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## Nuclear forensics within a Nordic context

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## **Abstract**

The NKS NUFORNOR project was initiated to strengthen the competence of nuclear forensics and to increase the collaboration within this field in the Nordic countries. The seminar “NKS-B seminar on nuclear forensics in Nordic countries” was arranged in Oslo October 5<sup>th</sup> -6<sup>th</sup>, 2015, with 26 participants from 7 countries and 17 presentations. The goal of the seminar was to provide information on the necessity and suitability of novel analytical techniques within Nordic nuclear forensics as well as exploring possibilities for collaboration across institutions and borders within the Nordic countries. Furthermore, selected analytical techniques such as QQQ-ICPMS, gamma/X-ray spectrometry with post processing of coincident signals as well as micro-RAMAN combined with XRD were tested on relevant samples. Recommendations for nuclear forensics in the Nordic countries include the need to leverage on existing knowledge at nuclear and environmental research institutes and international cooperation. Furthermore, new analytical techniques such as rare earth element elemental ratio analyses, micro-RAMAN and synchrotron based x-ray techniques should be included in the nuclear forensics toolbox. There is also a need of reference materials and proper uncertainty estimation procedures.

## **Key words**

Nuclear, analytical tools, international cooperation, material out of regulatory control, attribution process.

# **Final report with analytical recommendations for nuclear forensic purposes in the Nordic countries**

**Final Report from the NKS-B NUFORNOR activity (Contract: AFT/B(15)10)**

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## 1. Introduction

According to the IAEA (2015), nuclear forensics is “the examination of nuclear or radioactive material, or of evidence that is contaminated with radionuclides, in the context of legal proceedings under international or national law related to nuclear security”. The material could be recovered from capture of unused, often illicit materials, which is out of regulatory control, or from e.g. radioactive debris following a nuclear explosion from an improvised nuclear device (IND) or a radiological dispersal device (RDD). Internationally, nuclear forensics is a strong discipline encompassing several scientific fields and a broad spectrum of techniques that provide advanced tools to identify sources and production methods of materials and thereby attribute the materials and/or nuclear device to its originators. Furthermore, nuclear forensics can elucidate the key features of a detonated nuclear device. Thus, nuclear forensics should be a prioritized field within the Nordic countries, as the competence to characterize exposure from unforeseen nuclear or radiological events i.e. release of nuclear or radioactive material out of regulatory control is rather limited. Thus, the Nordic community would benefit from acquiring more knowledge and competence on nuclear forensics including the use of advanced analytical techniques. Importantly, such competence and training must be acquired prior to a real situation. Techniques associated with this field are continuously developing internationally, for instance within nano science, where submicron entities are focused. With new tools follows questions such as: why and how these techniques can be applied. Such knowledge can open up and provide insight into new fields not only related to nuclear forensics or even nuclear material, but it may also be associated to the oil/gas industry (e.g. behaviour of micrometre sized Ba(Ra)SO<sub>4</sub> particles) or sorption of radioisotopes on to surfaces (medicine, industry, nuclear waste). The point is that by learning and applying a variety of techniques, we can build competence and capability that are applicable to several fields within the radioprotection area of importance for the Nordic countries. Thus, synergistic effects should evolve where the competence on behaviour of nanometre to micrometre radioactive particles can be combined with problems in other fields. Nuclear forensics pose technically complex challenges both for the scientific and the law enforcement communities, and difficulties involved especially for attribution processes, should not be underestimated.

The goal of the present project has been to increase the awareness and sharing knowledge of the large potential for nuclear forensics to play a crucial role in the analysis of both pre- and post-detonation of nuclear and other radioactive material. This includes Material Out of Regulatory Control (MORC) and at the same time pointing at challenges and problems related to analysis of nuclear or radioactive material released from a source of unknown composition and origin, both in an internationally and in a Nordic perspective.

The overall objective of the project was to initiate a close collaboration between institutions in the Nordic countries within nuclear forensics, by maximising the exploitation of new and existing techniques and equipment across institutions and borders. The work particularly focused on:

1. Testing selected analytical techniques on relevant samples available in the participating laboratories. In the final report, we include results from this part of the project in chapter 2 entitled “Testing selected analytical techniques”.
2. The activity included also a seminar in October 2015 in Oslo, Norway entitled “NKS-B seminar on nuclear forensics in Nordic countries” with invited speakers from authorities (end users) and experts within nuclear analytical chemistry. The goal of the seminar was to provide information on the necessity and suitability of novel analytical techniques

within Nordic nuclear forensics as well as exploring possibilities for collaboration across institutions and borders internationally including within the Nordic countries. The seminar is summarized in chapter 3.

3. Develop procedures for cost effective nuclear forensic work adapted to specific nuclear scenarios and events including screening of exposure and recommended analytical techniques. The final project report includes the chapter "Recommended procedures and analytical techniques adapted for specific scenarios and nuclear events in the Nordic countries", which is based on the seminar contributions (chapter 4). Chapter 4 also includes some analytical recommendations for nuclear forensic purposes in the Nordic countries.

## **2. Testing selected analytical techniques**

Three of the project partners tested analytical techniques that can be useful in a nuclear forensics context. Norwegian University of Life Sciences (NMBU) developed and tested a method for analysis of Pu isotopes utilizing the relatively new Agilent 8800 ICP-QQQ-MS (dual quadrupole ICP-MS with octupole collision / reaction cell) with excellent analytical capabilities (Chapter 2.1). FOI utilized XRD and  $\mu$ -Raman measurements to characterize uranium oxides (Chapter 2.2), while Risø DTU developed and tested a gamma and x-ray spectrometry system with post-processing of coincident signals (Chapter 2.3).

### **2.1 Development and testing of a method for analysis of Pu isotopes using ICP-QQQ-MS**

Plutonium (Pu) is a mainly anthropogenic element generated by neutron irradiation of U isotopes in reactors and during nuclear detonations. Pu isotope ratios depends on irradiation intensity and time and allows for distinction between different sources, for instance weapon production, naval and commercial power production.

Accurate determination of Pu isotopes depends heavily on the removal of matrix elements and interfering elements. Due to the high abundance and mass proximity of uranium relative to Pu this element is of particular concern. U is present in most samples at a seven orders of magnitude excess over Pu (Tanner et al., 2004). The presence of U in the final sample hamper determination of  $^{238}\text{Pu}$  by mass spectrometry, moreover uranium hydrides interferes with the measurements of plutonium on mass 239. U is normally reduced through chemical separation, most often ion chromatographic and / or ion separation methods, however, these methods are time consuming and labour intensive. Moreover, due to the high natural abundance of U, orthogonal separations are often required to sufficiently reduce U in the final sample. This adds further to the work-load.

New ICP-MS instrumentation offering double consecutive mass spectrometers separated by gas collision cells provides the possibility of performing in-line interference removal. Depending on sample characteristics, this new technique could greatly reduce, or even eliminate the need for elaborate chemical pre-separation of the samples.

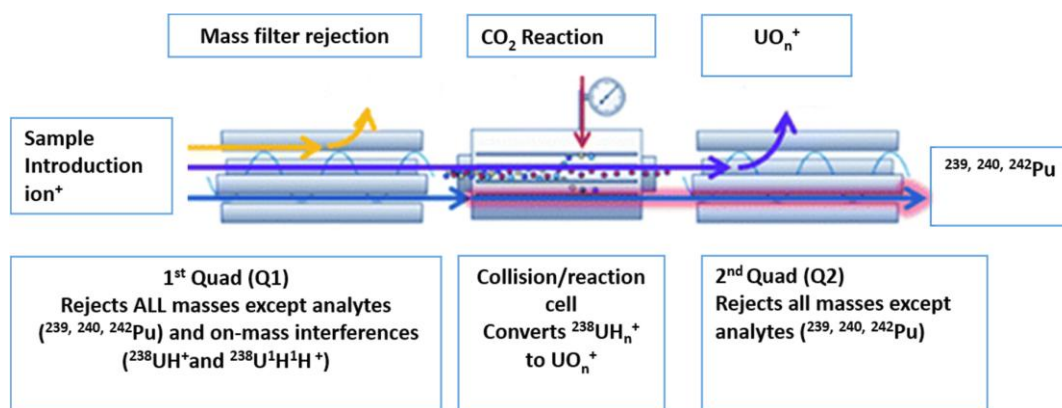


Figure 1. Schematic of ICP-QQQ-MS instrumentation (Balcaen et al., 2013).

### Materials and methods

A suite of different reference materials were employed for method validation; the reference materials analysed were IAEA384, IAEA300, NIST 4353, and in house standards Mayak 2626 and 3516. Yield monitors (1 g of a combined  $^{233}\text{U}$  (28.7 pg/g) and  $^{242}\text{Pu}$  (24.1 pg/g)-tracer) was added to all samples and blanks before digestion. IAEA 384 was transferred to PTFE-tubes using an additional 3x2 ml of concentrated  $\text{HNO}_3$  while 10 ml of  $\text{HNO}_3$  was added directly to the remaining samples.

All samples were left to soak overnight before addition of 4 ml 28 % HF. Digestion was conducted in an ultraclave unit (UltraClave 3, Milestone Ltd) at temperatures up to 260 °C for 20 minutes. After digestion, the samples were transferred to PTFE beakers and left to evaporate to dryness on a sand-bath.

The sample residues were then dissolved, and a single step anion exchange procedure, as described in Wilcken (2007), was employed to remove matrix elements and to reduce the presence of uranium in the sample solutions.

In this work,  $^{240}\text{Pu}/^{239}\text{Pu}$  isotopic ratios were obtained by mass spectrometry measurements using an Agilent 8800 ICP-MS triple quadrupole, as a part of a method development. Pu also reacts to  $\text{PuO}$ , and the method hardware was tuned for the highest possible signal with a 10 ppt  $^{242}\text{Pu}$  solution to maximize the  $\text{UO}_n$  formation and minimize the  $\text{PuO}$  formation. 0.32 mL/min of  $\text{CO}_2$  gave a good signal to noise for Pu vs. UH for 1 µg/L U. The mass balance was set to 93 % (of 260 amu) for best sensitivity of Pu masses. Normally, a quadrupole is most sensitive at mid mass of quad (130 amu for 8800). By setting the mass balance to 93% (242/260), the quadrupole is most sensitive for mass 242.

All measurements were performed with 32 ml/min  $\text{CO}_2$  as a reaction cell gas, causing formation of UO and transferring sample U to mass 254 and 270, while leaving Pu to be measured on-mass without U-interferences.

### Results and discussion

Results from analysis of selected reference materials are shown in table 1. Concentrations and atom ratios were found to be within the confidence intervals of the certified values, except for IAEA-300, likely associated with the low intensities due to low available sample size. The present method permits the reduction of U-interferences during analysis of Pu by ICP-MS, and the reduction in work load during sample pre-treatment.

Table 1. Concentrations and atom ratios in selected reference materials analysed in the present work. \* the available sample size of IAEA 300 was only 0.7 g.

Reference material	$^{239}\text{Pu}$ Bq/kg	$^{240}\text{Pu}$ Bq/kg	$^{239+240}\text{Pu}$ Bq/kg	$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio	Reference
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IAEA 384 98	17.5	107	0.049±0.001	Reference sheet
Fangataufa sediment	CI:85-105 88±5.24	15.1-18.7 17±3	CI:103-110 106±6	0.054±0.009 this work
IAEA 300* Baltic sea sediment	1.8±0.2	1.4±0.2	3.55 CI:3.44-3.65 3.2±0.3	0.22±0.05 Reference sheet this work
NIST4353 Rocky flats soil	9.5±0.4	2.1±0.2	16.8 CI:6-26.8 11.5±0.5	0.056 CI:0.053-0.06 0.059±0.006 Reference sheet this work

## 2.2 Characterization of uranium oxides using XRD and $\mu$ -Raman

Identification of solid phases of uranium compounds is important in many areas, one of them being nuclear forensics. Knowledge about the chemical phase is important since it might give clues, often in combination with other signatures, about the intended use and process history of a sample, and furthermore ultimately about the possible origin of a sample.

XRD and  $\mu$ -Raman were used to identify the chemical phases in uranium oxides. Due to the different penetration depths of the two techniques, they might give complementary information regarding the different compounds present in a uranium sample. Table 2 gives instrument and instrumental settings for the XRD measurements. Figure 2 shows diffractograms of three different uranium oxides (P14112, P14111 and P14110) compared with those of two reference compounds ( $U_3O_8$  and  $UO_2$ ).

Table 2. The XRD instrument used in the characterisation.

Manufacturer and model	Bruker D2 Phaser
Source and wavelength	Monochromatic Cu $k_\alpha$ -source ( $\lambda=1.54060\text{\AA}$ )
Device for reduction of the Cu $k_\beta$ -peaks	Ni-foil
Detector	1-dimensional Lynx Eye
Geometry	Bragg-Brentano $\theta/\theta$
Primary slits	0.2 mm



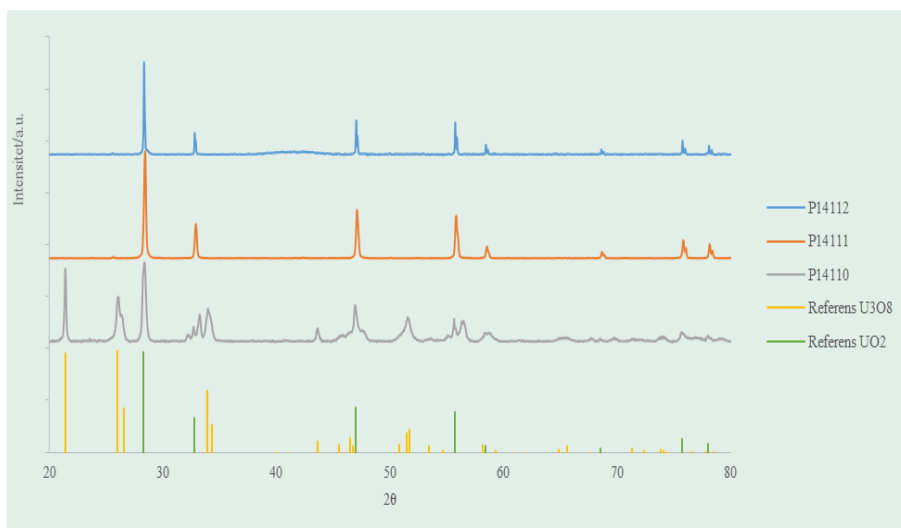


Figure 2. Diffractograms for three uranium samples and two reference compounds.

$\mu$ -Raman will, due to the shorter penetration depth in the sample, give information about the chemical phase present on the surface. In Table 3, the instrument used for the  $\mu$ -Raman measurements is presented. Figure 3 shows  $\mu$ -Raman spectra for the same samples as shown in Figure 1.

Table 3. The  $\mu$ -Raman instrument used in this work.

Manufacturer and model	Horiba-Jobin-Yvon HR 800 UV
Laser wavelength (nm)	514 785
Laser Characteristics (Lasing medium)	Argon ion (514 nm) Diode semi-conductor (785 nm)
Spot size of laser	With x100 objective: $\sim 0.4 \mu\text{m}^2$
Gratings (lines/mm)	300 for 785 nm 600 for 785 nm 1200 for 785 nm 600 for 514 nm 1800 for 514 nm
Spectral range ( $\text{cm}^{-1}$ )	$>4000$ (for 1800 lines/mm) Up to $\sim 3500$ (for 600 lines/mm) Up to $\sim 1700$ (for 300 lines/mm)
Focal distance of the spectrometer (cm)	80
Numerical Aperture (NA)	0.25 for x10 0.45 for x50 long work distance 0.75 for x50 0.9 for x60 water immersion 0.9 for x100 1.25 for x100 oil immersion
Output power (mW)	300 (785 nm) 50 (514 nm)
Slit ( $\mu\text{m}$ )	N/A <sup>a</sup>
Detector and operating temperature ( $^{\circ}\text{C}$ )	Air cooled electron multiplying CCD ( $-70^{\circ}\text{C}$ )
Typical integration time (range)	10 ms to infinity
Objectives	10x, 50x, 50x long work distance, 60x water immersion, 100x, 100x oil immersion

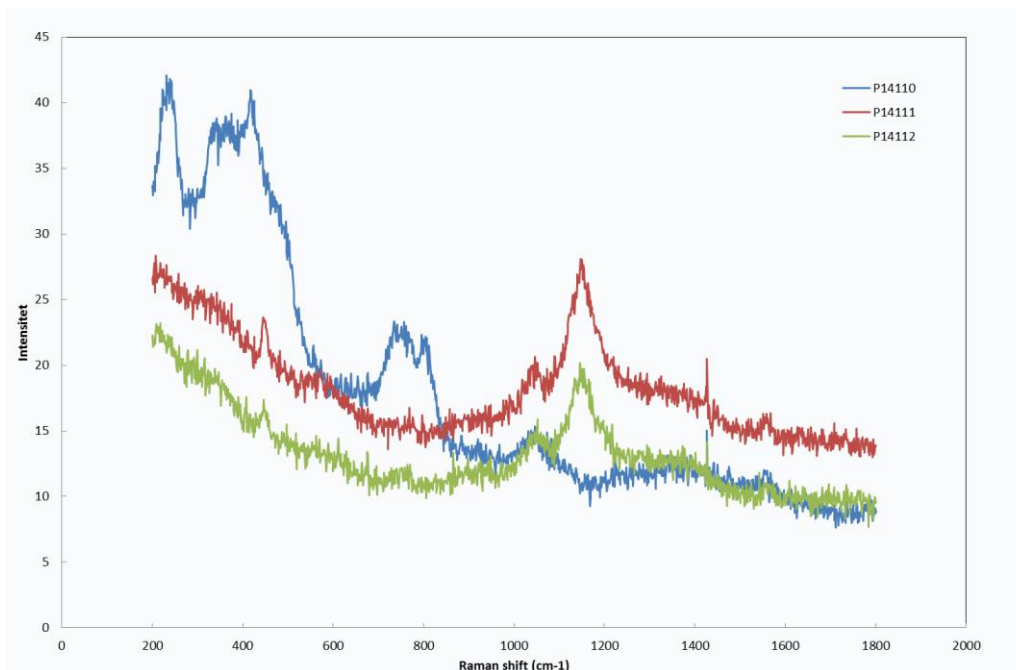


Figure 3.  $\mu$ -Raman spectra for the three uranium samples.

In Figure 2 it can be noted that one of the samples contains an ‘oxidized’ uranium oxide as well as the  $\text{UO}_2$ . However, in combination with Figure 3 it can be concluded that this phase is present on the surface of the sample, i.e. a surface oxidation has occurred and the sample is not completely oxidized to  $\text{U}_3\text{O}_8$ . This is important information since it tells about the history of the sample.

### 2.3 Gamma and x-ray spectrometry with post-processing of coincident signals

Non-destructive gamma spectrometry of unknown samples is an important tool in nuclear forensics. There are a great variety of detector types to be used with various performances in energy resolution, efficiency, mobility and energy span. Traditionally, laboratory based detector systems are made up of high resolution germanium detectors. In order to be able to reduce ambient background from surrounding sources substantial shielding is required. However, even with thick lead-shielding the cosmic muon/neutron component is difficult to reduce to levels which can only be achieved in underground laboratories at great depth (several hundred meters). The effect of the cosmic background component is a generally increase in the background continuum thus raising detection limits for all gamma emitters irrespectively of gamma energy. Another important contributor to the general background is the scattered radiation in the sample itself, surrounding shielding material as well as in the detector. The latter can be reduced by using large peak to Compton units while the scattered photons from the sample and shielding itself depends on geometry, sample size, composition and gamma energy.

An alternative method of reducing background is by active shielding using coincidence techniques. So called anti-compton systems have been designed to reduce the continuous background from scatter in the sample itself and from cosmic radiation provided interaction occurs in both detector systems. These techniques, using hardware coincidence circuitry, have been available for several decades. The major drawback of these systems is that the output data cannot be further manipulated, for instance if it would be discovered that timing properties was incorrectly set during acquisition. During the last 5-10 years focus has turned to acquire time stamped bulk data emitted from the sample and to later perform post processing. In this way coincident signals from several lines emitted by an isotope may be

used, coincidence settings relative to the background count rate may be changed and in general it opens up for a more flexible handling of the data output. Many of the relevant isotopes in nuclear forensics are cascade emitting (eg  $^{60}\text{Co}$ ,  $^{235}\text{U}$  etc) thus enabling coincident techniques to be used.

A LGe system consisting of two planar Ge-detectors with epoxy window was constructed (Fig. 4) in order to enable acquirement of coincident signals from gamma/x-rays. An entirely digital pulse-processing system was used to acquire signals from the two detectors and store output data in a text-file for later processing.



Figure 4. LGe system consisting of two planar Ge-detectors with epoxy window.

First measurements on the detector system have been done using a Co-60 test sample. As the detectors are of low efficiency, the current system does not perform its best on a Co-60 sample which was used for testing. The probability of both gammas being totally absorbed is very thus low. Coincidence effect is somewhat more pronounced in a NBL 103 sample where the Bi-214 609 and 1120 keV peaks emerge.

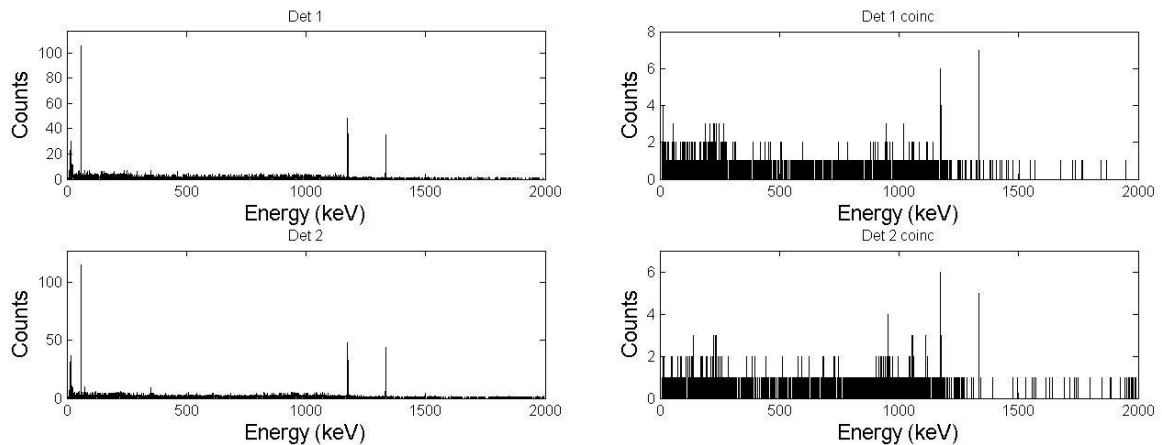


Figure 5. Co-60 source signal on both detectors (left) and the coincidence events on both detectors within 0.3  $\mu\text{s}$  (right).

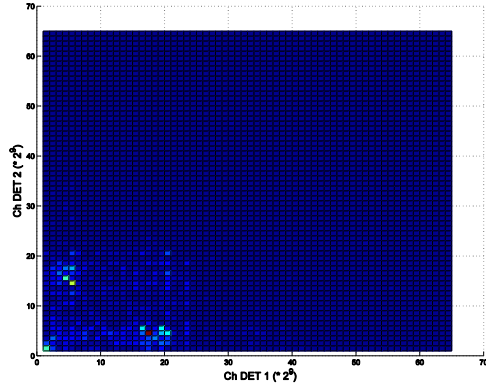


Figure 6. 2D coincidence map shows the events that reached the detectors within the coincidence interval. On x-axis is detector 1 channel number, on the y-axis the detector 2 channel number. Blue areas are zero counts while progressively warmer colors indicate higher count rate, full-energy peaks shown as red.

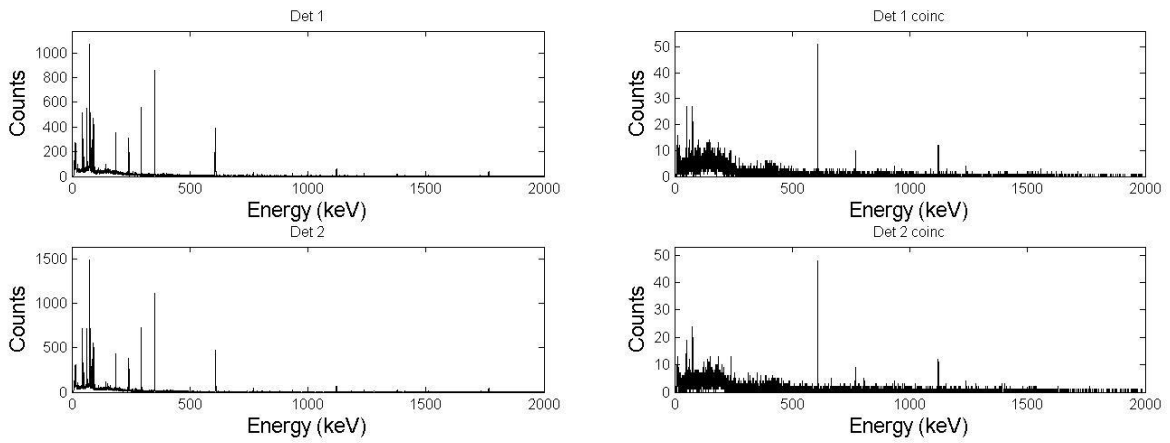


Figure 7. NBL 103 sample total spectra from both detectors (left) and corresponding coincidence spectra (right) showing pronounced Bi-214 609 and 1120 peaks.

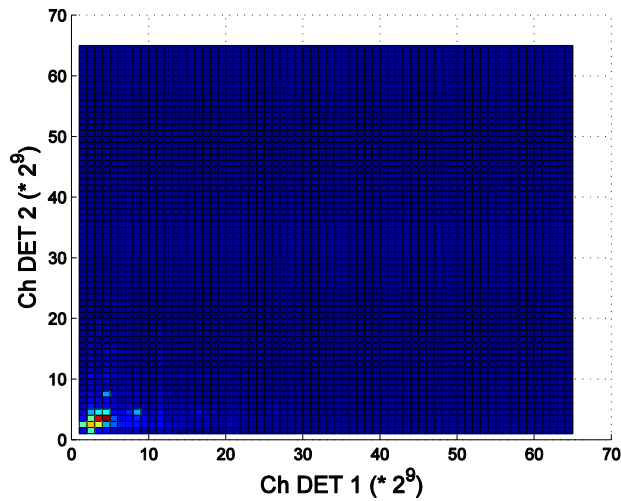


Figure 8. NBL 103 2D representation.

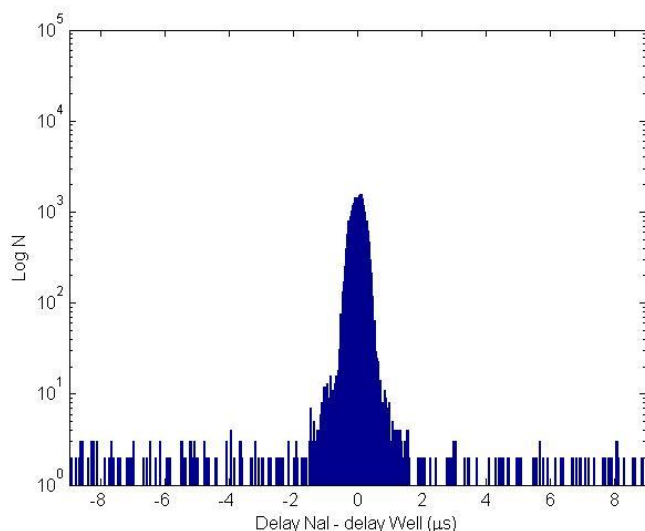


Figure 9. Distribution of coincidences in the NBL 103 sample. Time 0 is when signal is detected in detector 1. The detectors are of similar size so most of the coincidence events occur within 0.3  $\mu$ s.

### 3. Seminar report

The NKS-B seminar on nuclear forensics in Nordic countries was successfully arranged in Oslo, October 5.-6. 2015 with 26 participants from 7 countries and 17 presentations. The participants were mainly members of the NKS organisations and the invited speakers. The invited speakers David Smith, IAEA Nuclear Security, Michael Curry, US State Department, Anne-Laure Fauré, French Alternative Energies and Atomic Energy Commission (CEA), France, Per Reppenhausen Grim, Emergency Preparedness Denmark and Hari Toivonen, HT Nuclear Oy, Finland, contributed significantly to the meeting and recommendations in chapter 4 is largely based on their presentations.

### 4. Analytical recommendations for nuclear forensic purposes in the Nordic countries

Nuclear forensics has received much international attention recently and several important contributions to the literature have been published (Fedchenko, 2015; IAEA, 2015; Moody et al., 2015). According to Fedchenko (2015), nuclear forensic investigations or analyses (NFA) are typically divided into 4 stages: “sample collection and categorization, characterization of the material in the sample, interpretation of the results and reconstruction of the history of the material or an event to which the material relates”. Technical insights into the origins and histories of evidentiary specimens can be obtained through elemental, isotopic, chemical and physical signatures (Moody et al., 2015). A wide array of analytical tools can be used in nuclear forensics. Table 1 lists of many of the applicable techniques and what kind of information the different techniques can provide. The individual techniques can be sorted into three broad categories (Table 1): bulk analysis tools, imaging tools and solid-state speciation techniques. Among the latter category, many are also micro-/nano-analytical imaging tools such as nanometer- and micrometer focused synchrotron radiation based techniques that deserve more attention by the nuclear forensics scientists as they can provide unique information, especially in relation to the development of nanometer resolution techniques.

Table 4. Analytical tools applicable for nuclear forensics (IAEA, 2006; Salbu et al., 2015)

Measurement goal	Technique	Type of information
Survey	Gamma and X-ray spectrometry	Isotopic

Elemental and isotopic bulk or single particle analysis	Chemical assay	Elemental
	Radiochemistry/Radioanalytical counting methods	Isotopic, elemental
	AMS	Isotopic
	TIMS	Isotopic, elemental
	ICP-MS	Isotopic, elemental
	GD-MS	Isotopic, elemental
	XRF	Elemental
	XRD	Crystallography
	GC-MS	Trace organic constituents
	Infrared	Molecular
Imaging	Visual inspection	Macroscopic
	Optical microscopy	Microscopic
	SEM with microanalysis	Structure, phases and elemental composition and distribution
	TEM with microanalysis	Elemental composition and distribution
	Micro focused PIXE	Elemental distributions/ratios
	Nano-SIMS, SIMS	Size distribution and isotope ratios
	LA-ICP-MS, LAMMA	Elemental and isotopic composition and spatial distribution
Solid state speciation	Analytical TEM (transmission electron microscopy)/STEM with microanalysis, electron diffraction, EELS, HAADF	Size distribution of colloids. Element composition, crystalline structure, chemical bonding and Z-contrast imaging
	Nano- and micro focused XRF	Elemental composition and 2D distribution (depth information)
	Confocal $\mu$ -XRF	Elemental composition and 3D distribution
	Nano- and micro focused XANES	Oxidation state (distribution)
	Nano- and micro focused XRD	Nano- and micro focused XRD
	Nano- and micro focused tomography	Spatial distribution of density, elements, oxidation states
	EXAFS	Structure of non-crystalline materials
	Micro-RAMAN (Pointurier and Marie, 2013)	Spatial distribution of molecular species

Abbreviations: AMS: Accelerator Mass Spectrometry; TIMS: thermal ionization mass spectrometry; ICP-MS: inductively coupled plasma mass spectrometry; GD-MS: glow discharge mass spectrometry; XRF: X ray fluorescence analysis; XRD: X ray diffraction analysis; GC-MS: gas chromatography-mass spectrometry; SEM: scanning electron microscopy; TEM: transmission electron microscopy; SIMS: secondary ion mass spectrometry; LA-ICP-MS: Laser Ablation ICP-MS; LAMMA: Laser Microprobe Mass Analyzer; STEM: Scanning Transmission Electron Microscope; EELS: Electron Energy Loss Spectroscopy; HAADF: High Angle Annular Dark Field; XANES: X-ray Absorption Near Edge Structure spectroscopy; EXAFS: Extended X-ray Absorption Fine Structure spectroscopy.

Key challenges in nuclear forensics analysis include (IAEA, 2015):

1. Capabilities to initially detect and categorize exposure from a source of unknown origin, i.e. seized material at a site of incident (which is important in order to guide further investigations).
2. Chain-of-custody procedures and safe handling of contaminated material upon receipt in a laboratory, i.e., well documented analytical procedures for measurement and characterization of the main as well as minor constituents of the material including isotope ratios.

3. Characterize the exposure i.e. nanometre to micrometre sized radioactive particles with respect to composition, size, structure, oxidation state and element/isotope/atom ratios utilizing analytical equipment which is fit-for-purpose.

To characterize the exposure from a source of unknown origin, a variety of techniques such as X-ray spectrometry, and microanalytical techniques e.g., synchrotron radiation based nano and micrometer X-ray techniques can be applied, improving the information that can be obtained if the techniques are implemented in nuclear forensics analysis. However, it is worth mentioning that neither IAEA nor the most influential books recently published on nuclear forensic analysis mentions synchrotron techniques (IAEA, 2015; Moody et al., 2015). Information of elemental composition and isotopic/atom ratios can for instance be obtained by MS techniques (AMS, ICP-MS, TOF-SIMS) while oxidation states and crystallographic structures of the carrying actinide matrix can be determined using TOF-SIMS (Hocking et al., 2013),  $\mu$ -Raman and XRD (see chapter 2.2) or synchrotron radiation based x-ray techniques (Batuk et al., 2015; Lind et al., 2007; Salbu et al., 2003; Salbu et al., 2001).

Other important capabilities are the detection of nuclear material using portable equipment and measurements of impurities in e.g. nuclear material, which might point to production processes etc.

#### **4.1 Recommendations from the seminar participants**

Nuclear forensics is an effective capability for states to criminalize nuclear and other radioactive material out of regulatory control and also to remedy vulnerabilities within nuclear security. David Smith, IAEA, Division on Nuclear Security argued in his talk at the NUFORNOR seminar that in a time of unprecedented change and competing national priorities, a critical challenge for nuclear forensics is sustainability. Thus, it is essential that partner disciplines are enabled to do nuclear forensics tasks. Furthermore, training, research as well as international cooperation such as the Nuclear Forensics International Technical Working Group (ITWG), Global Initiative to Combat Nuclear Terrorism (GICNT) and International Atomic Energy Agency (IAEA) are crucial. Reporting from the IAEA International conference on Advances in Nuclear Forensics, July 7<sup>th</sup>-10<sup>th</sup> 2014 (CN-218), Smith highlighted the outcomes stating that there is 1) continued development of subject matter experts and experienced practitioners, 2) advancement of new analytical tools and methods, 3) sustainment of technical nuclear forensic capabilities once developed and 4) strategic international engagement. The path forward for nuclear forensics should be, according to Smith, to focus on 1) nuclear forensic science, 2) to meet law enforcement requirements, 3) increase confidence of new practitioners and 4) develop consistent implementation. In the context of the present NUFORNOR project, the focus on nuclear science is highly relevant as nuclear forensics science is important for innovation as well as engagement with international partners. Smith listed several scientific advancements in the wake of nuclear forensics:

- rapid and complete radiochemical separations
- precision and accuracy with respect to age dating (e.g.,  $^{230}\text{Th}/^{234}\text{U}$ )
- nanoscale particle characterization of nuclear materials (e.g., SIMS)
- source and origin identification by exploiting halogens, rare earths and minor isotopes
- advanced computer simulations (burn up of spent fuel)
- new nuclear forensic analytical reference materials (e.g.,  $^{229}\text{Th}$ ,  $^{134}\text{Ba}$ )
- link signatures across nuclear fuel cycle to thermodynamics

These examples clearly show the relevance for nuclear forensics to leverage on existing knowledge at nuclear and environmental research institutes. On the other hand, the acceptability of nuclear forensics findings are strengthened by scientific peer review.

Michael Curry of the US State Department gave a very interesting lecture on international nuclear forensics cooperation in which analytical tools were mentioned implicitly in an international capability discussion.

Anne-Laure Fauré from CEA/DAM Ile de Paris gave a very good talk on characterization of nuclear materials with focus on SIMS analysis of U materials. She stressed the need of collaborations between countries and institutes as well as round robin exercises in order to facilitate scientific exchange. Development of reference materials is also important for nuclear forensics. Henrik Ramebäck, FOI, pointed out that LRGS RIID is in need of a spectroscopic reachback capability for a reliable identification of nuclear and other radioactive materials. Furthermore, he stressed that a proper uncertainty estimation is required in gamma spectrometric measurements of nuclear materials and that U isotope determination by means of gamma spectrometry with high efficiency geometries require TCS corrections. Pablo Lebed, CERAD/NMBU presented a promising technique for geolocation by means of rare earth metal elemental ratio measurements on ICP-MS. Brit Salbu and Ole Christian Lind, CERAD/NMBU argued that all aspects of particle characteristics as part of the signature should provide highly relevant information within nuclear forensic. They also presented particle characterization techniques (e.g. synchrotron radiation based x-ray techniques with nm to  $\mu\text{m}$  resolution, see table 1) they are using within radioecology that should be highly useful also in nuclear forensics.

## 6. Conclusions

The Nordic community would benefit from acquiring more knowledge and competence on nuclear forensics and the analytical techniques available within this discipline. Nuclear forensics poses a technically complex challenge both for the scientific and the law enforcement communities and difficulties involved especially for attribution processes, should not be underestimated. In the present project, the seminar “NKS-B seminar on nuclear forensics in Nordic countries” was arranged to provide information on the necessity and suitability of novel analytical techniques within Nordic nuclear forensics as well as exploring possibilities for collaboration across institutions and borders within the Nordic countries. The present report includes recommendations from the seminar participants including suggestions to include new analytical tools. Challenges in nuclear forensics analysis include handling of nanometer to micrometer radioactive particles, availability of the best suitable or required analytical equipment, cross-contamination issues etc. A variety of techniques such as mass spectrometry, gamma/X-ray spectrometry and nanometer- or micrometer focused analytical techniques such as synchrotron radiation based X-ray techniques may together with early detection systems, improve the information that can be obtained if implemented in nuclear forensics analysis.

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## APPENDIX I: Seminar programme

### NKS-B seminar on nuclear forensics in Nordic countries

Hotel Thon Hotel Vika Atrium, Oslo  
October 5<sup>th</sup> and 6<sup>th</sup>, 2015

#### PROGRAMME

##### Day 1

12:30-13:30	<b>Lunch</b>	
13:30	Brit Salbu and Ole Christian Lind	Welcome
13:40-17:00	<b>Session 1: Nuclear forensics - from an international to a Nordic perspective</b>	<b>Chair: Henrik Ramebäck, FOI, Sweden</b>
13:40-14:25	David Smith, IAEA Nuclear Security	Nuclear forensics as part of a Nuclear Security Infrastructure
14:25-15:10	Michael Curry, US State Department	International nuclear forensics cooperation
15:10-15:30	<b>Coffee and fruits</b>	
15:30-16:00	Per Reppenhausen Grim, Emergency Preparedness Denmark	Status of nuclear forensics in Denmark
16:00-16:30	Hari Toivonen, HT Nuclear Oy, Finland	Nuclear security and forensics in Finland
16:30-16:45	Henrik Ramebäck, FOI, Sweden	Research and development at FOI relevant to nuclear forensics
16:45-17:00	Inger-M. Eikermann, NRPA, Norway	Status of nuclear forensics in Norway
17:00-17:15	Gisli Jonsson, GR, Iceland	Nuclear forensics capabilities in Iceland
17:15-17:45	General discussion	<b>Chair: Gisli Jonsson, GR, Iceland</b>
19:00-	<b>Dinner</b>	Louise Restaurant & Bar Aker Brygge - Stranden 3, Oslo

##### DAY 2

08:45	<b>Coffee</b>	
09:00-11:30	<b>Session 2: Historic nuclear events of relevance to nuclear forensics</b>	<b>Chair: Per Roos, DTU, Denmark</b>
09:00-09:30	Brit Salbu	Source terms and release scenarios
09:30-10:00	Jerzy Bartnicki, MET, Norway	Modelling atmospheric dispersion of pollution from nuclear accidents and detonations with the SNAP model
10:00-10:30	Per Roos, DTU, Denmark	Characterisation of the Thule terrestrial Pu-U particles
10:30-11:00	NN, NRPA	International collaboration on nuclear forensics
11:00 -11:30	Discussion	
11:30-12:30	<b>Lunch</b>	
12:30-15:45	<b>Session 3: State-of-the-art and novel techniques</b>	<b>Chair: Inger Eikermann, NRPA, Norway</b>
12:30-13:15	Anne-Laure Fauré, CEA, France	Nuclear forensics at CEA/DAM Ile de France - Focus on SIMS particle analysis
13:15-13:45	Henrik Ramebäck	Precision and accuracy in gamma spectrometric measurements of nuclear materials
13:45-14:15	Ole Christian Lind	Combining speciation and source identification techniques
14:15-14:35	<b>Coffee and fruits</b>	
14:35-14:55	Pablo Lebed	Isotopic signature of selected lanthanides for nuclear activities profiling
15:55-16:15	Ortec	To be announced
16:15-16:35	Michel Ceuppens, Canberra	To be announced
16:35-16:55	<b>General discussion: Future perspectives</b>	<b>Chair: Brit Salbu, NMBU/CERAD, Norway</b>
16:55-17:00	Ole Christian Lind	Closing of the meeting

## **APPENDIX II: List of seminar participants**

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Title	Nuclear forensics within a Nordic context
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Abstract max. 2000 characters	<p>The NKS NUFORNOR project was initiated to strengthen the competence of nuclear forensics and to increase the collaboration within this field in the Nordic countries. The seminar “NKS-B seminar on nuclear forensics in Nordic countries” was arranged in Oslo October 5<sup>th</sup> -6<sup>th</sup>, 2015, with 26 participants from 7 countries and 17 presentations. The goal of the seminar was to provide information on the necessity and suitability of novel analytical techniques within Nordic nuclear forensics as well as exploring possibilities for collaboration across institutions and borders within the Nordic countries. Furthermore, selected analytical techniques such as QQQ-ICPMS, gamma/X-ray spectrometry with post processing of coincident signals as well as micro-RAMAN combined with XRD were tested on relevant samples. Recommendations for nuclear forensics in the Nordic countries include the need to leverage on existing knowledge at nuclear and environmental research institutes and international cooperation. Furthermore, new analytical techniques such as rare earth element elemental ratio analyses, micro-RAMAN and synchrotron based x-ray techniques should be included in the nuclear forensics toolbox. There is also a need of reference materials and proper uncertainty estimation procedures.</p>
Key words	Nuclear, analytical tools, international cooperation, material out of regulatory control, attribution process.