October 25, 2023

Vapour Pressure of Xe Isotopes & The XeStill Project

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Motivation

- In the past, understand the triple point (TP) of xenon to be used as a **standard temperature**
  - Fail to produce credible data

Tew, W. L., Intl J of Thermophysics (2008), 29

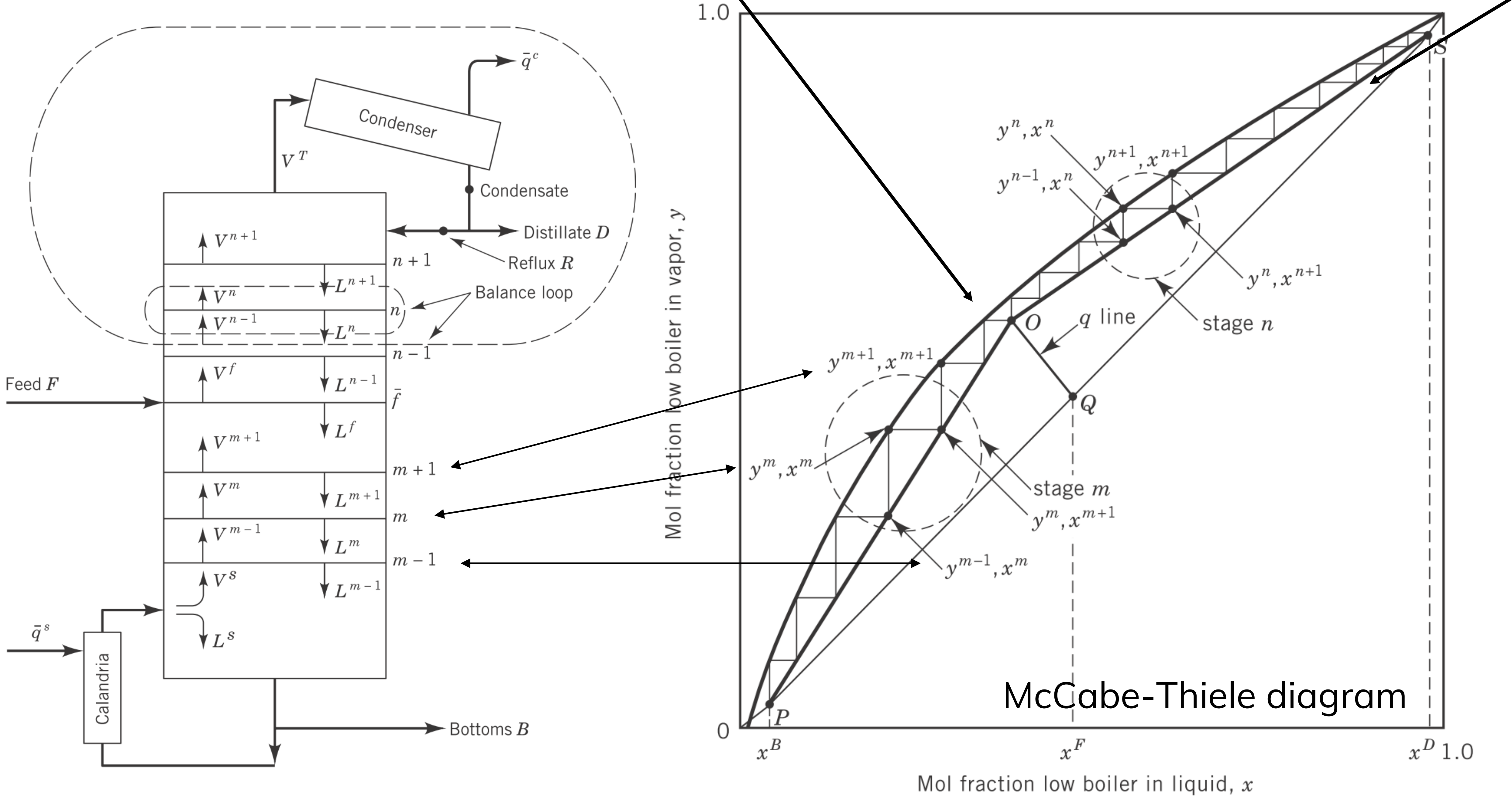
- More recently, need of **xenon enrichment** for $0\nu\beta\beta$ searches
  - Natural xenon: ~80% $^{128-132}Xe$, 8.9% ^{136}Xe
  - Only approach employed to enrich is via centrifugation: costly power consumption
  - Distillation uses gravity and isotopic differences in vapour pressures

- Cryogenic distillation was used to produce heavy water in the SNO experiment

Concepts of (Cryo) Distillation

Equilibrium line
(vapor pressure differences)

Operation line
(packing/trays, number of stages)



(Isotopic) separation parameters:

- Number of stages: N
- Height equivalent theoretical plate:
  - HETP = column height / N
- Vapor pressure differences
  - $\frac{\delta p}{p} = (p_h - p_l)/p$
- Vapour pressure isotopic effect:

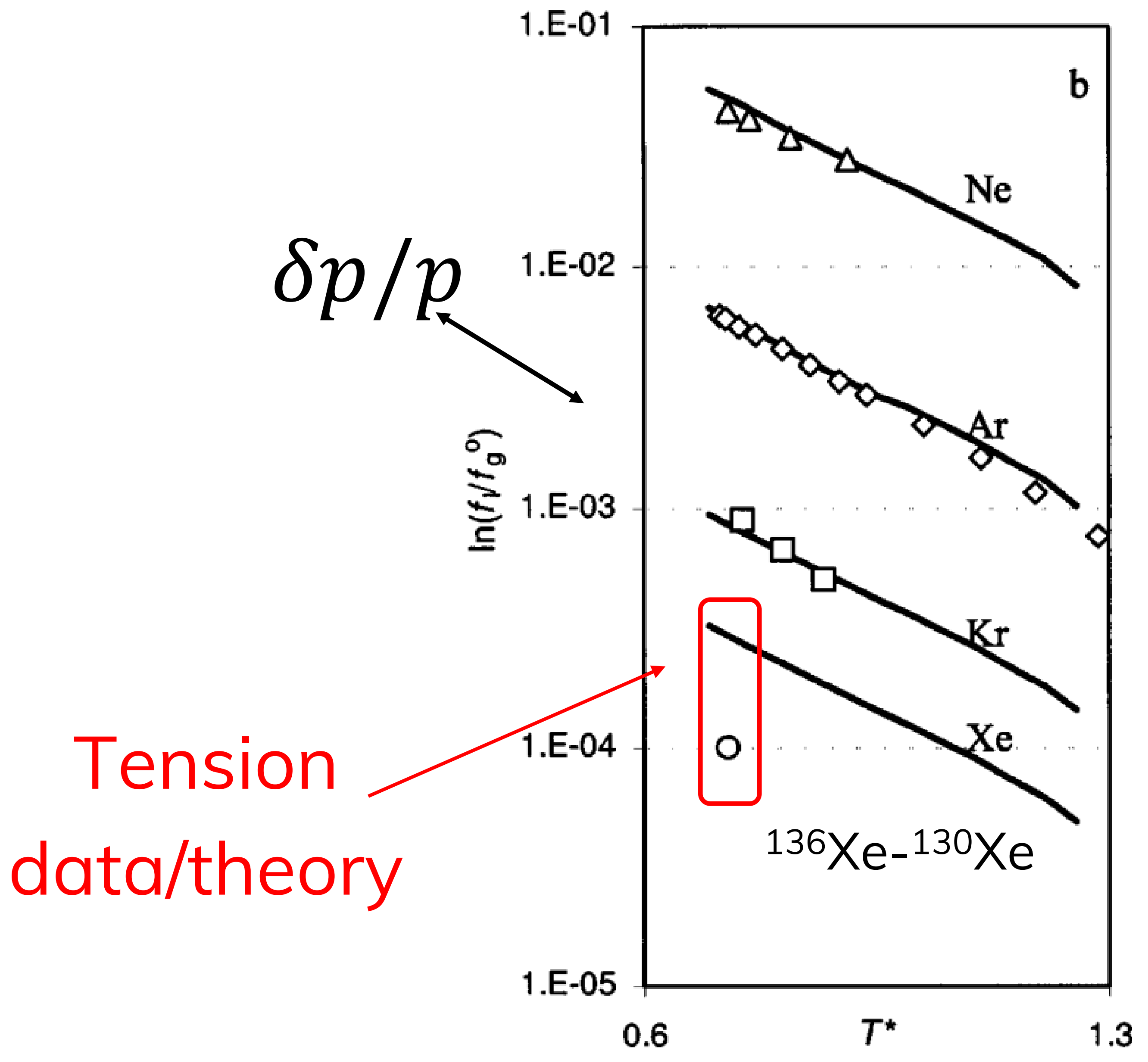
$$\mathrm{VPIE} := \ln\left(\frac{p_l}{p_h}\right) \simeq \ln\left(\frac{f_c}{f_g^0}\right) \simeq \ln(\alpha_{l-h})$$

J. Fair and H. Kister, "Distillation",

Kirk-Othmer Encyclopedia of Chemical Technology (2018)

Existing Data / Models

- Data exists for some fluid isotopes at various temperatures: ^{22}Ne-^{20}Ne, ^{40}Ar-^{36}Ar, ^{84}Kr-^{80}Kr
- Two thermodynamic models describe these fairly well

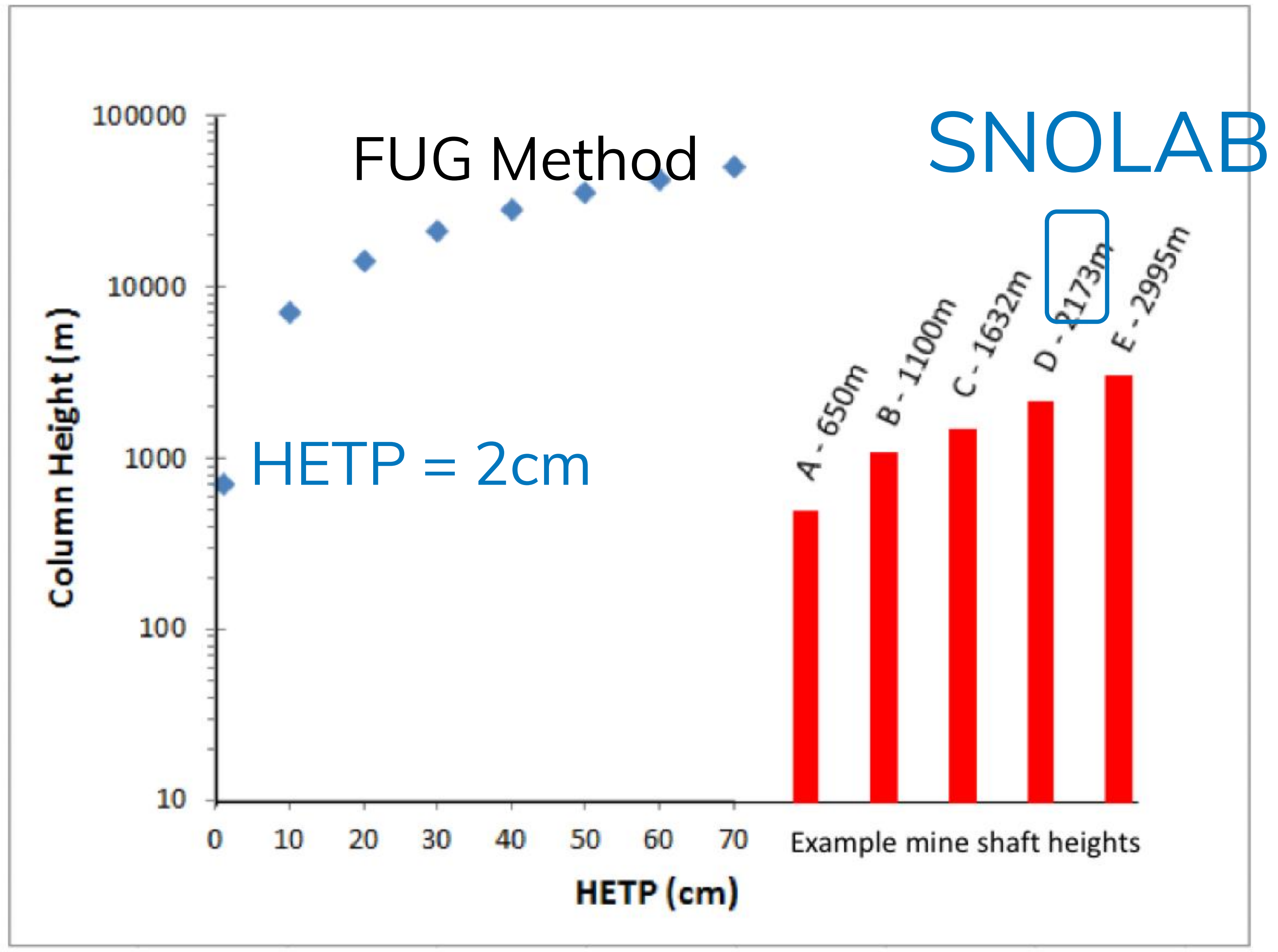


J. Canongia-Lopes et al J. Chem. Phys. 118, 5028 (2003)

$$\ln \alpha_{l-h} = \frac{\hbar^2}{24}\frac{1}{\varepsilon \cdot \sigma^2}\frac{\Delta m}{m_l m_h}\left(\frac{A}{T^{*2}} + \frac{B}{T^*} + C\right) \propto \Delta m/m^3$$

Theoretical prediction is 1/3 of Kr:

$\delta p/p \sim 0.1\ \%$ → sensitivity need of 0.01%

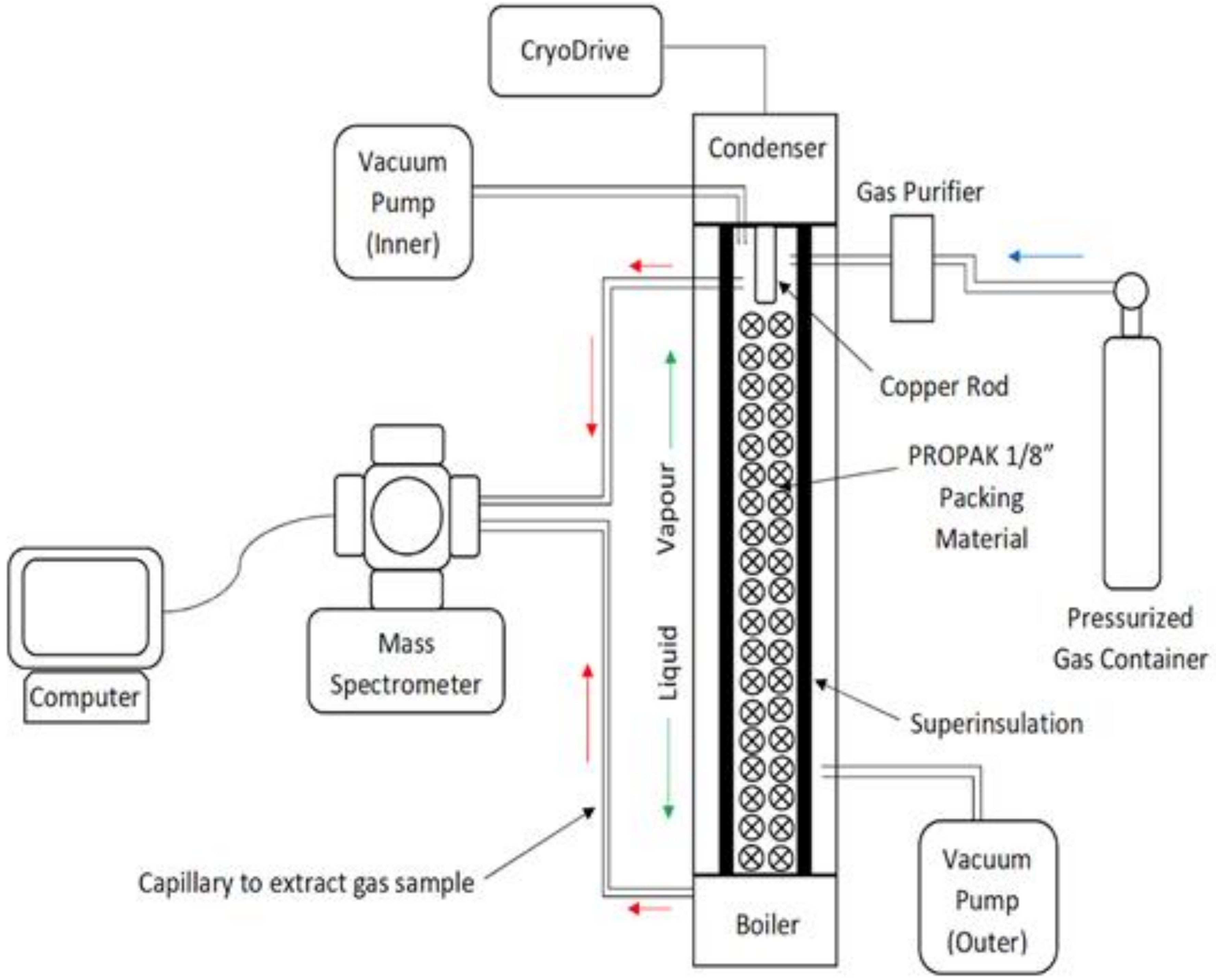


H.O. Back et al 2017 JINST 12 P09033

Still Setup @ Carleton

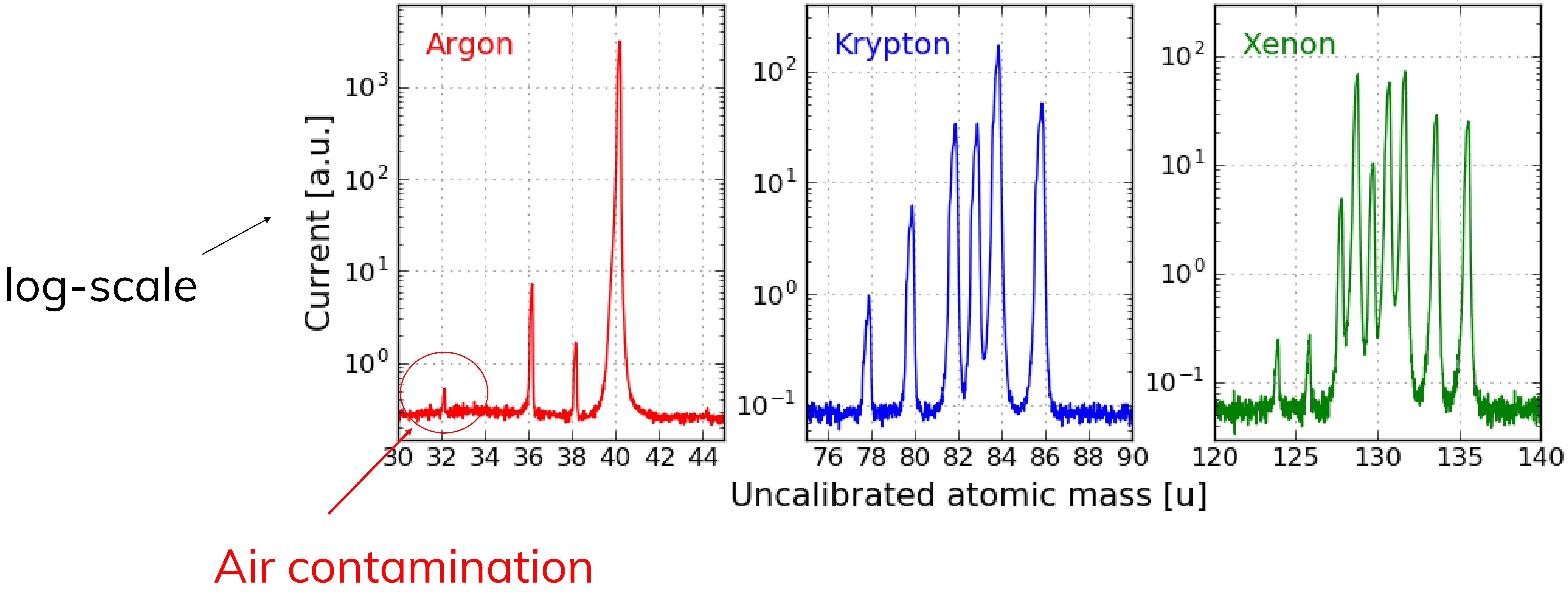
- 1.8 m high cryogenic still
- 25 mm inner diameter vacuum insulated reflux column filled with PROPAK (random) packing

- One condenser placed at the top and re-boiler at the bottom
- 100% reflux
- Pressure of 105 kPa



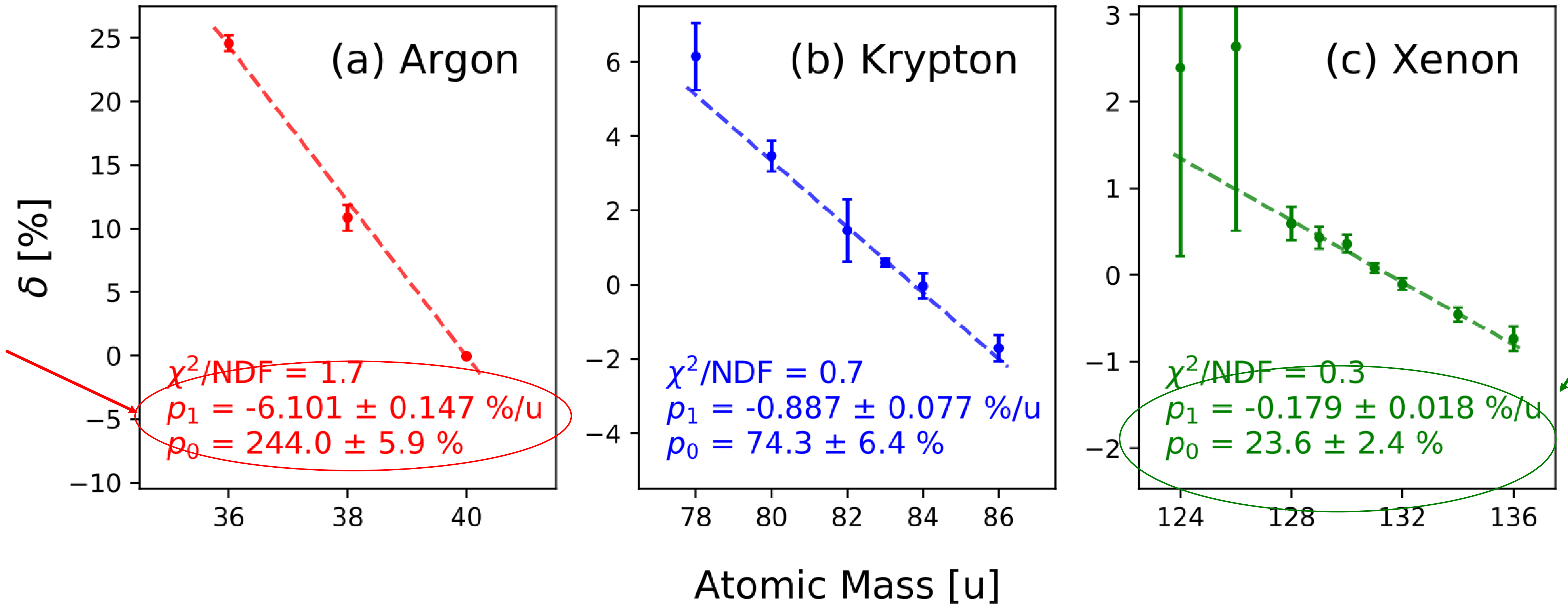
Mass Spectra

- EXTREL quadrupole: 19 mm bore, mass resolution of 1:2000
- Focus on single ions



Analysis Procedure

~4% stat. error



~10% stat. error

$$\delta = \frac{f_{i,top} - f_{i,bot}}{(f_{i,top} + f_{i,bot})/2} \times 100$$

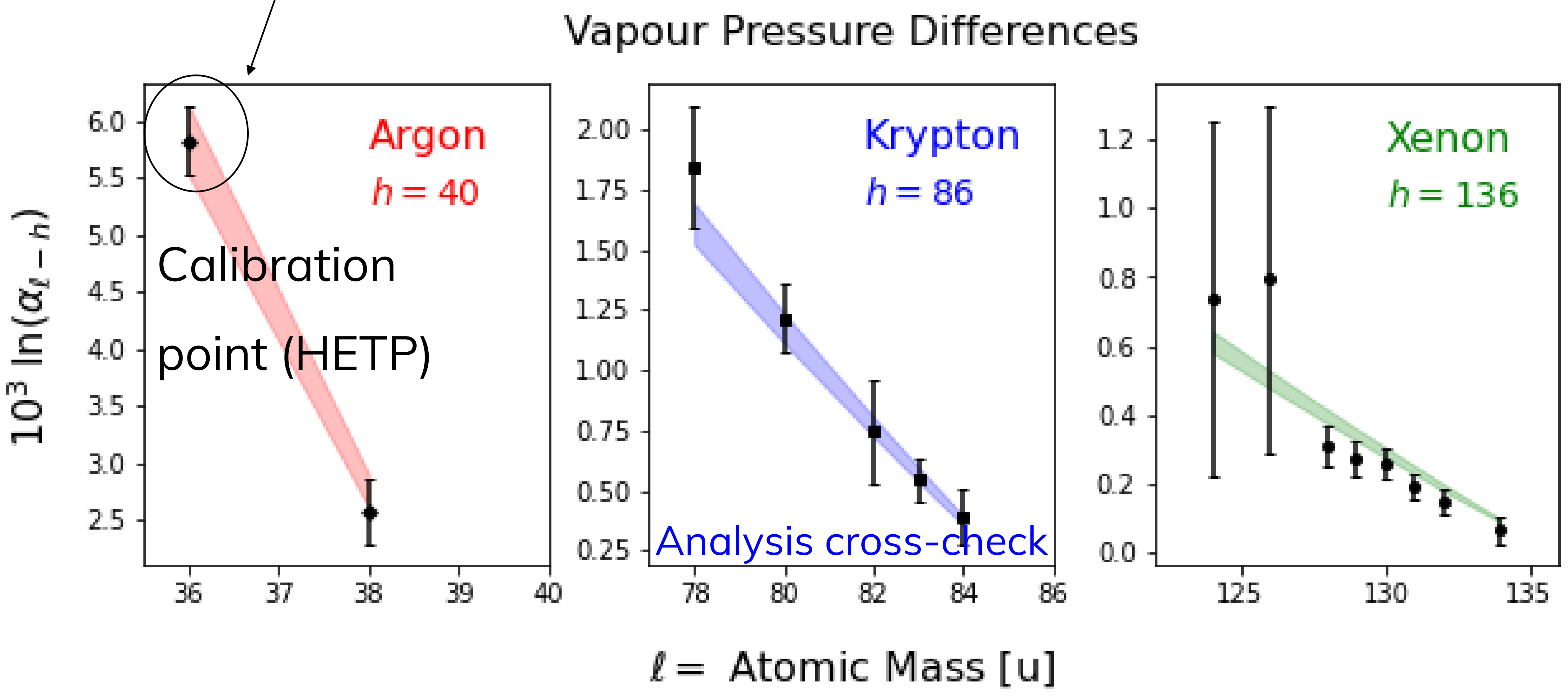
⟷ Measurement of relative enrichment top-bottom per isotope

Initial Results

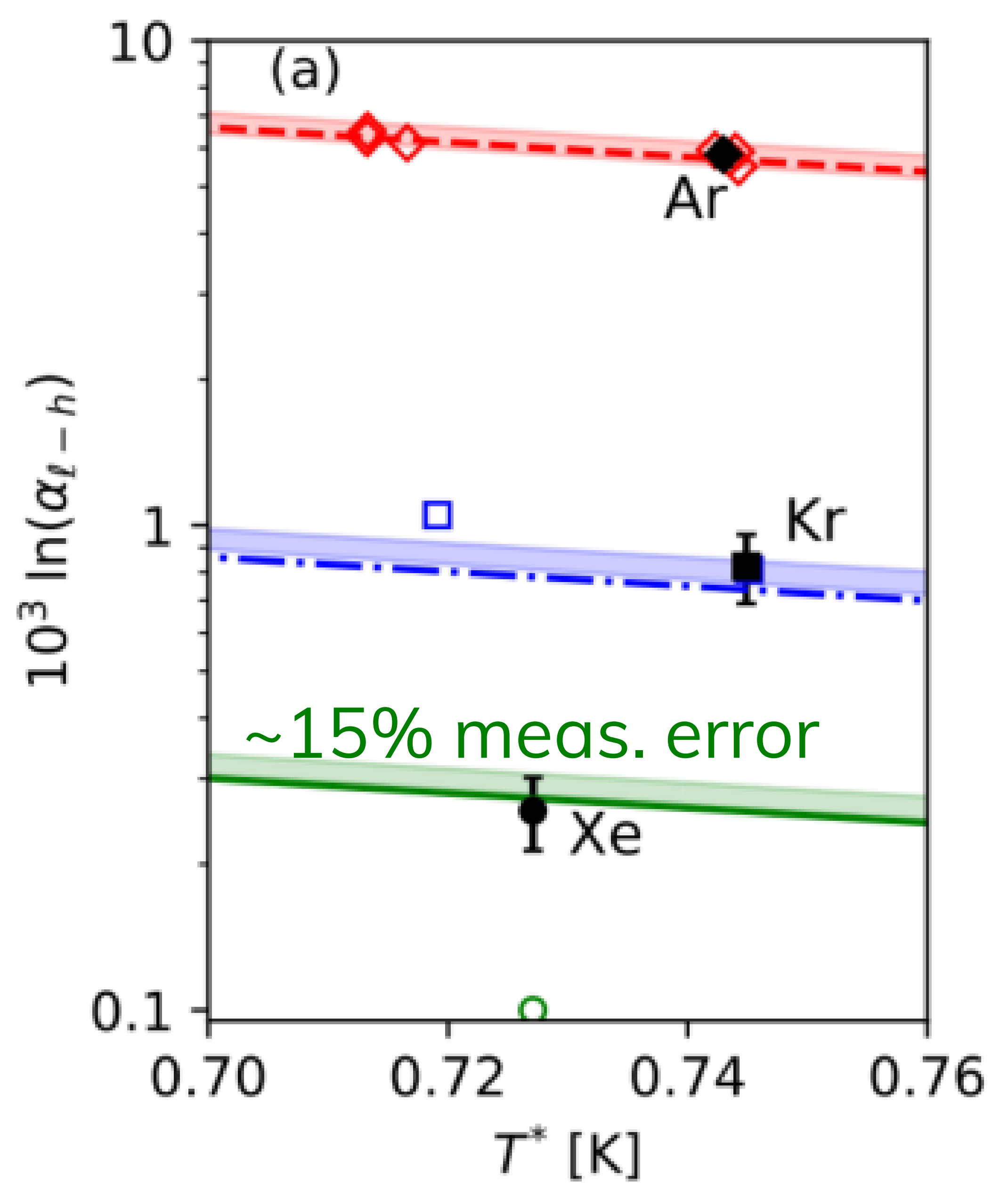
100% reflux, Fenske equation: $N(t) = \frac{\ln((\delta_{l-h})(t))}{\ln((\alpha_{l-h})(t))}$ ↔ $\delta_{l-h} = \frac{(1+\delta_l/200)/(1-\delta_l/200)}{(1+\delta_h/200)/(1-\delta_h/200)}$.

HETP = 4.23 ± 0.25 cm

N = 42.5 ± 2.5 stages



A. Alamre et al, ACS Omega 2020, 5, 45, 28977–28983



Still @ SNOLAB

Located in the Cryopit, 8x increase, ~14m high

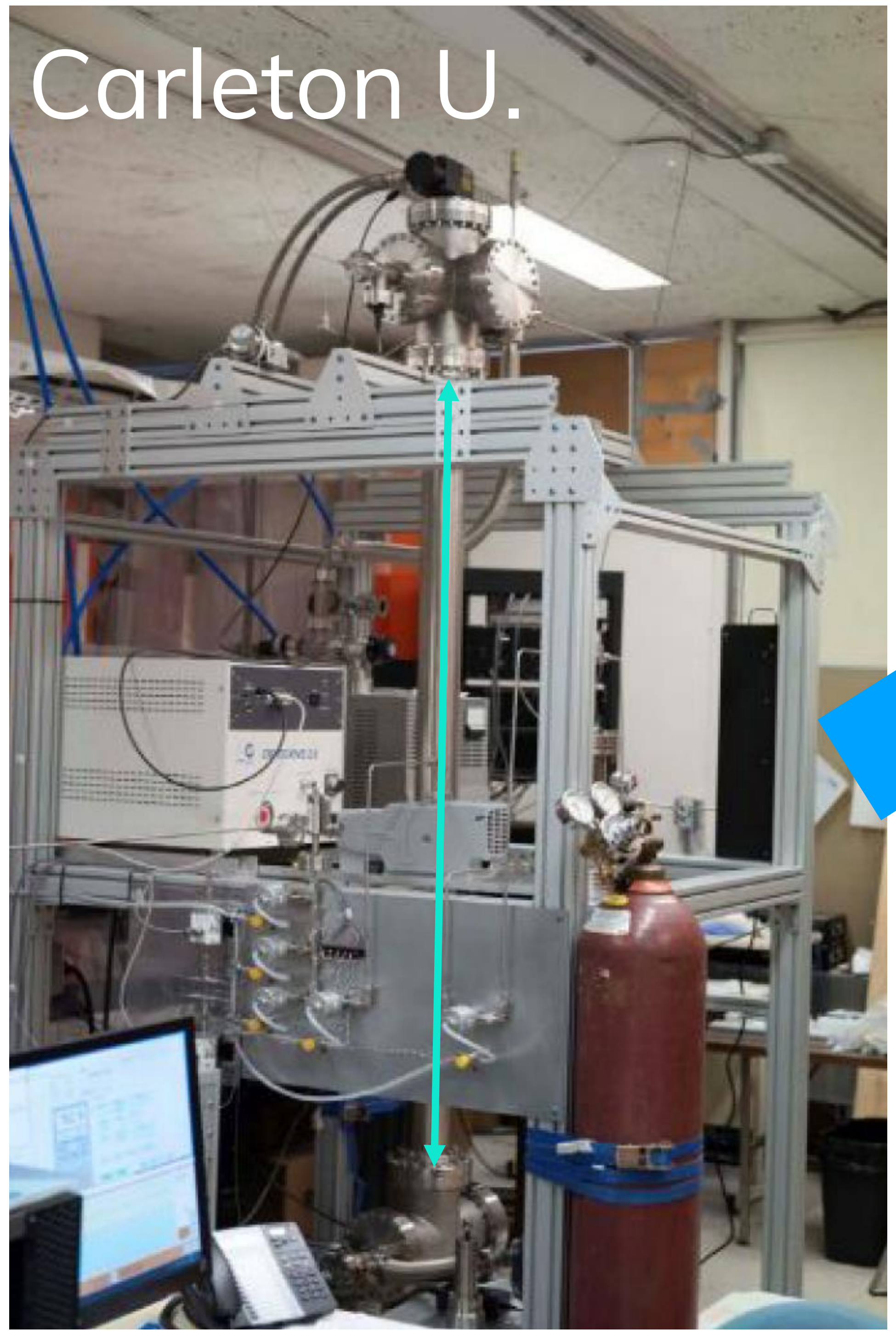


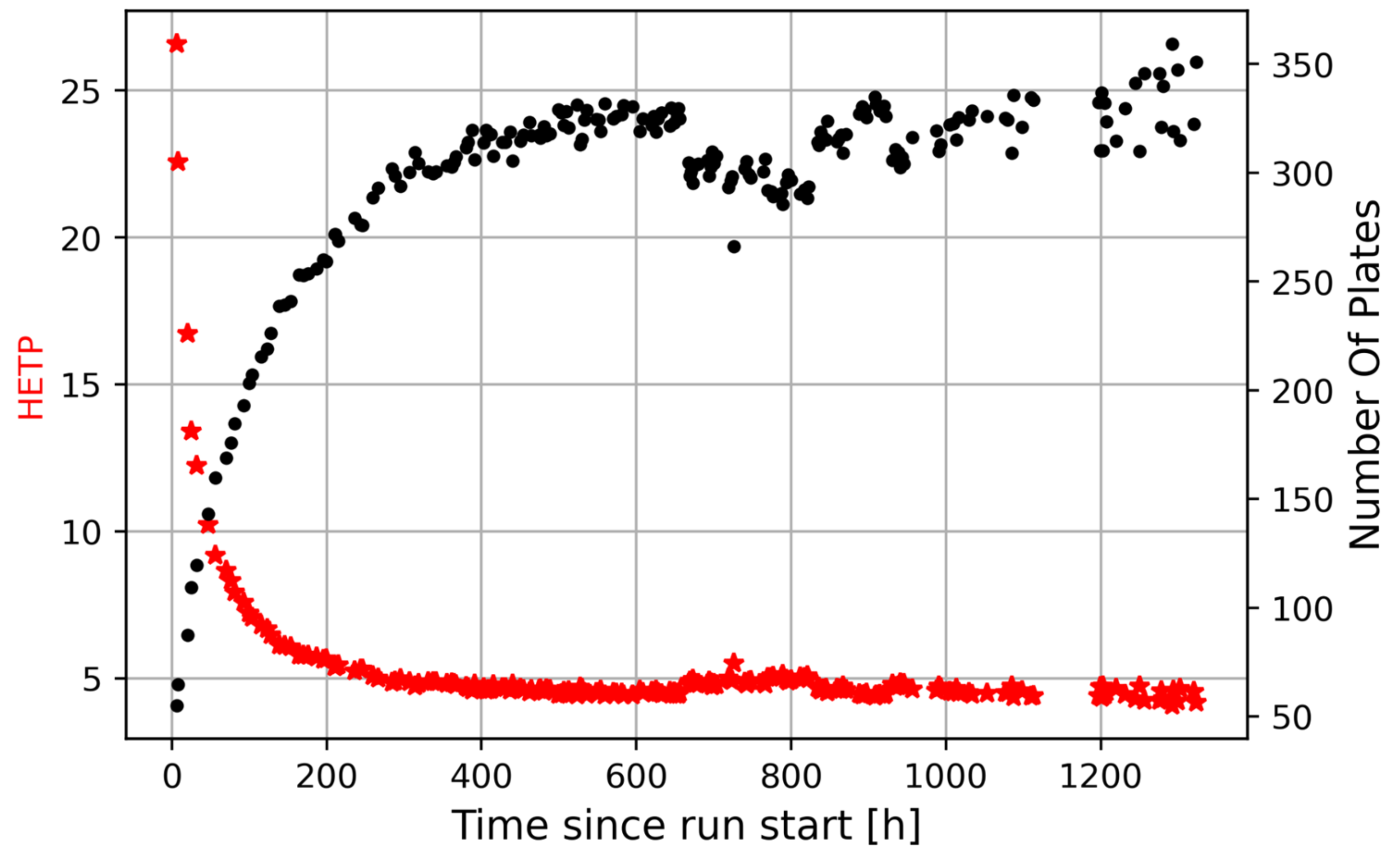
Photo courtesy of Andrea Pocar

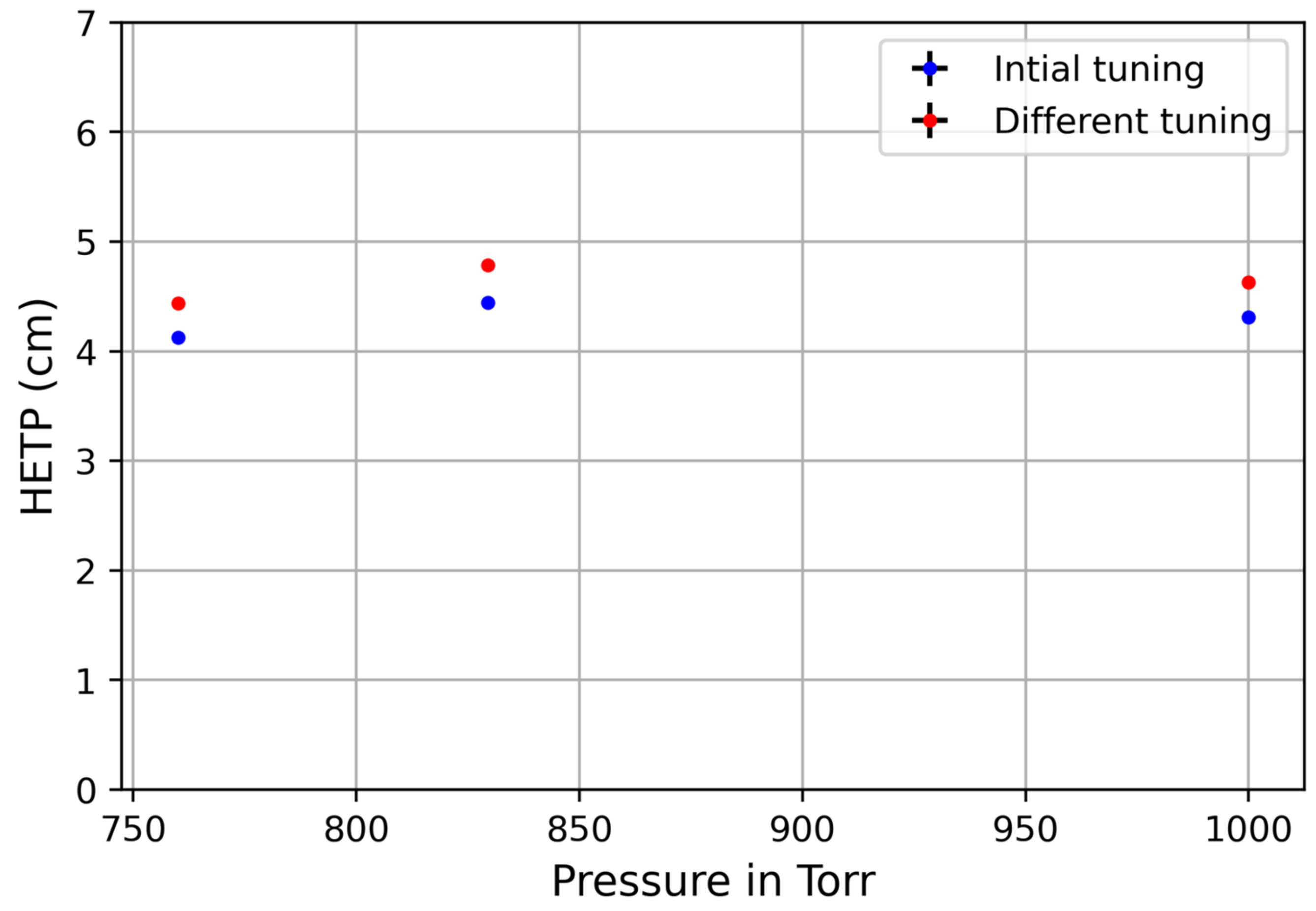
SNOLAB Results (so far)

- MSc thesis (A. Emara, 2023) measured HETP for argon

SNOLAB vs Carleton
Number of stages: 328.7±0.9 (vs 42.5±2.5)
HETP: 4.29 ± 0.13 cm (vs 4.23 ± 0.25 cm)

- Data for krypton obtained this summer 2023, on-going analysis, validation of HETP
- Running for 2 years, weeks or months at a time
- Challenges running underground:
    - Long evolution time (~month vs week)
    - Operations, hardware maintenance
    - Precision measurements, systematics





XeStill Team

Carleton University:

- David Sinclair (PI)

University of Windsor

- Caio Licciardi (PI), Drake Wickman (MSc student)

SNOLAB:

- Steffon Luoma (scientist), Ashley Mathewson (project engineer)

Former members:

- *Jason Anstey, Jeff Mason (technical staff), Vance Strickland (project engineer)*
- *Abo-Bakr Emara (Carleton, MSc graduate), Amal Alamre (Carleton, MSc graduate), Brandon Death (Carleton, undergrad thesis)*
- *Dr. Ibtesam Badhrees*
- *SNOLAB scientists: Erica Caden, Jeter Hall, Steve Linden, Brian Morissette*

Conclusions

Cryogenic distillation is a potential approach for xenon enrichment

Distillation relies on vapour pressure differences between xenon isotopes

- Very small, requires precision measurements for optimal design
- Estimated column heights ~100-1000s meters

Delivered results at 15% error, consistent with existing expectations

Working on precision measurements with higher column at SNOLAB's Cryopit

- Successful calibration measurement
- Validation is on-going
- Xenon measurements expected in Winter 2024