A high-resolution laboratory-based $\beta$-$\gamma$ coincidence spectrometry system for radioxenon measurement

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T3.2-482
• The UK CTBT Radionuclide Laboratory (GBL15) is certified for Particulate and Noble Gas measurements as part of the International Monitoring System (IMS)

• To deliver the Noble Gas measurements, a SAUNA II IMS Lab system is used for noble gas re-measurements to quantify $^{133}\text{Xe}$, $^{135}\text{Xe}$, $^{131m}\text{Xe}$ and $^{133m}\text{Xe}$

• GBL15 has the remit to research new detection technologies to improve detection sensitivity and accuracy

• A PIPSBox detector has been configured for coincidence measurements with multiple high-purity germanium (HPGe) detectors to evaluate its performance as an option for a future operational laboratory system

• This work looks to determine the **optimal detection limits** achievable for this type of system

• This work is in collaboration with scientists from the University of Surrey and the National Physical Laboratory (NPL)
System Overview

- PIPSBox detector with 2x Mirion 6530 carbon-window BEGe detectors
- Acquisition data collected in time-stamped list-mode for each detector (2xPIPS, 2xHPGe) using custom acquisition software
- Data processed using C++/ROOT custom tools
- Advanced coincidence post-processing in Python/ROOT to generate spectral projections
- **Electron-photon coincidences are combined from all four gain-matched detectors to create a near-4π detector geometry and maximise the detection efficiency**

Fig. 1. AWE PIPSBox-HPGe research detector photographs, with detector cables and gas lines
Measurement & Analysis Overview

- Spike samples prepared by Seibersdorf Laboratories containing $^{133}$Xe, $^{131m}$Xe, $^{133m}$Xe and $^{135}$Xe and a separate sample with pure $^{131m}$Xe
- Gas injected to the PIPSBox and quantified by HPGe measurements of the gas vial
- Perform acquisition and archive list-mode data
- Determine $4\pi$ detection efficiencies
- Extract coincidence projections and generate energy-gated-coincidence summed spectra
- Calculate the MDAs

Fig. 2. Glass vial containing radioxenon sample prepared by Seibersdorf laboratories, received by GBL15.
Fig. 3. electron-photon coincidence matrix from a measurement of $^{131m}$Xe (with small $^{133}$Xe contamination) and inset: zoomed to the X-ray region. Each photon signal energy is used as a gate to project an electron spectrum (see right).

Straggled/backscattered conversion electrons. Should we clean out this signal or make use of it?
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Fig. 4. Summing individual projections to generate a summed projected coincidence spectrum for $^{131m}$Xe
What about when we have a mixed xenon sample? Does this really work?

Can we generate ‘optimised’ spectra for each signal, using the different X-ray energies of Cs and Xe?

Fig. 5. Electron-photon coincidence matrix for a radioxenon sample containing $^{133}\text{Xe}$, $^{135}\text{Xe}$, $^{131m}\text{Xe}$ and $^{133m}\text{Xe}$
Gating on the Xe X Kα gives a $^{133}$Xe β- continuum 20x lower than that in the Cs X Kα. Most of the contribution is from scattered conversion electrons.

Fig. 6. Coincidence matrix and projected spectra using the X(Kα) energy gate for Cs and Xe.
(Same can be applied to $K_{\beta}$ X rays)

Fig. 7. Coincidence matrix and projected spectra using the $X(K_{\beta})$ energy gate for Cs and Xe
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### Total X-ray gate: 20-40 keV (a low resolution photon spectrometer)

More selective signals means less interference.

Summing projections means greater detection efficiency.

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**Fig. 8. Optimising the signal by summing spectra from multiple photon energy gates**

- **Cs X $K_{\alpha+\beta}$**
- **Xe X $K_{\alpha+\beta}$**

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**Putting an End to Nuclear Explosions**

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- $^{133}$Xe is prevalent in the atmosphere due to civil nuclear processes contributing to the global radioxenon background, and high-activity $^{133}$Xe samples are often measured on the IMS.

- $^{131m}$Xe is an important radionuclide - because the 3-isotope ratio plots do NOT allow for clear discrimination between a nuclear explosion and a civil source in many cases.

- On the 4-isotope ratio plot, $^{133m}$Xe/$^{131m}$Xe is the most effective way to determine the possible source 'type' from IMS radioxenon measurements.

- High-resolution spectroscopy means the effect from $^{133}$Xe on the detection limit of $^{131m}$Xe and $^{133m}$Xe is reduced.

- Both metastable isomers are more readily detected using $\beta$-$\gamma$ coincidence, rather than $\gamma$-singles.

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### Table: Isotope 4π MDA (mBq)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>4π MDA (mBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}$Xe</td>
<td>1.0</td>
</tr>
<tr>
<td>$^{131m}$Xe</td>
<td>1.0</td>
</tr>
<tr>
<td>$^{133m}$Xe</td>
<td>1.0</td>
</tr>
<tr>
<td>$^{135}$Xe</td>
<td>4.0</td>
</tr>
</tbody>
</table>

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**Fig. 9.** Detection limit (critical limit - $L_2$) of $^{131m}$Xe as a function of the $^{133}$Xe measured activity concentration.

**Fig. 10.** 4-isotope ratio plot showing the civil nuclear processes domain and explosion domain.

See Kalinowski et al. for more information on the 4-isotope ratio plot. [https://doi.org/10.1007/s00024-009-0032-1](https://doi.org/10.1007/s00024-009-0032-1)

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• Improved sensitivity of $^{131m}$Xe in real/environmental samples can enhance our dataset during an event of interest. Re-measurement at a laboratory could IMPROVE the sensitivity compared to the station, for some isotopes. With delayed re-measurements, perhaps the laboratories can detect isotopes that were not detected in the station measurement…

Fig. 9. Detection limit (critical limit - $L_c$) of $^{131m}$Xe as a function of the $^{133}$Xe measured activity concentration

Fig. 10. 4-isotope ratio plot showing the civil nuclear domain and explosion domain.
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Electron Energy Resolution

Photon Energy Resolution

High-resolution photon detectors provide the signal discrimination required (Cs X-rays / Xe X-rays)

High-resolution electron detectors improve discrimination of $^{131m}$Xe and $^{133m}$Xe but this is not the most important interference

Low-resolution electron detectors are more efficient

Beginning GEANT4 simulations of HPGe+Plastic scintillator coincidence setup

New measurements with SAUNA/Xe-I beta-cell and GBL15 HPGe detectors

Current GBL15 system & most IMS-type systems

New measurements have recently been published:
Qi Li et al. Nuclear Inst. And Methods in Physics Research, A 988 (2021) 164939
Conclusions and Future Work

- Detector geometry has been optimised to improve detection efficiency by enclosing the PIPSBox with two HPGe detectors.
- High-resolution β-γ coincidence spectrometry can improve sensitivity to metastable isomers $^{131m}\text{Xe}$ & $^{133m}\text{Xe}$ with a high background signal of $^{133}\text{Xe}$, when compared to a NaI(Tl)+Plastic set up.
- High-resolution γ-ray detector means it is possible to resolve Cs and Xe X-rays and create separate projections.
- Signals from $K_\alpha$ and $K_\beta$ can be used selectively and the projections summed.
- PIPSBox geometry is excellent for getting near-4π geometry.
- More work required to determine whether the drop-off in detection efficiency is worth the enhanced electron energy resolution – Testing plastic+HPGe system with measurements and GEANT4 simulations.
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Thanks to those that have contributed to this project:

Marc Abilama (NPL), Rob Shearman (NPL), John McLarty (AWE), Graham Galvin (AWE), Phill Scivier (AWE)

Please visit the corresponding poster:

*Measurement of gaseous fission products on an electron-photon coincidence detector system*

(P3.1-485)