



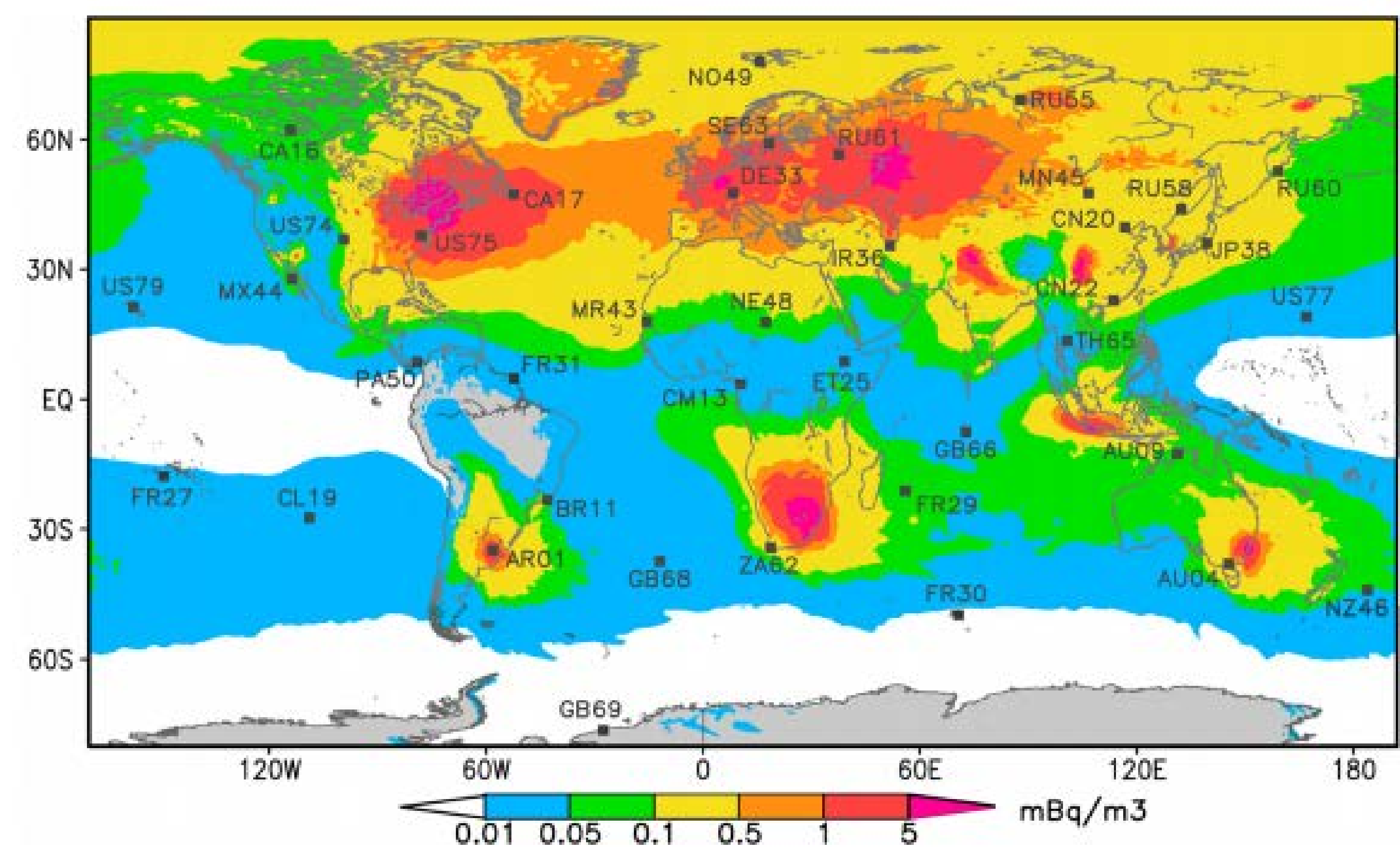
The PTS and the international community have expressed interest in the ability to obtain xenon samples using the output of industrial oxygen plants, which could be three orders of magnitude or more larger than xenon samples currently provided by International Monitoring System (IMS) stations. Precise characterization of regional backgrounds is a motivation for very large volume samples and the enhanced sensitivity they could provide. Such samples would be too large for measurement in the existing detection systems and, as such, a new optimized method for analysis needs to be investigated. For many years the Pacific Northwest National Laboratory (PNNL) has been developing ultra-low-background internal-source gas proportional counters (ULBPCs) for use in a variety of applications; these ULBPCs function as beta spectrometers and have the capacity to analyze significantly larger gas samples than the traditional detectors used in the IMS and the potential to offer significantly improved sensitivity. PNNL has investigated the use of ULBPCs for measuring large-volume radionuclide samples to assess optimal performance parameters (e.g. operating pressure, count gas blend, etc.), sensitivity levels, and ability to discriminate multiple radionuclide isotopes. Results from these investigations will be presented.

Goals and Objectives

- Determine if the recently-developed Pacific Northwest National Laboratory (PNNL) ultra-low-background proportional counter (ULBPC) can enable measurements of worldwide radionuclide backgrounds by using large-volume xenon (LVXe) samples.
- This work focused on the following questions:
 - What is the largest volume of xenon that can be measured in an ULBPC?
 - What spectroscopic and background performance is possible with a LVXe gas load?
 - Can methods be developed for discriminating multiple radionuclide isotopes in a large sample measured via beta spectroscopy?

Motivation

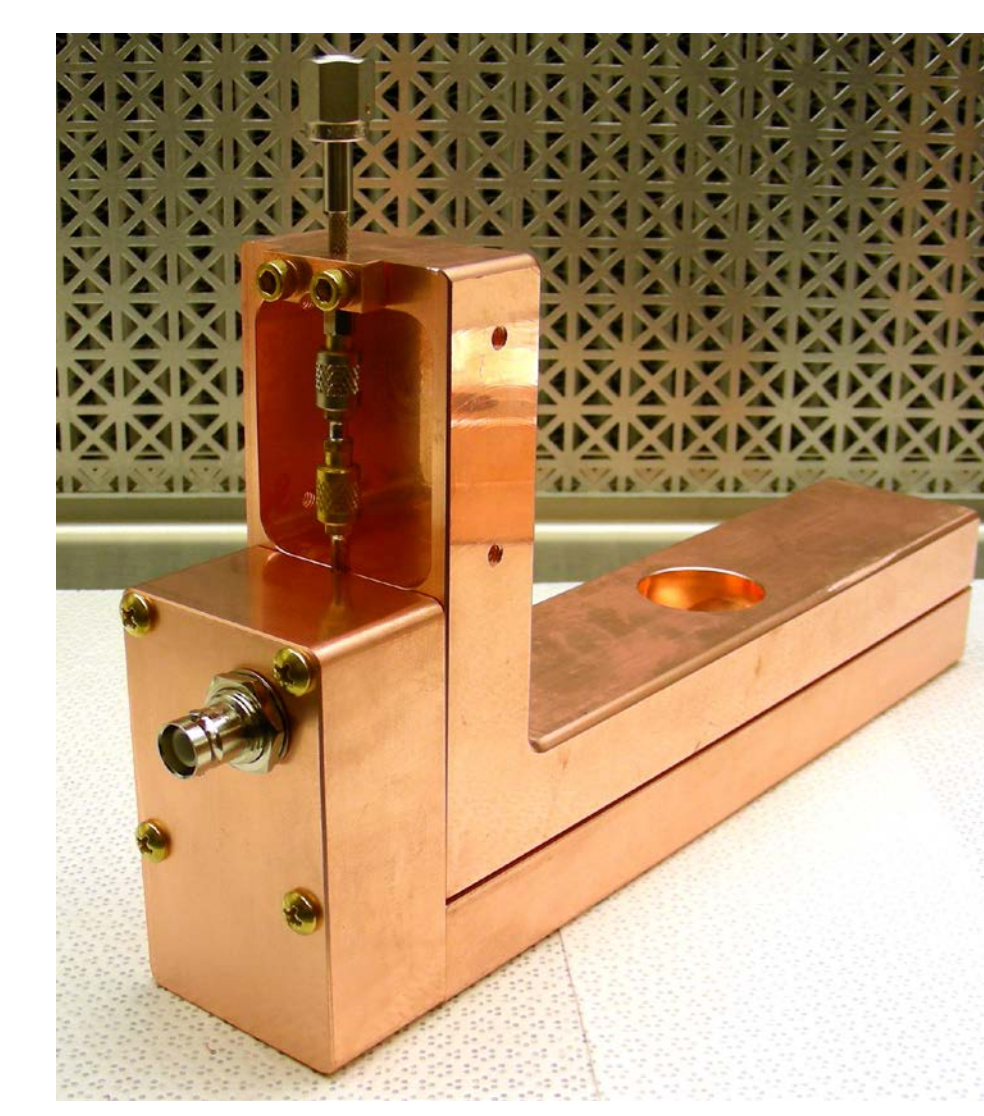
- The baseline sensitivity level for a LVXe measurement is the ability to routinely quantify the background radionuclide concentrations around the world.



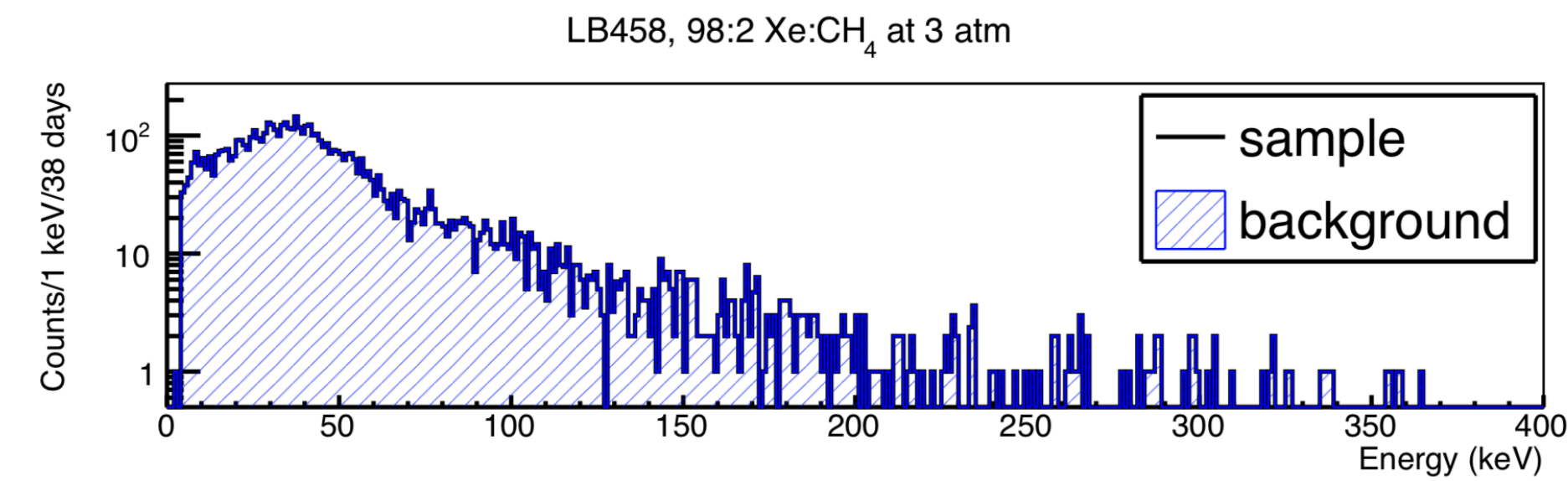
Annual average activity concentrations of ^{133}Xe calculated from two years of simulated data, at ground level. Note that the radionuclide spikes observed from anthropogenic sources results in an average elevation above the natural background levels of **0.01 to 0.05 mBq/m³**, but there are days when the local concentration is consistent with natural levels. [1]

Ultra-Low-Background Proportional Counter (ULBPC) Measurements

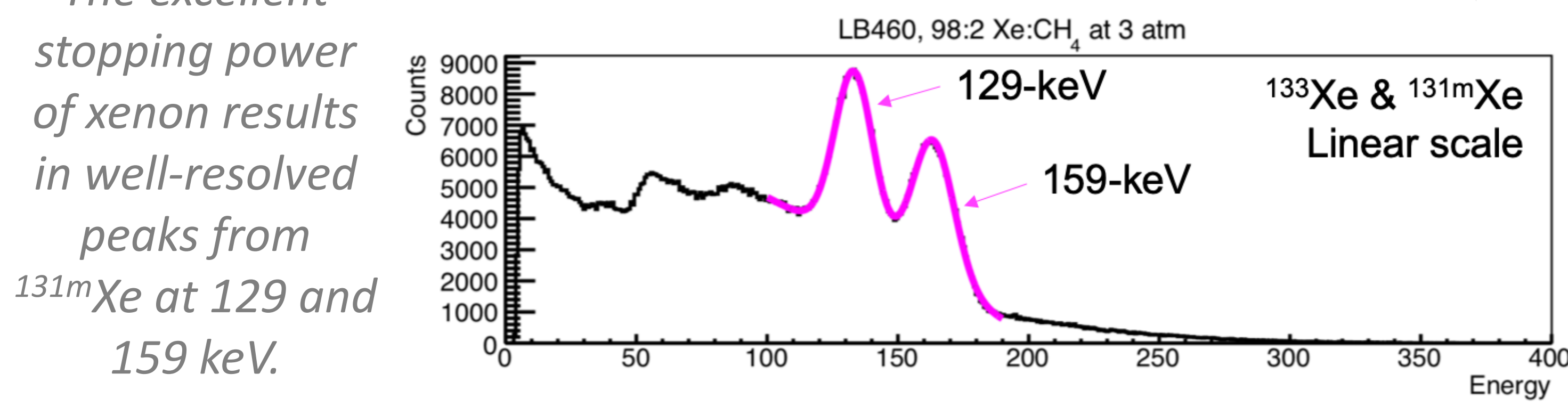
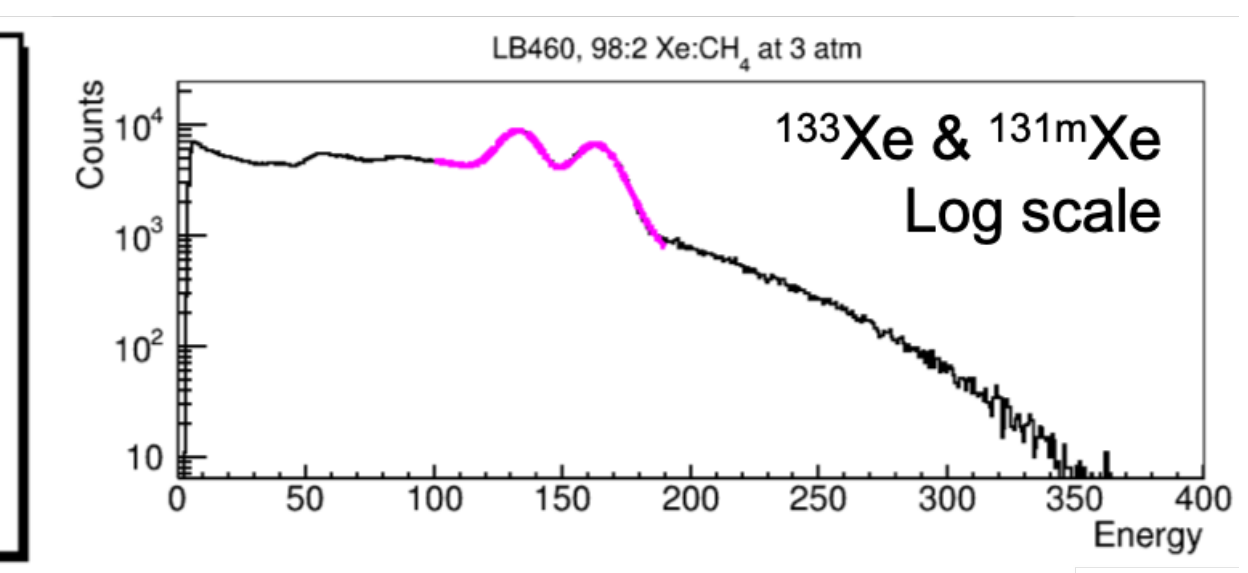
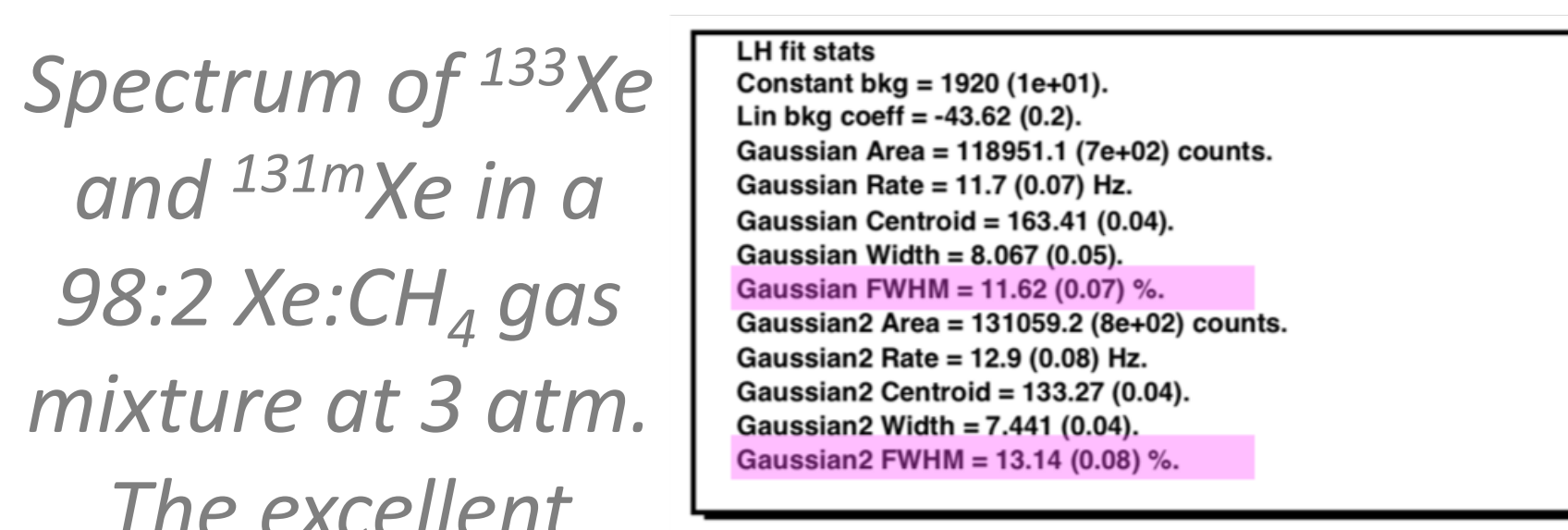
- PNNL has developed an ultra-low-background measurement capability utilizing 100-cc ULBPC and the PNNL Shallow Underground Laboratory (SUL) [2,3].
- Previous measurements of radionuclide in these detectors used a small spike of radionuclide activity (<1cc) on top of a carrier blend of P10 (90% Ar, 10% CH₄) at pressures ranging from 3 to 10 atm.
- A parametric study was performed to evaluate various gas blends including mixtures of P10 and Xe, along with Xe containing a small percentage of CH₄.
 - Gas blends were evaluated for resolution, dead time, and pulse pile-up.
 - A blend of 98:2 Xe:CH₄ was chosen and detection sensitivity evaluated with ^{133}Xe + $^{131\text{m}}\text{Xe}$ and background measurements.



The ULBPC, with 100-cc internal gas volume.



Background spectrum from 100-cc ULBPC with 3 atm of 98% Xe and 2% CH₄.



- Using the half-life for ^{133}Xe and the measured background rate of 158 counts per day in the ROI, the minimum detectable activity is calculated to be **2 mBq** for a 12-hour measurement.

Sensitivity Analysis

- Inputs:
 - Consider a background sensitivity goal of 0.01 mBq/m³
 - Constrain operating pressure of ULBPC to 3 atm
 - Operating gas composition of 98% Xe and 2% CH₄
 - Nominal internal volume of a ULBPC is 100 cm³
 - Detection efficiency is expected to be approximately 88% for ^{133}Xe , $^{131\text{m}}\text{Xe}$, based on MC modeling
- For an operating pressure of 3 atm and a detection efficiency of 88%, the active xenon volume is 259 cm³.
- In whole air, xenon is present at the concentration of 87 ppb (0.087 cm³ per m³ of air), giving an equivalent air volume within the detector of 2,977 m³ of air.
- Radionuclide backgrounds at 0.01 mBq/m³ will thus produce a signal of **30 mBq** in a ULBPC; this is much higher than the 2 mBq sensitivity demonstrated for a 12-hour measurement.
- For the calculated sensitivity of 2 mBq for a 12-hour measurement, a LVXe sample counted in a ULBPC can thus expect to be sensitive to **0.00067 mBq/m³**, well below expected environmental levels.

Conclusions

- The PNNL ULBPC has more than sufficient sensitivity with a 3-atm load of a 98:2 Xe:CH₄ blend to measure radionuclide at expected atmospheric concentrations.
- Additional sensitivity may be achieved, if desired, by operating at higher pressures (detector limit is 10 atm) but would require more careful optimization of quench fraction and operating voltage.

References

1. Kalinowski, M.B., et al., *Discrimination of Nuclear Explosions against Civilian Sources Based on Atmospheric Xenon Isotopic Activity Ratios*. Pure and Applied Geophysics, 2010. **167**(4): p. 517-539.
2. Aalseth, C.E., et al., *Design and construction of a low-background, internal-source proportional counter*. Journal of Radioanalytical and Nuclear Chemistry, 2009. **282**(1): p. 233-237.
3. Aalseth, C.E., et al., *A shallow underground laboratory for low-background radiation measurements and materials development*. Review of Scientific Instruments, 2012. **83**(11): p. 113503-10.

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 The views expressed here do not necessarily reflect the opinion of the United States Government, the United States Department of Energy, or the Pacific Northwest National Laboratory.