to be useful and could be applied for the nuclear fingerprint. Hence it is necessary to do furthermore research and development and to keep close cooperation with other sciences whether new characteristic parameters can be investigated.

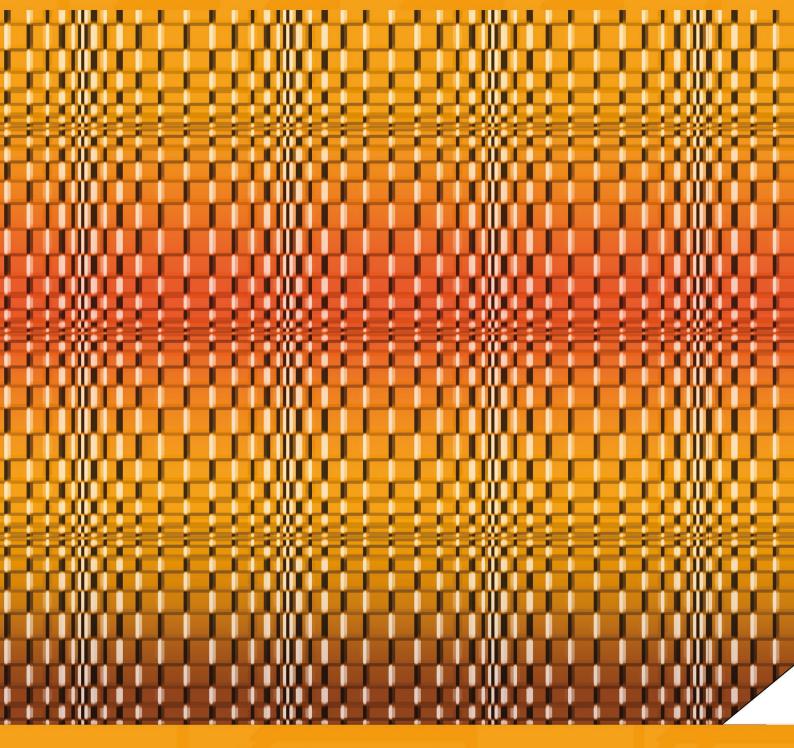
6. Acknowledgement

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A celebrative cover for the 40th anniversary of ESARDA. As a side note, the cover is inspired by the *Abbey Road* LP recorded by the Beatles in the same year: 1969.





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