USING MICROPHOTONIC NUCLEAR FORENSICS TO MEDIATE NUCLEAR SECURITY AND NUCLEAR SAFETY

ID #159

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TALK OUTLINE

- Introduction and Definitions
- Situational Analysis (Motivation)
- Microphotonic Nuclear Forensics
  - (With Machine Learning)
- Example Applications
  - (i) LRS: Quantitative imaging of uranium in nuclear aerosol microparticles
  - (ii) LIBS: Trace analysis of fission products in high level nuclear waste
- Conclusion and Prospects

INTRODUCTION (AND DEFINITIONS)

Nuclear safety aims at preventing accidents

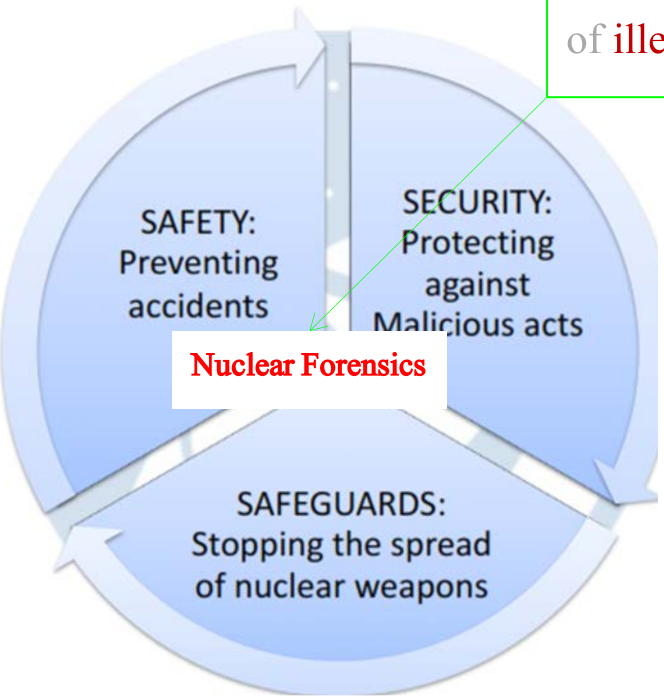
Nuclear security aims at preventing theft of NRM (NSS No. 20)

Nuclear safeguards aims detecting NRM out of statutory control

Nuclear forensics (NF) probes the relation between origin and intended use of NRM: pursued in support of criminal investigation of illegal use, transfer or disposal.

POST 9/11 HIGHLIGHTED THE RISKS THAT NSAs POSE TO INTERNATIONAL SECURITY

There is now a renaissance in nuclear power programs (NPP) in Africa (potential for nuclear proliferation)



Nuclear Forensics

- Powerful tool to respond to cases of nuclear security events
- In itself “deterrence” against nuclear security threats

Safety and Security have a different focus, but a common purpose: protecting human life and health and the environment from the harmful effects of ionizing radiation (a common philosophy of defense in depth)

SITUATIONAL ANALYSIS (MOTIVATION)

Nuclear Security Threats (Examples)

- Sabotage of a nuclear or radiological facility
- Sabotage of a shipment of NRM
- Illicit trafficking of NRM
- Theft of nuclear or radioactive material

- Radiological Dispersal Device (RDD)
- Improvised Nuclear Device (IND)
- Radiological Exposure Device (RED)

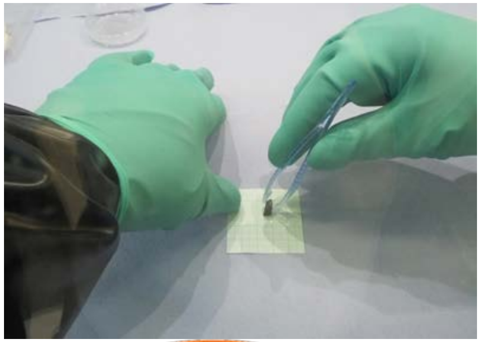
IAEA Incident and Trafficking Database (ITD)

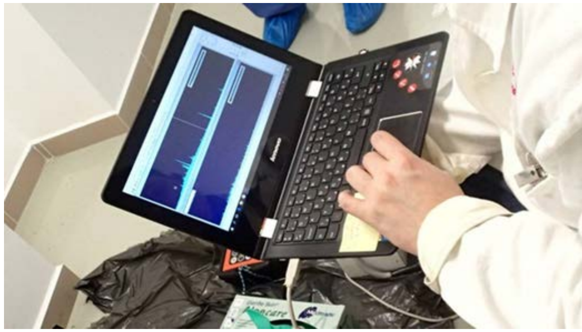
	Improvised Nuclear Device (IND)	Radiological Dispersal Device (RDD)	Radiological Exposure Device (RED)
Required material	Plutonium or Highly Enriched Uranium	Radioactive source and explosives (or other dispersal method)	Radioactive source
Availability of material	Nuclear fuel cycle	Commercially – medical or industrial	Commercially – industrial
Nuclear physics and technology “know-how”	Complex but available	Minimal	Minimal

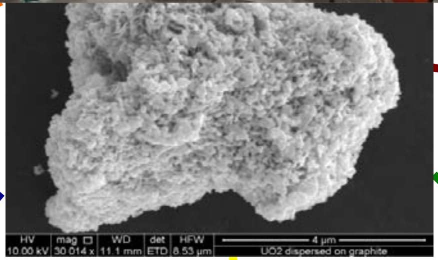
3305 incidents were confirmed to the IAEA from 1993 to 2017

- 8% confirmed or likely act of trafficking, malicious use or scam/fraud
- 28% undetermined act of trafficking or malicious use
- 64% confirmed or likely absence of an act of trafficking or malicious use

As materials move through the **nuclear fuel cycle** the **various NF signatures** are created, modified and destroyed; so each step provides information that can be used to constrain source







What is the material?

Where was it produced?

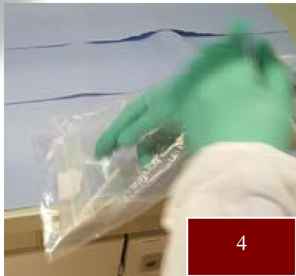
NF Signatures (Examples)

- Isotopic (U, Pu, O, Pb, Sr, Nd, S)
- Chemical (e.g. REE-, trace metals)
- Morphological attributes of NRM.

How was it produced?

Estimated date of production?

What is its intended use?



Current techniques (radiochemical, radiometric) are insufficient for these tasks

MICROPHOTONIC NUCLEAR FORESNICS

Group's research line (NF): development of multimodal spectroanalytical and imaging protocols that marry trace and spatially-resolved chemical, elemental, microstructural, speciation, and morphological information for direct rapid NF analysis and attribution of NRM (we emphasize the science behind detection, analysis, and attribution).

We use laser Raman spectromicroscopy (LRS), laser induced breakdown spectroscopy (LIBS) & laser ablation molecular isotopic spectrometry (LAMIS), energy dispersive X-ray flourescence spectrometry (TXRF) spectrometry.

Microparticle Analysis (Examples)

- Various U oxides
- UO_2F_2 particles
- Pu oxide particles
- U-ore concentrates

Versatility Analytical Attributes

- Speed (e.g. single laser shot)
- Simple operation, systems
- Portable (in *situ*, for *in-field* NF)
- Small sample requirements
- Minimal or no sample preparation

But identity and distribution of the NF signatures does not appear in a straightforward way, owing to the weak signals and interpretative challenges of the high-dimensional data (combine with machine learning).

MACHINE LEARNING -MNF

How to recover the sparse and weak analyte signals (spectra, images) of the trace nuclear forensics (NF) signatures and to explore them in multivariate space, in relation to attribution.

- Doesn't rely on predefined peak shape ('peakless' analysis)
- Works with subtle peaks (weak signals buried in the background)
- Can deal with spectral overlaps and matrix effects robustly
- Applicable even for spectra/ images from different measurement conditions
- Need for spectral resolution of system are significantly relaxed
- Once developed, can perform rapid and stable subsequent analyses

- Greater sensitivity
- Versatility/speed

- Many inputs induce an effect
- Many effects derived from one input

- Data description (exploratory multivariate analysis)
- Classification and discrimination
- Pattern recognition and correlation
- Regression/multivariate calibration

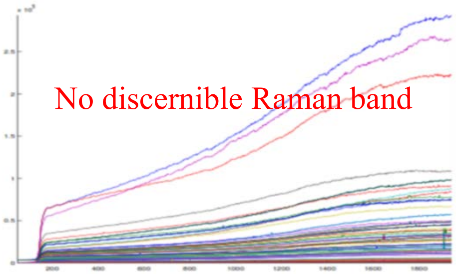
Example 1: Quantitative imaging of U in microparticles

Fine particles from nuclear activities immobilized and incorporated with other actinides in the atmosphere

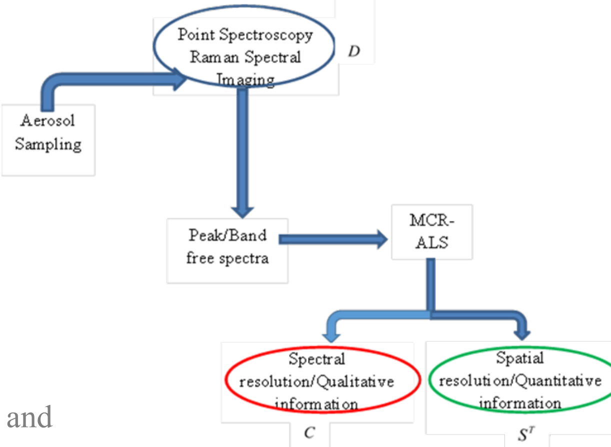
Determine particle formation process

Monitor undeclared nuclear activity,

Respond to anthropogenic releases


No discernible Raman band

3 Raman bands: uranyl chloride (814 and 854 cm^{-1}) and uranyl nitrate (868 cm^{-1}) recovered by MCR-ALS


Aerosol Sampling

Point Spectroscopy Raman Spectral Imaging

D

Peak/Band free spectra

MCR-ALS

Spectral resolution/Qualitative information

C

Spatial resolution/Quantitative information

S^T

8 cites Mrima Hill HBRA (Kenya)

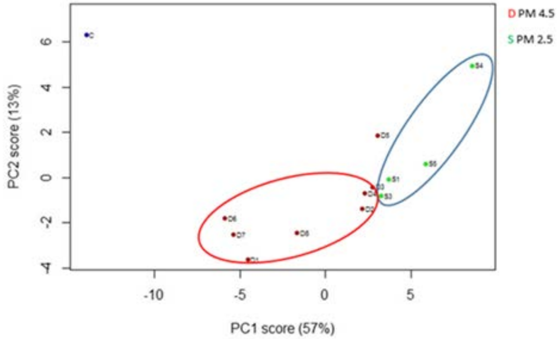
Wabwile J, M., Angeyo H. K., Dehayem-Massop A. K. Exploring Raman Microspectrometry Combined with PCA and MCR-ALS for Size-Resolved Aerosol Analysis for Uranium in a Model Nuclear Atmosphere. Journal of Environm Radioactivity. *In Press.*

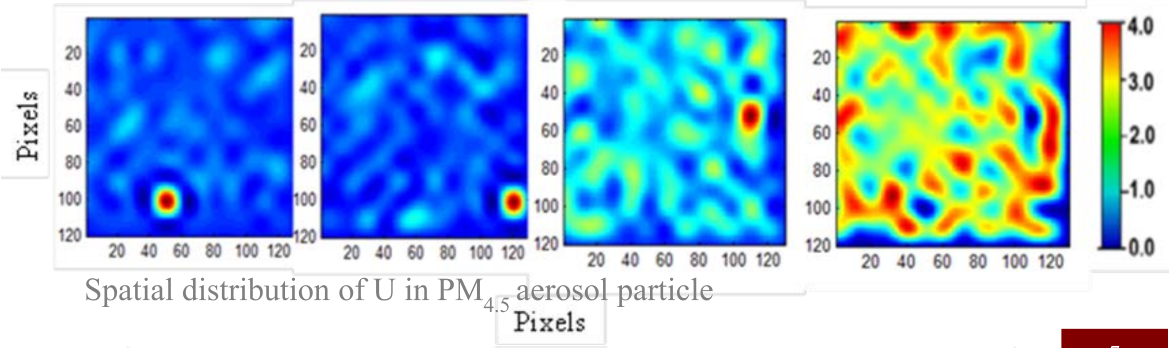
PCA recognized patterns in the aerosols based on their sampling source

Depending on sampling site uranium in the aerosols ranged 50-200 ppb

$PM_{2.5}$ characterized by high sea salt ions (Indian Ocean sea spray).

U more enriched in the 4.5 μm than the 2.5 μm size fraction.



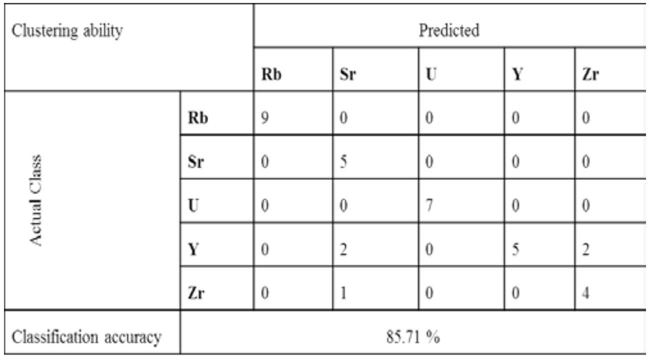


Spatial distribution of U in $PM_{4.5}$ aerosol particle

Example 2: Radiological Crime Scene Management

LIBS analysis and attribution of fission products in high level nuclear waste (HLNW)

- ANN calibration for trace analysis of Y, Sr, Rb, Zr: > 95% accuracy.
- Based on FP PCA and SVM differentiated nuclear from non-nuclear waste.
- Y, Zr, U most responsible for the observed grouping and thus they are powerful NF signatures for HLNW



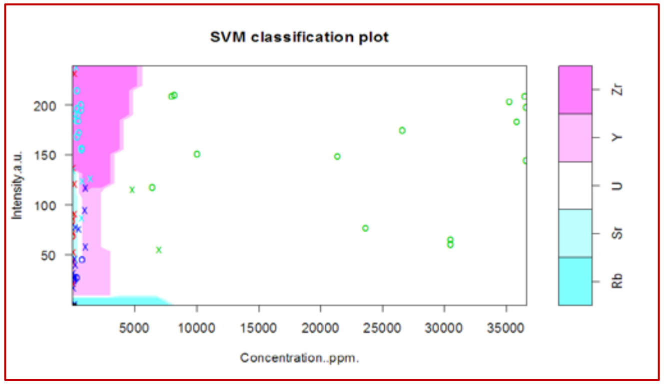
Clustering ability		Predicted				
		Rb	Sr	U	Y	Zr
Actual Class	Rb	9	0	0	0	0
	Sr	0	5	0	0	0
	U	0	0	7	0	0
	Y	0	2	0	5	2
	Zr	0	1	0	0	4
Classification accuracy		85.71 %				

ANALYZED

Vitrified glass (∅3 mm)

HLW Nuclear powders

HLW liquid (2 μL)

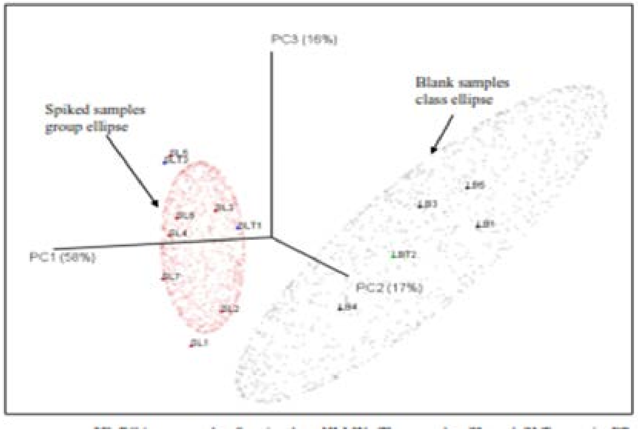


Application Examples

Post-detonation debris

Explosion powders

Liquid drops from RCS



Onkangi J. N., Angeyo K. H. Exploring Machine Learning Enabled LIBS Towards Forensic Trace Attributive Analysis of Fission Products in Surrogate High Level Nuclear Waste. Journal of Applied Spectroscopy. *In Press.*

CONCLUSIONS AND PROSPECTS

- Proof-of-concept for a ML-enabled LIBS methodology for direct rapid analysis and attribution of nuclear FP in HLNW especially in glass debris and mL liquid droplets.

- Raman microspectrometry- MCR-ALS has utility for quantitative spatial distribution of U in individual aerosols: a powerful tool to monitor undeclared nuclear activity, respond to anthropogenic releases, and analyze NRM at radiological crime scenes.

- Leveraging analytical spectroscopy and /imaging with machine learning increases the range and complexity of material analysis challenges that can be realized in NF.

- NF is essential in achieving an integrated approach to nuclear safety and security

Thank you for your attention

ACKNOWLEDGEMENTS

- Research Line
- TWAS
- ISP (Sweden)
- INSEN
- IAEA: CRP-J013
- ACP Org Committee