

Analysis of Radioxenon Detections in the UK

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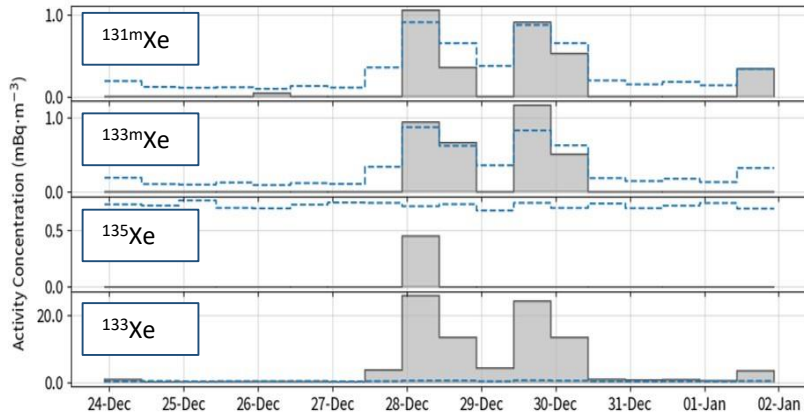
Fig. 1. SAUNA II IMS Lab system at the UK CTBT Noble Gas Laboratory (GBL15), based at AWE Aldermaston

An IMS-like noble gas system is in operation at AWE (Aldermaston, UK) and can collect and measure the radioxenon content in environmental air samples. When operated in this mode, data produced is analysed at the UK National Data Centre (NDC) as part of the in-house radionuclide (RN) analysis pipeline. This work discusses a number of significant detection events analysed using the operational system deployed at the UK NDC, which includes atmospheric transport simulations and a real-time stack-monitoring data feed from a nearby medical isotope production facility (MIPF) in Belgium. A comparison of the expected radionuclide contributions with measured detections is presented, including a comparison of the isotopic ratios for the radioxenon isotopes of interest (^{133}Xe , $^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$, ^{135}Xe).

Two modes of operation:

LAB Mode: For the re-analysis of samples from IMS stations.
IMS Mode: For sampling, processing and measuring samples collected from the local air, operated on an ad-hoc basis and data processed at the UK NDC

The SAUNA system was operated in IMS mode on an ad-hoc basis throughout 2020. High activity concentration, multi-isotope detections have been recorded and investigated. Fig. 2 shows the time series for the first set of detections (Period 1), which arrived in two distinct 'plumes'. All four isotopes of interest were detected in the first plume. The UK NDC RN analysis pipeline flagged several detections for review in the database and the results of the investigation are presented here.



Period	Period start	Period end
1	24-Dec-2019	02-Jan-2020
2	20-Mar-2020	25-Mar-2020
3	01-Apr-2020	15-Apr-2020
4	16-Apr-2020	30-Apr-2020

Fig. 2. Time series activity concentration results (grey histogram) and minimum detectable concentrations (MDC) (dashed blue line) for four radionuclides measured at GBL15 during period 1

The first piece of work is to investigate a possible source region by considering the meteorological conditions around the time of the key detections, and which nuclear facilities may be located such that their emissions could contribute to the detections at GBL15.

There are dozens of reactor facilities in the region, including nuclear power plants (NPPs) and medical isotope production facilities (MIPFs). The first effort involves trajectory models from HYSPLIT – a low-fidelity but fast process. This indicates the source region may include IRE (Belgian MIPF).

Fig. 3. Map of the UK and parts of mainland Europe, showing GBL15 (triangle) and IRE, Belgium (red star), UK NPPs (circle), French NPPs (square), Dutch NPP (yellow plus) Dutch MIPF (red plus) and German NPPs (cross).

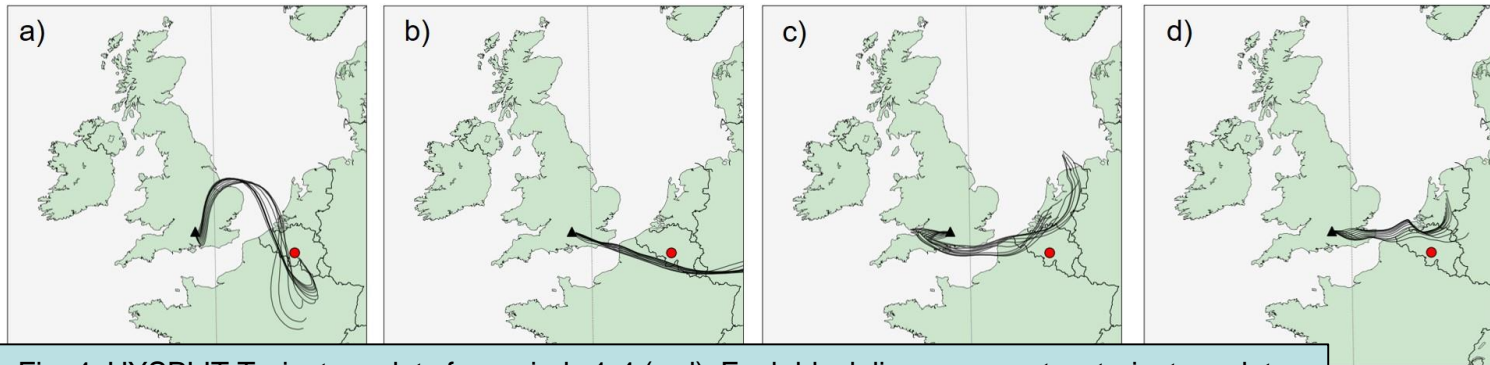
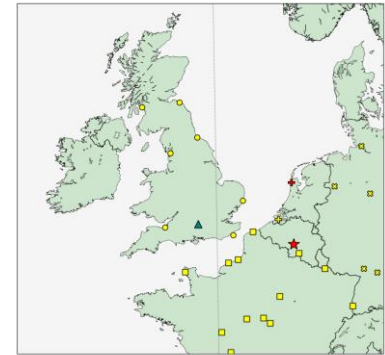


Fig. 4. HYSPLIT Trajectory plots for periods 1-4 (a-d). Each black line represents a trajectory plot for a 1-hour period of the collection. Red circle is IRE MIPF in Belgium

Could the detections be due to emissions from IRE?
 To test this hypothesis, we consider the results of forward atmospheric transport and dispersion (ATDM) simulations from IRE....

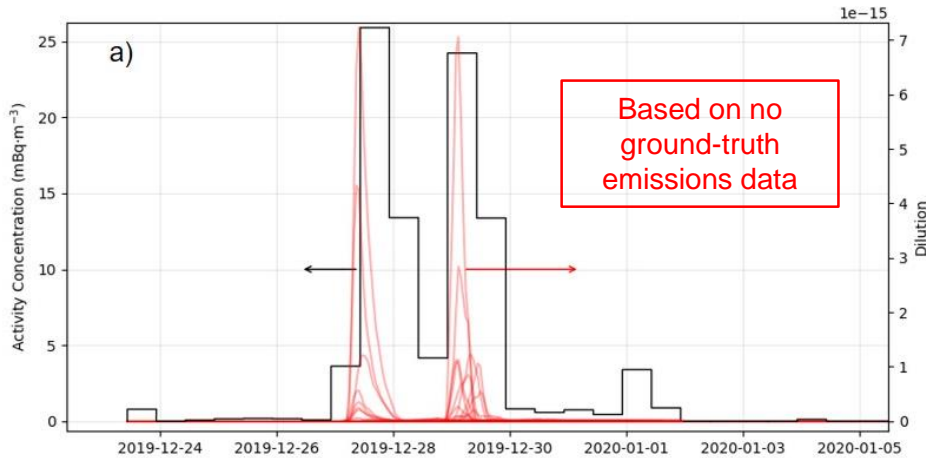


Fig. 5. Measured (**black**) ^{133}Xe activity concentrations and simulated IRE dilution factors (**red**) at GBL15 during the first period. Each simulated contribution is based on a one-hour emission with an emission rate of 1 Bq/hr.

Simulations vs. measurements (without STAX data)

Fig.5 shows the comparison of **simulated contribution** from IRE with the measured activity concentration (A_c), based on no emissions data.

There is clearly a good match in time between the arrival of simulated contributions from IRE and the detections at GBL15.

Simulation details: HYSPLIT; 0.25° GFS met-data; 0.125°, 1 hr sampling resolution; 100,000 particles/hour; 30 days simulation

The UK NDC has access to stack-monitoring data via STAX (see Poster by Auer et al T2.4-211).
At the time of this study, data was available from IRE.

Can the STAX emissions data be used to verify the detections by applying the emission magnitude to estimate the GBL15 activity concentrations...?

STAX DATA (IRE)

Can we use STAX data to explain the detections in the UK?

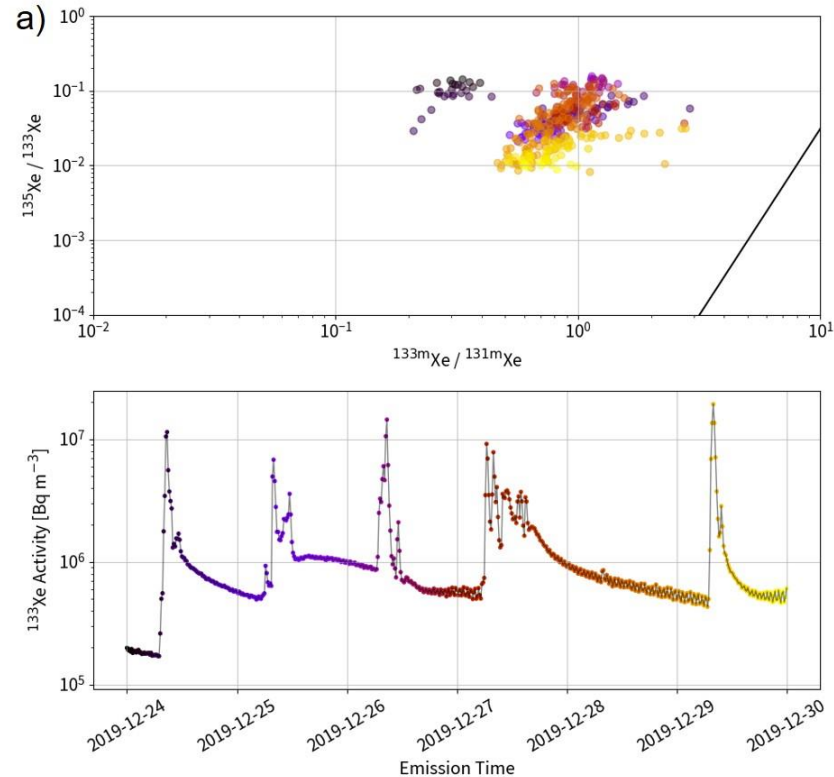
UK NDC STAX Pipeline:

1. STAX data is downloaded at the UK NDC
2. Spectral data files are converted to .CNF files
3. Analysed using the GBL15 Analysis Python code
4. Spectra archived and data stored in a database

Fig. 6 shows a section of data from the UK NDC STAX database, showing the ¹³³Xe activity concentration time series and 4-isotope plot. The data is colour-mapped to show the evolution of the ratios with time.

This data is used to generate input data for HYSPLIT simulations.

Fig. 6. STAX profile and ratio analysis, showing the evolution in the 4-isotope ratio over time for dates in December 2019. The black line on the 4-isotope plot represents the discrimination line (see Kalinowski *et al.*).



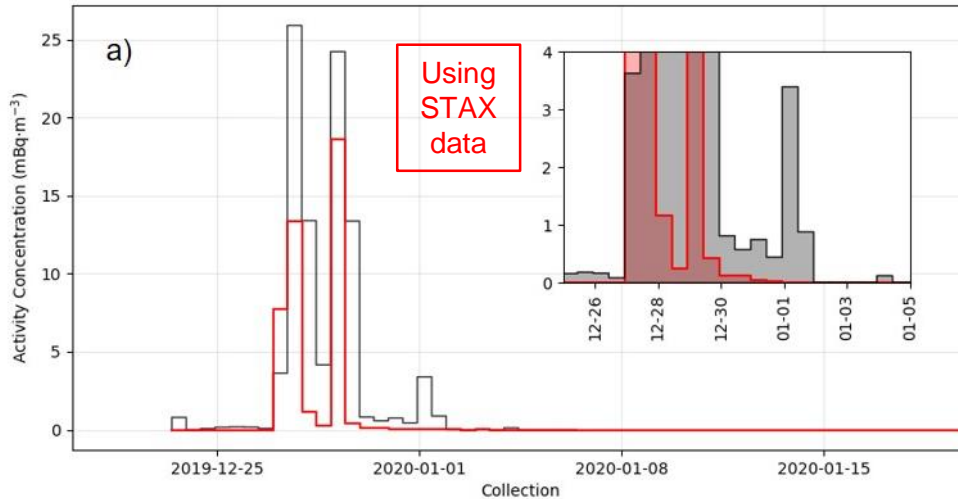


Fig. 7. Comparison of measured (**black**) and simulated (**red**) ¹³³Xe activity concentration at GBL15 for period 1. Inset shows a zoomed and shaded view of the same data

Simulations vs. measurements (with STAX data)

Fig 7. shows the comparison of simulated ¹³³Xe A_c with the measured A_c

The simulated activity concentrations agree well with the measured, but generally underestimate the measured activity concentrations. The final plume (01-01) is not predicted at all.

This is a real-world example of using STAX data to calculate the effect of a MIPF on an IMS-like system in order to determine the possible source of radioxenon.

The ATDM introduces significant (and uncalculated) uncertainty to the estimation.

How do the isotopic activity ratios compare between the STAX-measured emissions data, and the measured ratios at GBL15....?

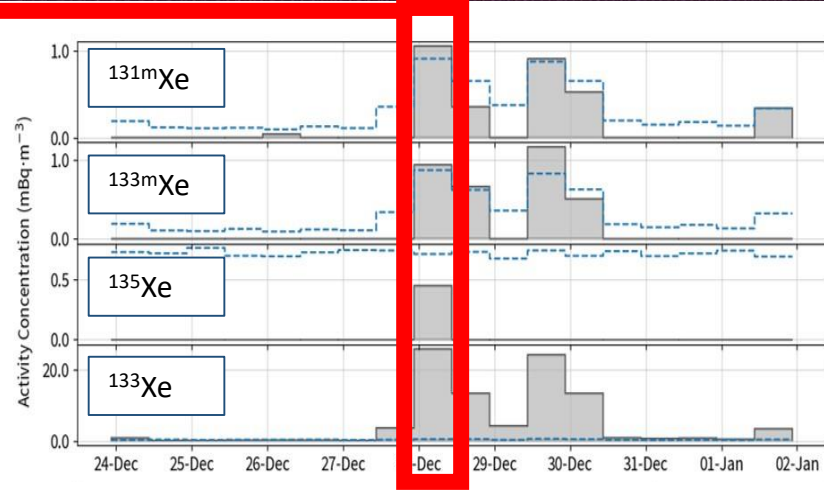
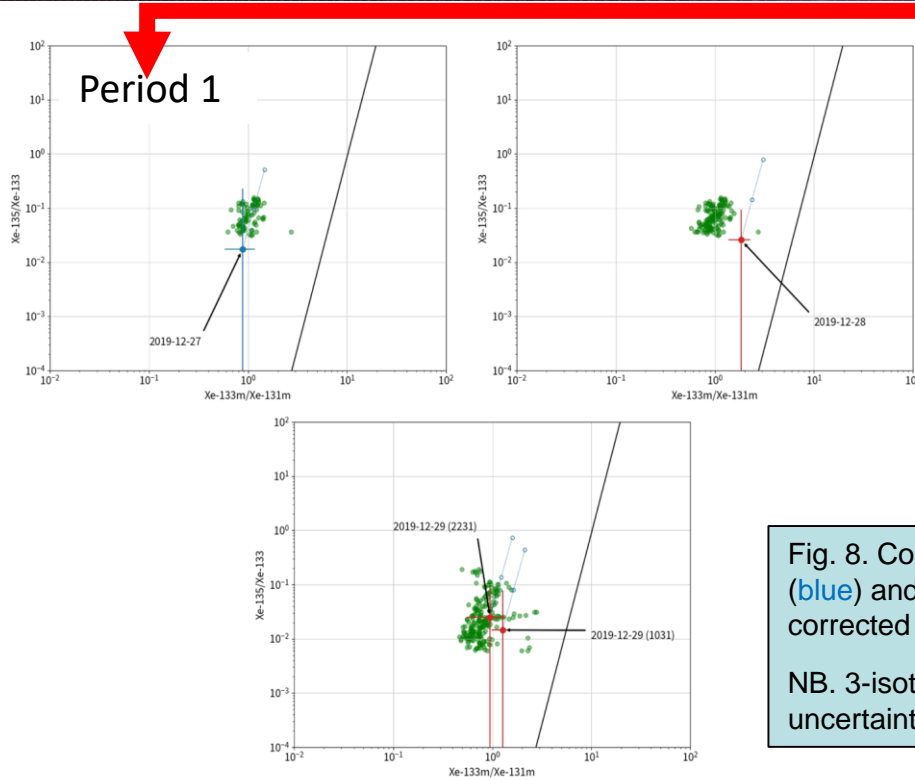


Fig. 8. Comparison of STAX ratios (green) with measured 4-isotope ratios (blue) and 3-isotope ratios (red). The blue lines represent the decay-corrected measurement for 24 & 48 hours.

NB. 3-isotope ratios are calculated using one L_c measurement and the uncertainty lines are expanded to show this

- Multiple detections at AWE, containing mixed isotopes
- Preliminary ATM indicates a possible source region includes Belgium
- Belgian MIPF (Institute for Radioelements - IRE) provide stack emissions data via the STAX programme
- STAX data used here can explain the detections at AWE, which agree well in time and magnitude for ^{133}Xe
- Isotopic activity ratio analysis indicates the detected ratios agree well with the stack ratios.
- The simulations generally underestimate the measured activities, which may indicate more complex ATDM calculations are required, or that there may be another (unknown) source contributing to the detections.
- RN event analysis relies heavily on ATDM simulations. Further research into this area can improve the Community's capability to characterise and understand the effects of the global radioxenon background on the IMS.

More detail on the detections presented in this work (and others) can be found in the corresponding Journal of Environmental Radioactivity article: <https://doi.org/10.1016/j.jenvrad.2021.106629>