



Novel Applications of TRIUMF's Accelerated Beams with ASPIRE

Chris Charles

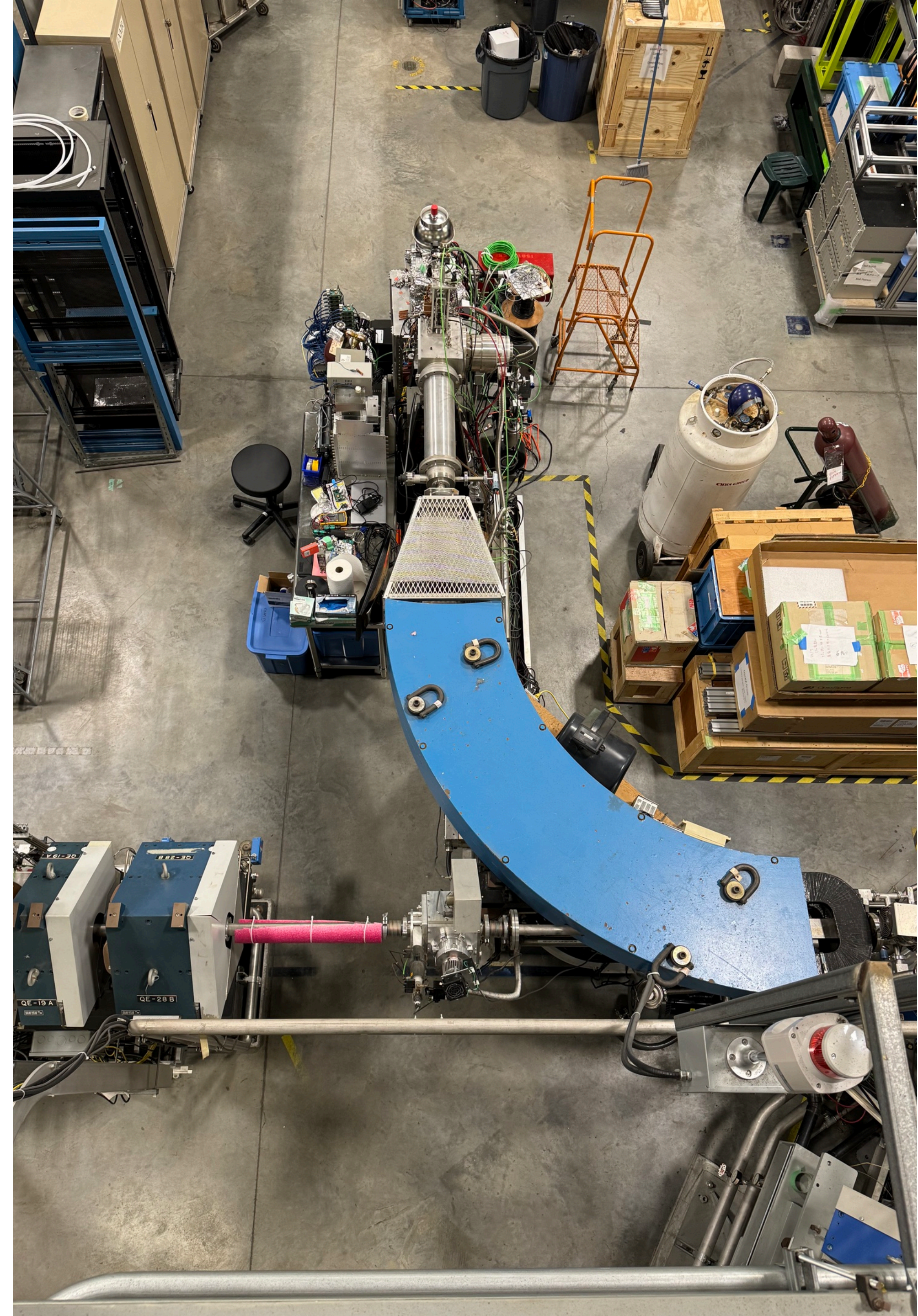
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TRIUMF Science Week 2024

Monday, July 22, 2024 @ 10:25-10:45 am

Accelerator Science Session, MOB Auditorium



**Discovery,
accelerated**

Today

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- What is ASPIRE ?
 - ✓ Introduction to the new ASPIRE experimental facility at TRIUMF.
- 3 upcoming ASPIRE projects:
 - ✓ Fission-track dating enhancement of geologic mineral grains.
New “regime” for fission-track age-dating in geology (1st beam time in August).
 - ✓ Astrochemistry on icy interstellar grains.
Production/degradation of complex molecules on grain surfaces in space.
 - ✓ Radiolysis of water & origins of life.
Implications to natural production of organic molecules by radiation.

Acknowledgments

(no particular order)

TRIUMF

- Marco Lovera
- Brian Minato
- Friedhelm Ames
- Ray Mendoza
- Gelo Ramon
- Martin Alcorta
- Dave Prevost
- Oliver Kester
- Shaun Georges
- Olivier Shelbaya
- Nigel Smith
- Dimo Yosifov
- Mike Vogel
- Doug Preddy
- Derek Orth
- Edi Dalla-Valle
- Peter Kunz
- Jens Lassen
- Aurelia Laxdal
- Devon Joseph
- Scott Kellog
- William Huang
- Heidi Awad
- Everton Shaw
- Tiffany Angus
- Hannah Dahl
- Chloe Beaumier-Martin
- Camille Belanger-Champagne
- RIB Operations Group
- Spencer Kiy
- Hayden Klassen
- Scott Kajioka
- Brad Noakes
- Pauline Dela Zilwa
- Paul Jensen

SFU Chemistry / TRIUMF

- Corina Andreoiu
- Devon Joseph
- Nabyl Merbouh

TRIUMF Life Sciences (expt. #L-169)

- Brooke McNeil
- Joe Huser
- Vicky Hanemaayer
- Gloria Botelho

uWaterloo Phys / Chem

- Scott Hopkins
- Ariana Pearson

Kwantlen Polytechnic U.

- Ron Murray

Yale, Earth & Space Inst.

- Phil McCausland

UCalgary Earth Sciences

- Eva Enkelmann
- Birk Haertel
- Akeek Maitra

3

uOttawa Earth Sciences

- Oliver Warr

UofT Earth Sciences

- Min Song
- Barbara Sherwood-Lollar

UWO Earth Sciences

- Roberta Flemming

UBC Chemistry

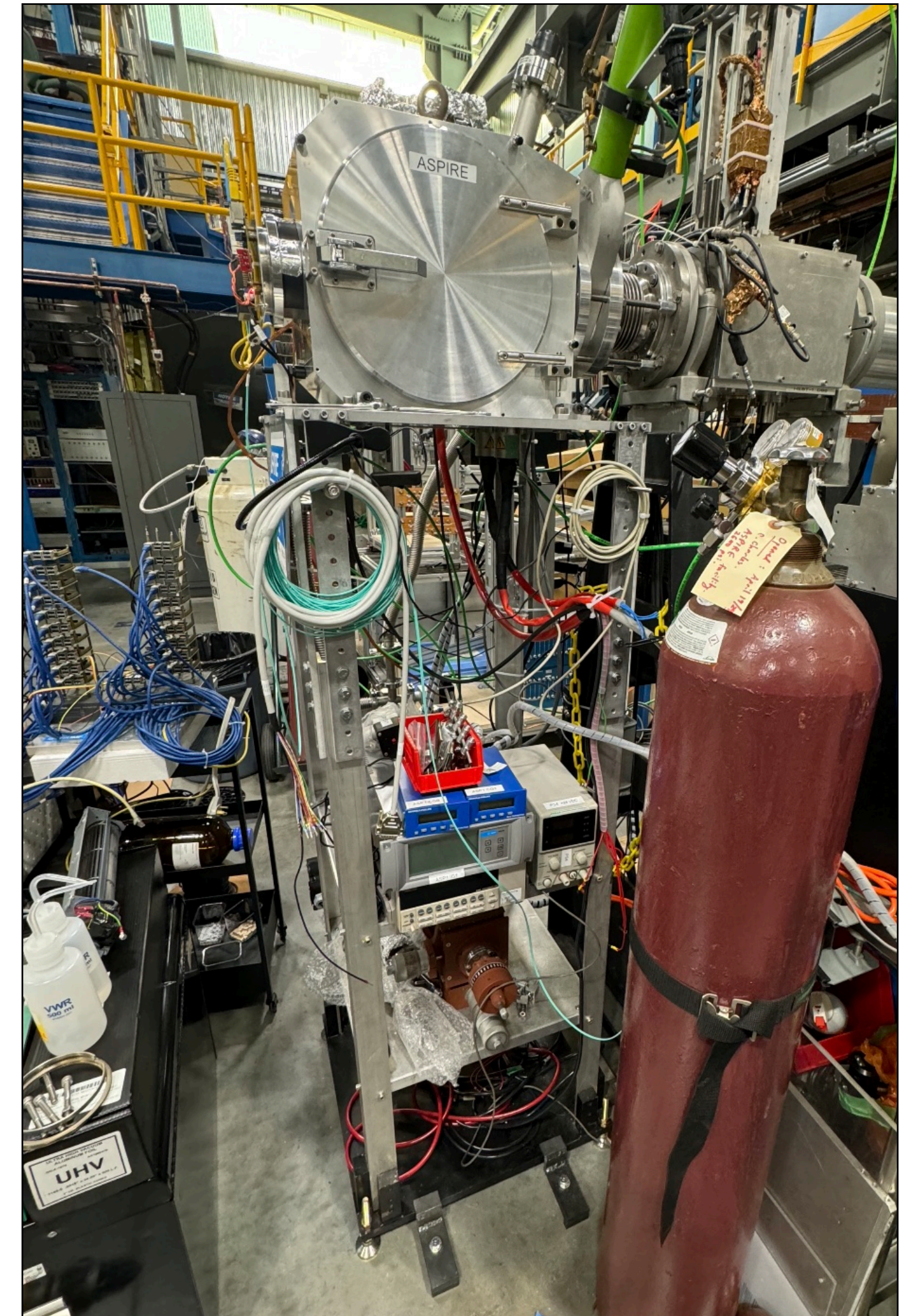
- Ilsa Cooke
- Elsa Yuan
- Arie Irving-Hughes

& BCIT IT Systems Tech.

ASPIRE



Astrochemistry & Planetary
materials Irradiation
Experimental facility



ASPIRE — location in ISAC-I (HEBT1)

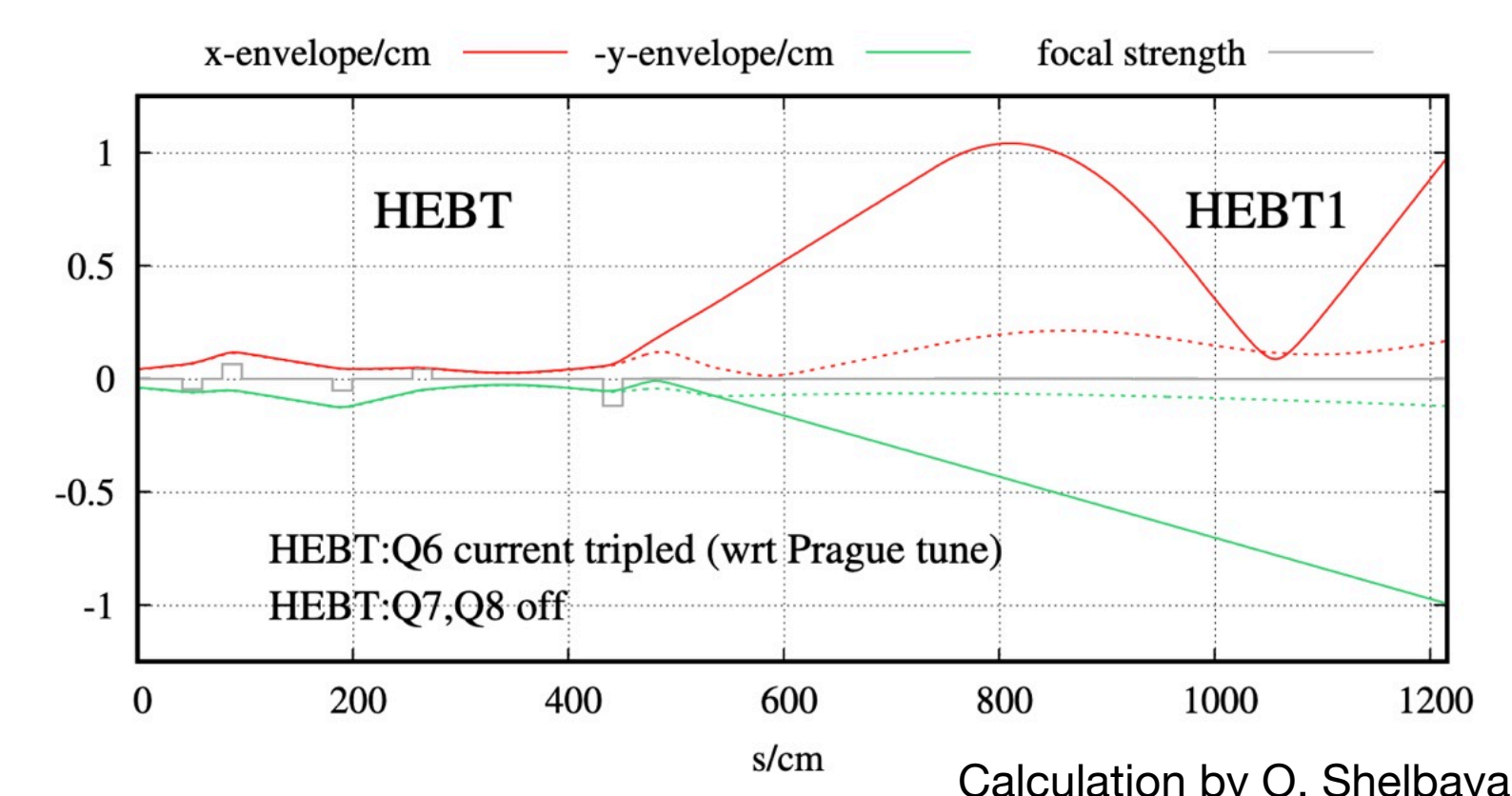
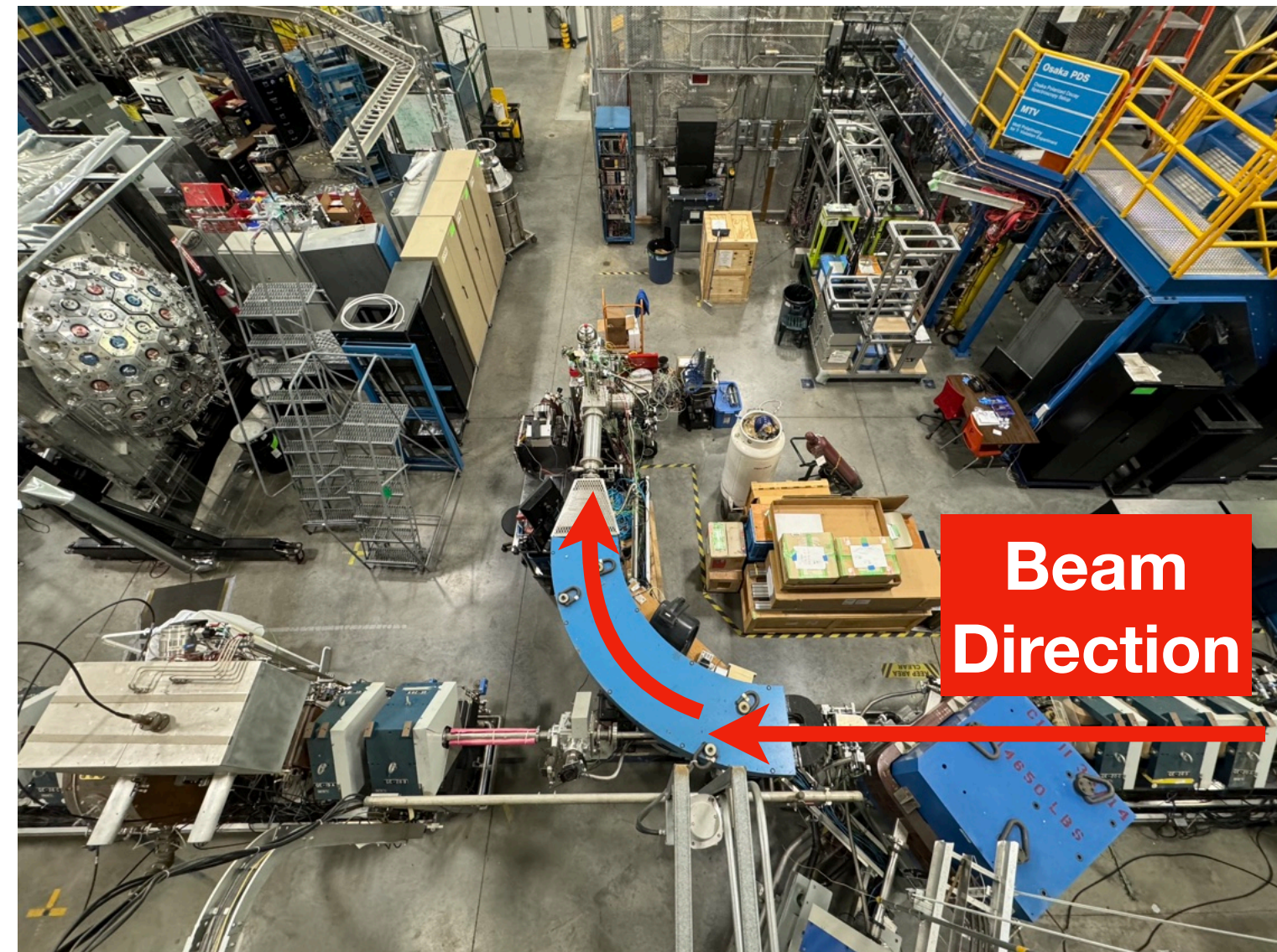
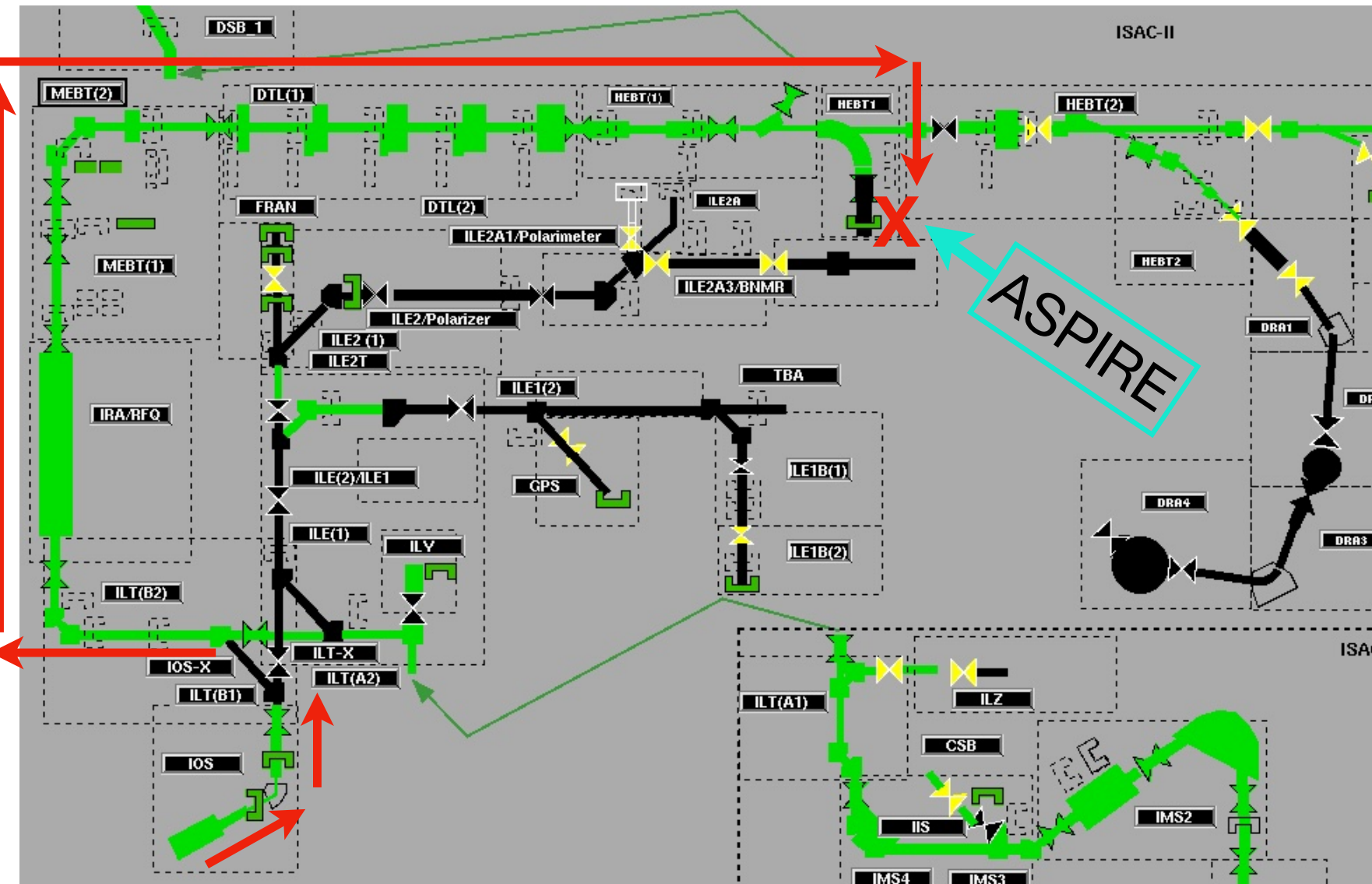
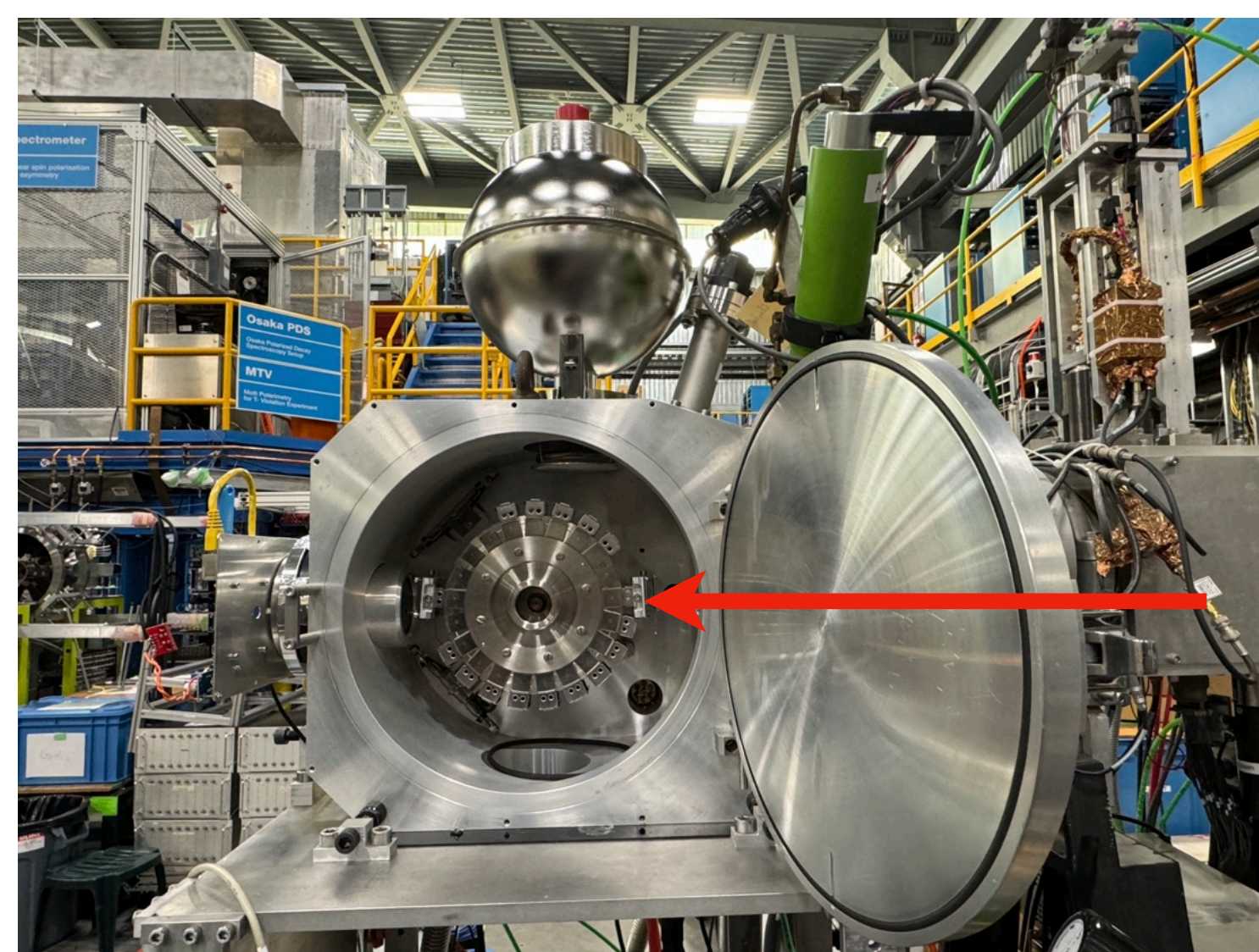
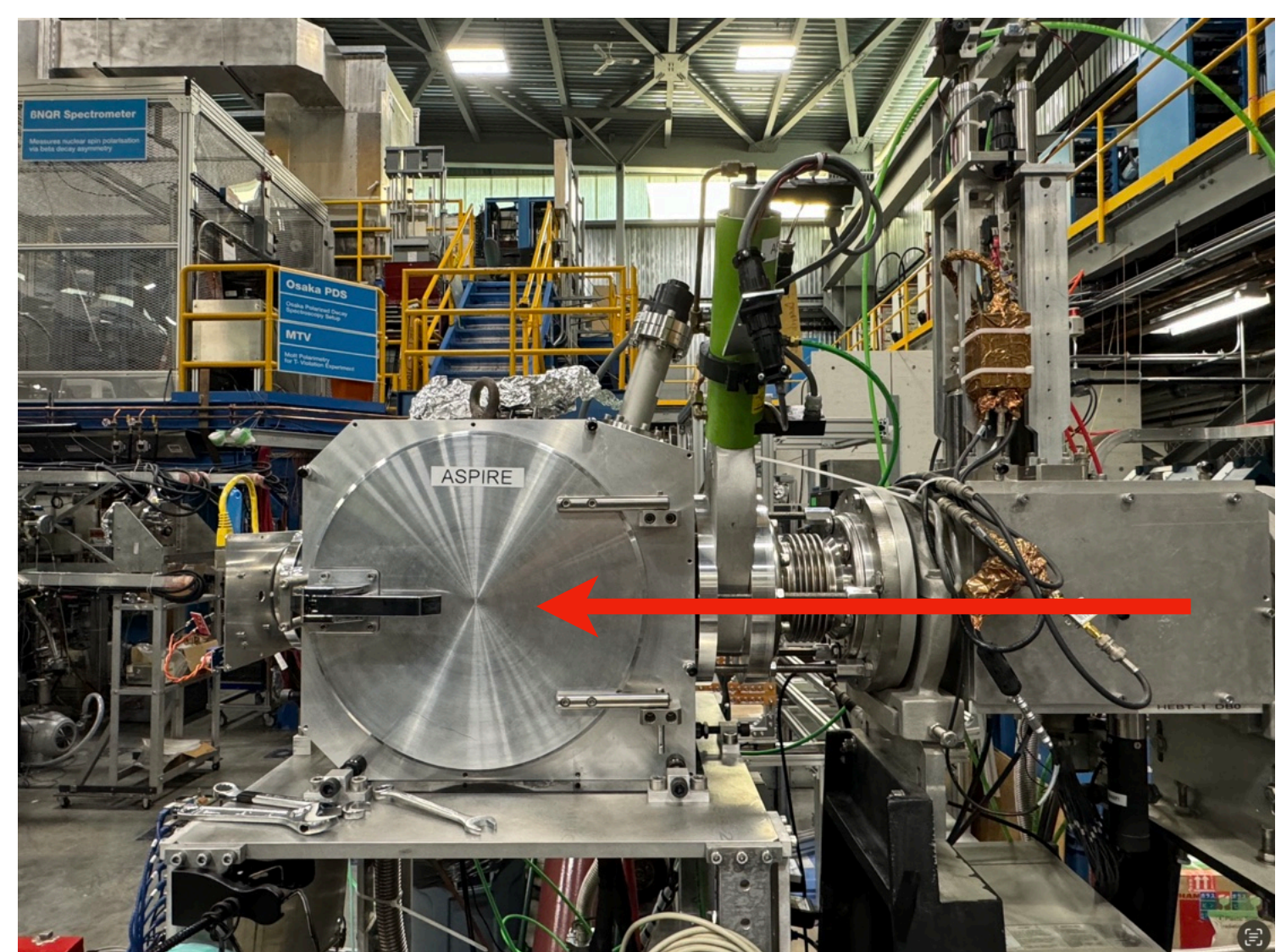


Figure 2: TRANSOPTR simulation[1] of accelerated beam through ISAC-DTL, using up to Tank-4, for an E/A of 1.0 MeV/u.



Sample(s)

1.5 MeV/u (max)
HEBT-1 in ISAC-I

- Solid
- Liquid
- Gas
- Icy/frozen
- ... combinations...

OLIS stable beam
i.e. $^{84}\text{Kr}^{+15}$ HEBT +21

Scientific purpose of ASPIRE

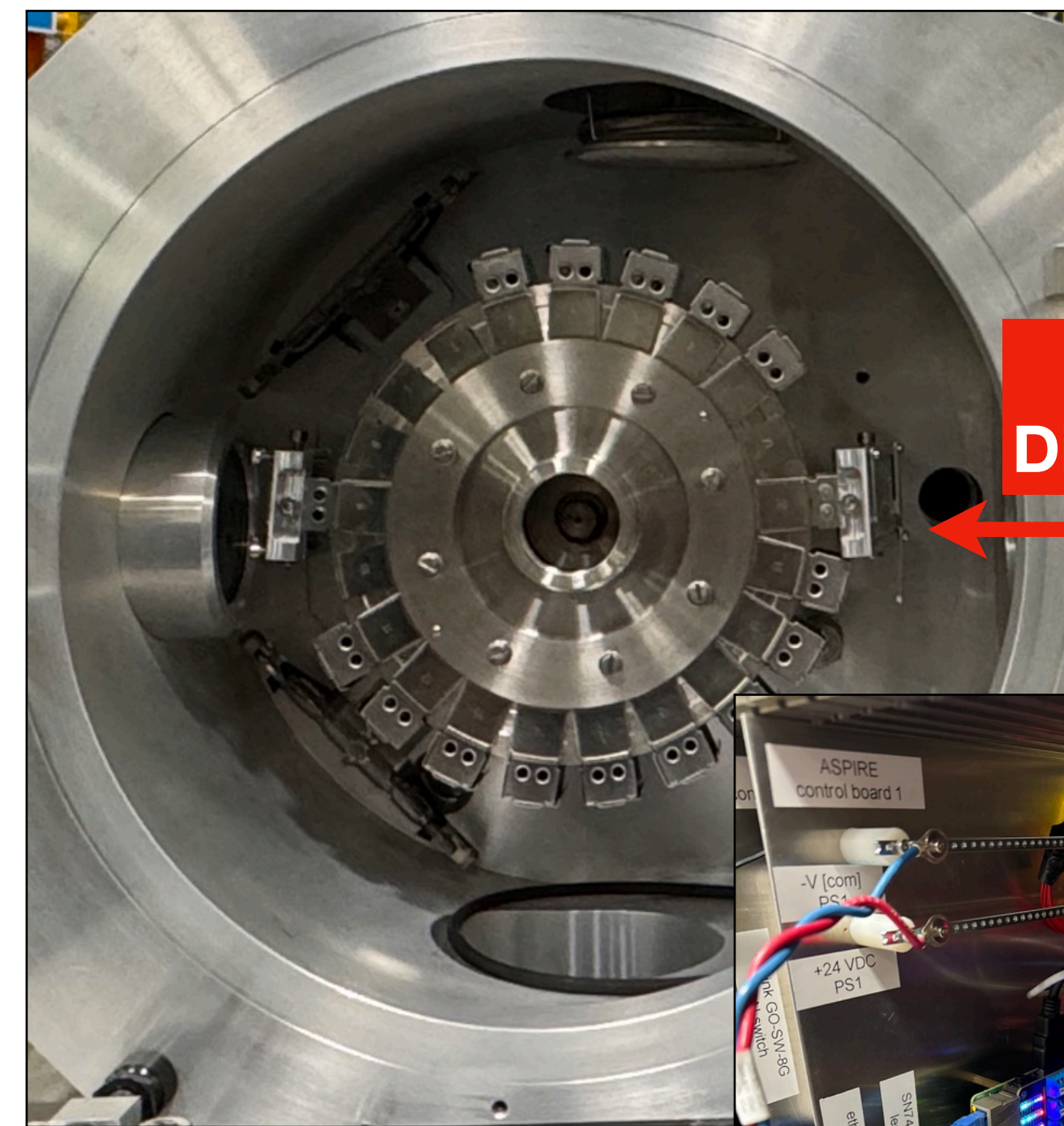
Non-Exhaustive List of Science using TRIUMF Accelerators & Radioisotopes

- Irradiate diverse types of samples with ISAC accelerated beams.
- Enable leading-edge interdisciplinary science with TRIUMF accelerators, of high benefit to the wide scientific community.
- Explore new “non-traditional” research areas for TRIUMF + collaborators.

FIELD	SCIENCE PROBLEM(S)	COLLABORATORS
Astrochemistry	(i) interstellar, nebular, and PPD element-isotope fractionations, enrichments, and depletion processes, (ii) radiation-induced molecular chemistry on interstellar icy grains , (iii) solar wind studies & implications for SS or exoplanet chemistry.	Ilsa Cooke (UBC) Eva Enkelmann (uCalgary)
Cosmochemistry	(i) irradiation of meteoritic materials, (ii) early solar system chemistry, (iii) planetary/exoplanetary atmospheres, (iv) PPD homogeneity vs heterogeneity & canonical ratios of CAIs and chondrules for radioisotopic chronometers .	Scott Hopkins (uWaterloo) Oliver Warr (uOttawa)
Radiolysis	(i) water radiolysis, origins of life studies (organic molecules from inorganic starting materials) , (ii) degradation of contaminants in polluted water, (iii) fundamental water radiolysis physics/chemistry, (iv) uWaterloo FEL & ARIEL E-Linac science.	Barb Sherwood-Lollar (UofT) other...
Terrestrial Geology	(i) improving fission track age-dating of geological minerals , (ii) beam-induced damage effects in geologic minerals with applications to radiometric chronometry.	TRIUMF INTERNAL: Nigel Smith Peter Kunz Camille Belanger-Champagne Life Sciences Division
Other	(i) irradiation of electronics and advanced materials with heavy accelerated beam, (ii) material science applications, (iii) medical or virus research with heavy accelerated beams.	other...

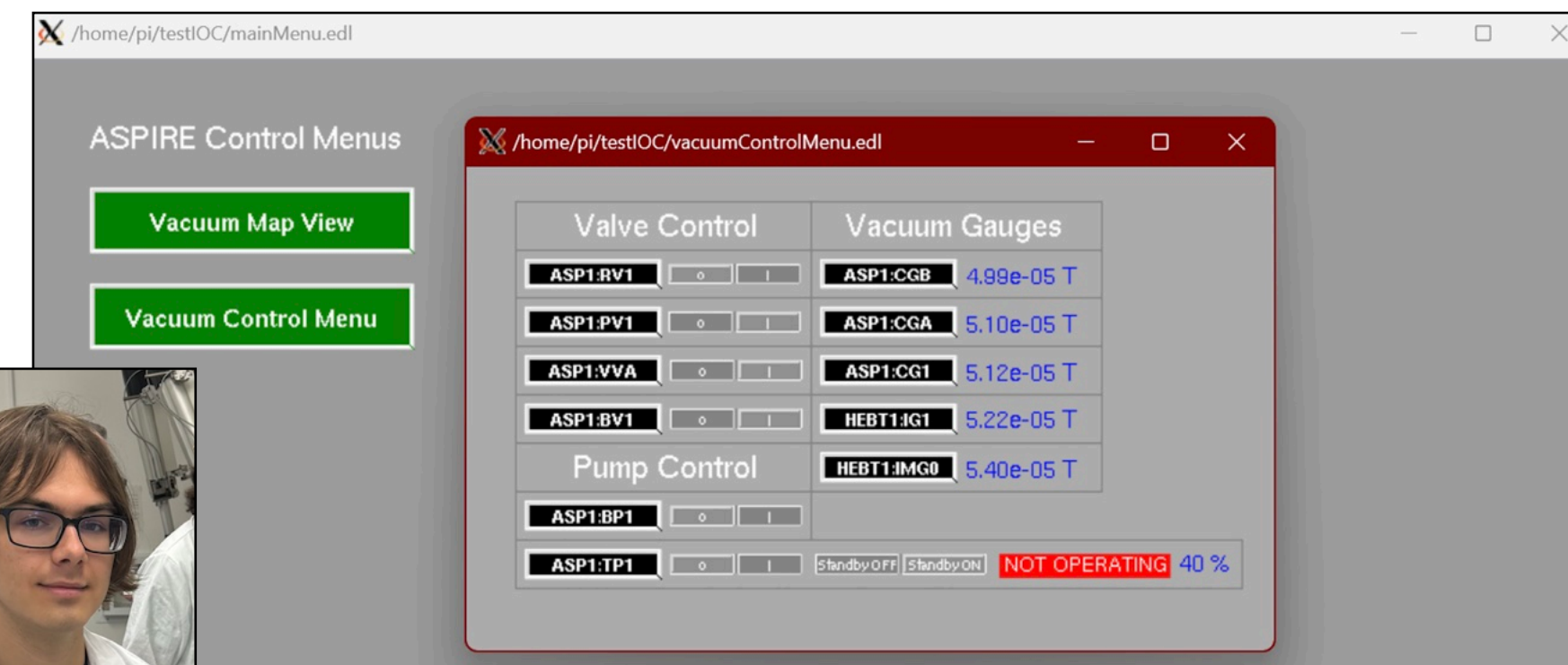
Some ASPIRE Details

- Highly customizable vacuum chamber for diverse/unique experiments, unique sample “targets”, diagnostics or analytical instruments, etc.
- Significant number of signal input-output capabilities on the main vacuum chamber (and HV compatible).
- Stepper motor & multi-position barrel for multiple sequential sample irradiations (can be removed and other target configurations installed).
 - ✓ *Custom* Zn-P-Ag phosphor screen + Raspberry Pi low-light camera diagnostic (to see beam shape & distribution).
 - ✓ *Custom* Faraday Cup at the sample plane, to assist with tuning.
 - ✓ *Custom* control system has been developed. Running **Pi-EPICS**, programmed by Hayden Klassen (student) for monitoring and controlling devices connected to the system, data acquisition.
 - ✓ Many possibilities for customization & expansion.



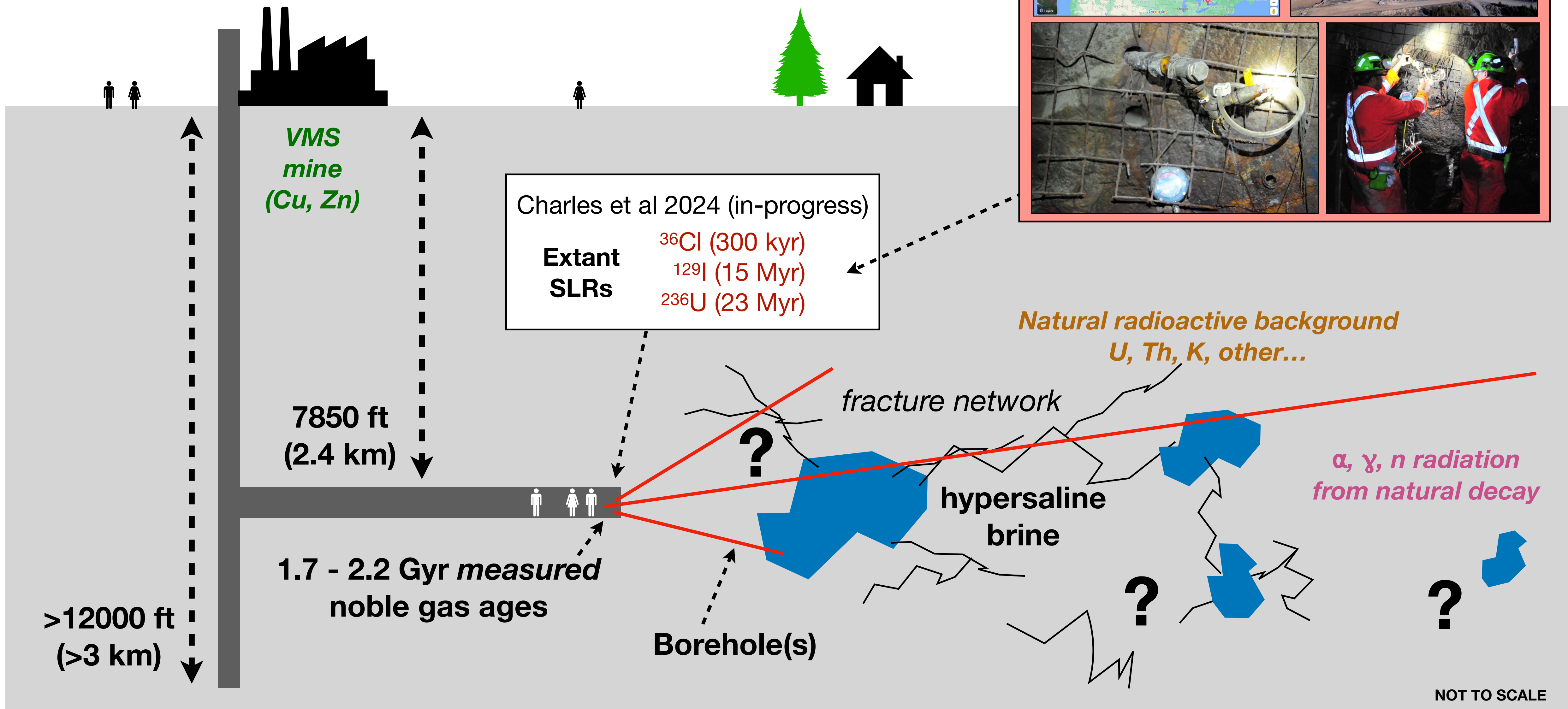
Beam
Direction

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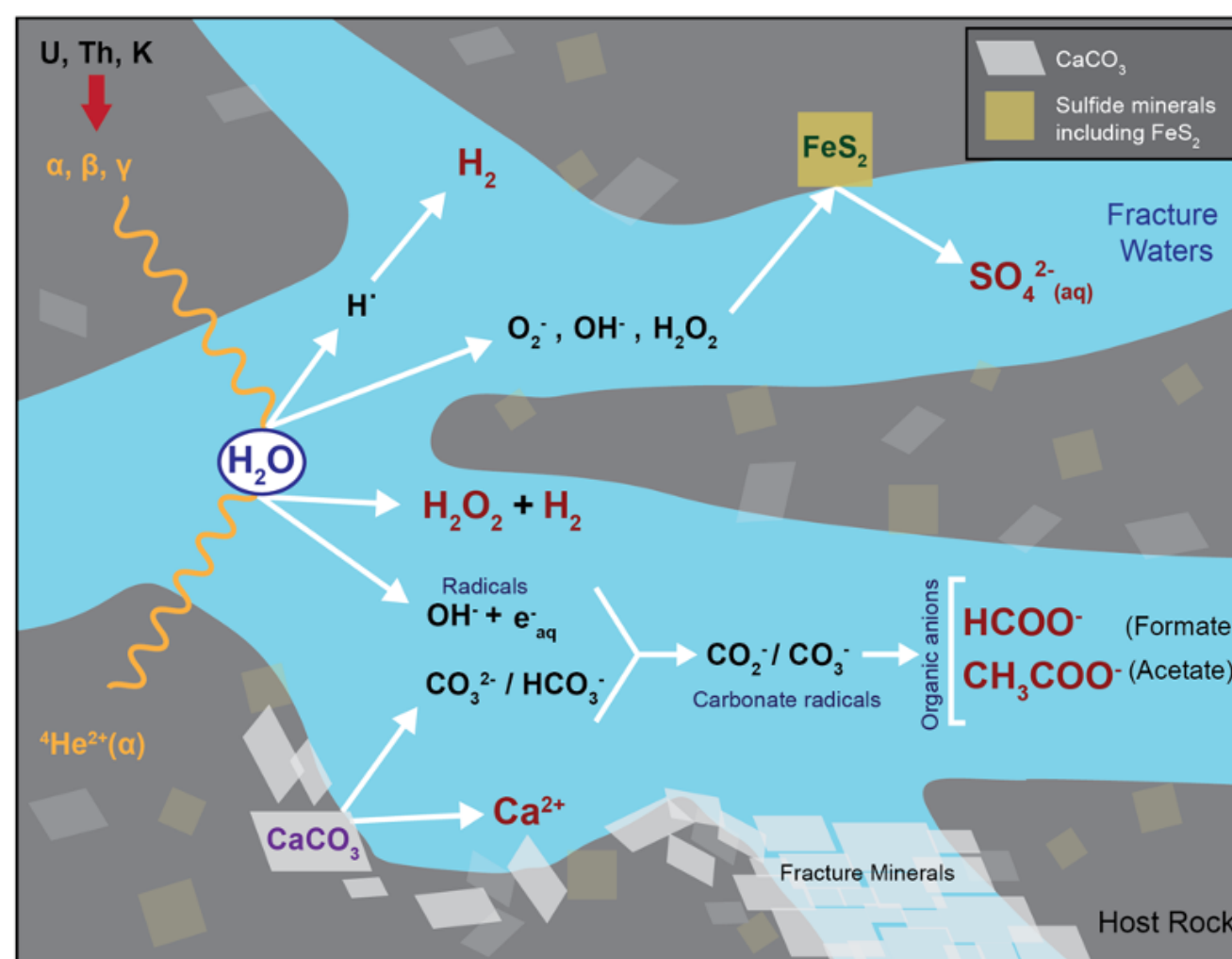
Radiololysis

Kidd Creek Mine, Timmins, ON
— simplified fracture network

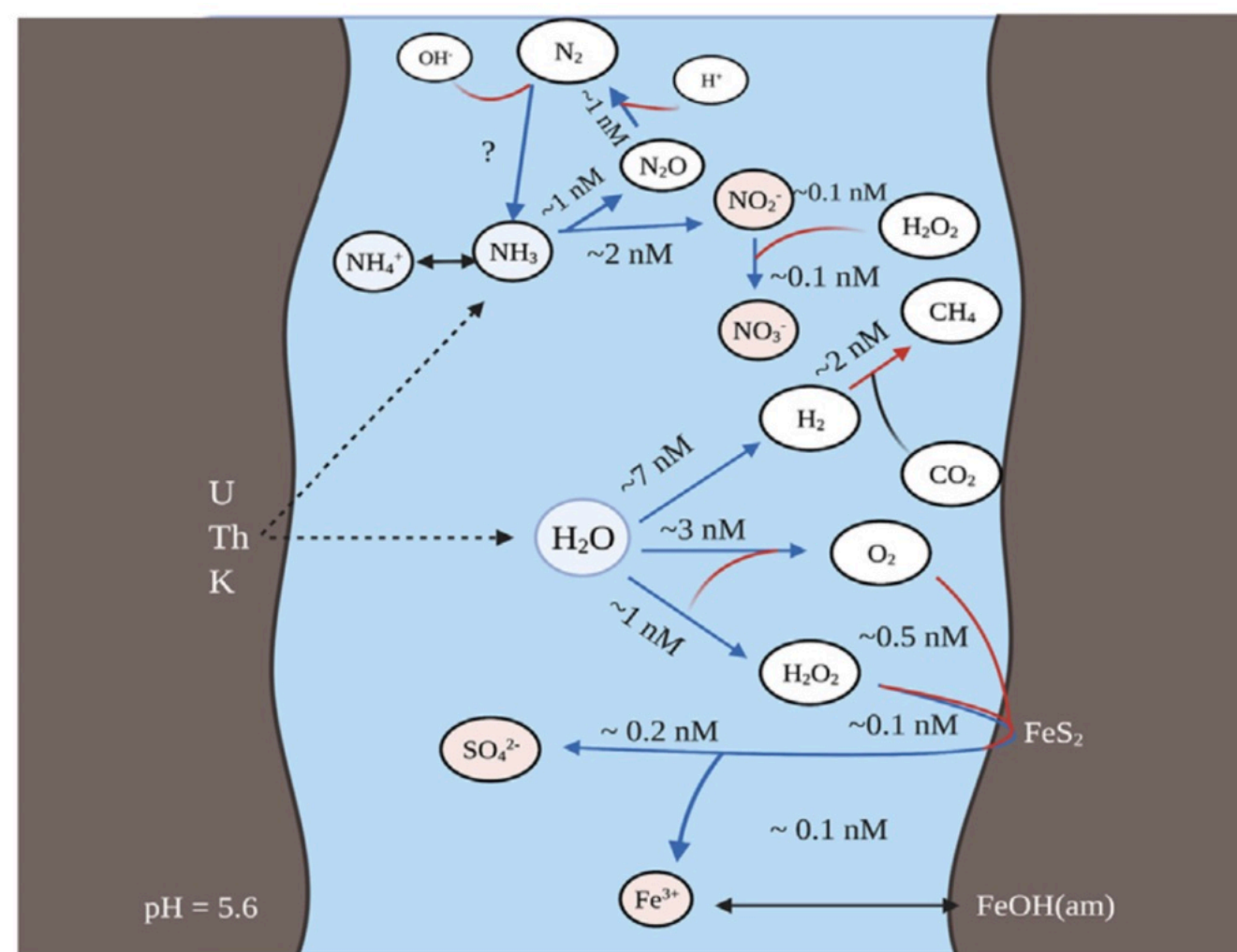


Radiolysis

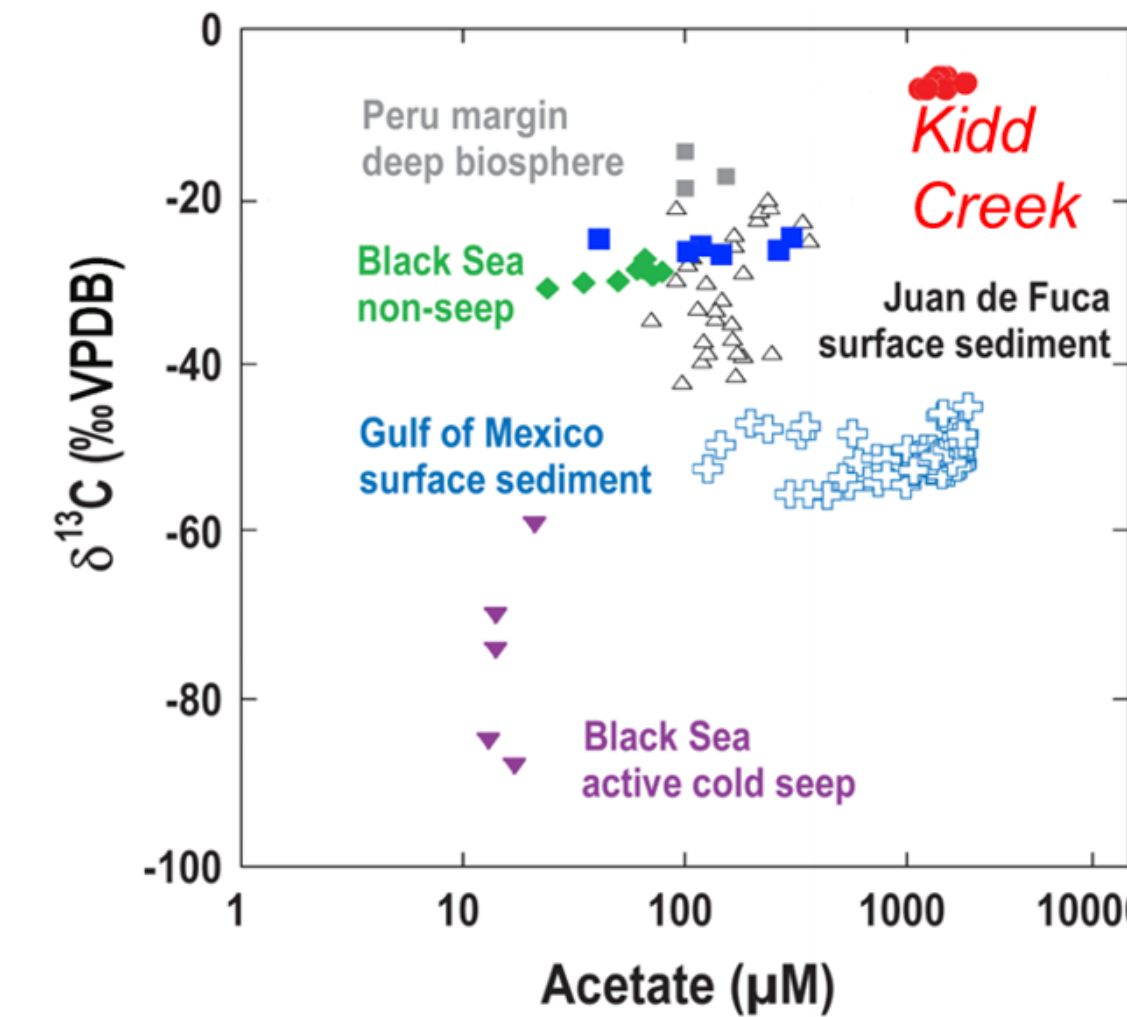
— Organic and inorganic molecules in deep fracture waters via radiolytic reactions



Sherwood-Lollar, Heuer et al, 2021 GCA



Nisson,... Sherwood-Lollar... et al. 2023 GCA



Sherwood-Lollar, Heuer et al, 2021 GCA

Costagliola et al (2017)
J. Phys. Chem.

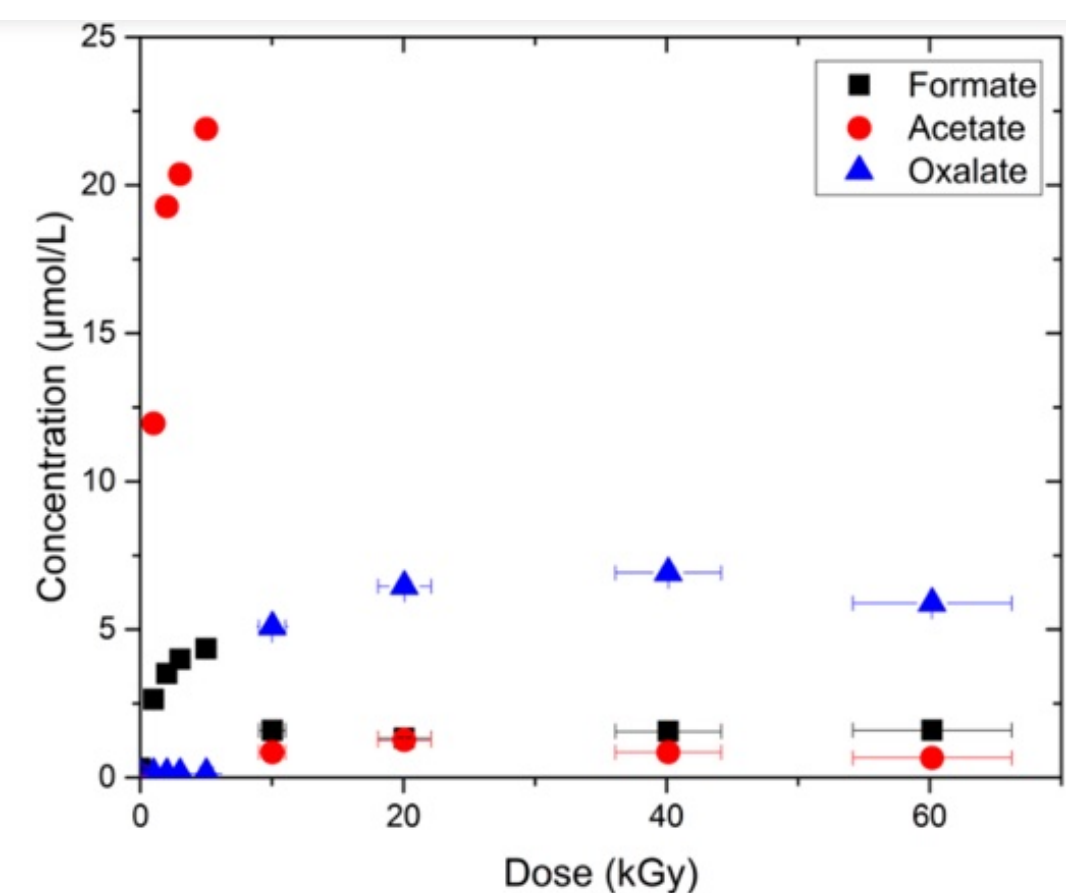


Figure 5. Organic anions production from the irradiation of calcite suspensions B20 (0.2 g) in water (atmosphere: Ar; 60.7 MeV helium ion irradiation; $I = 10$ nA).

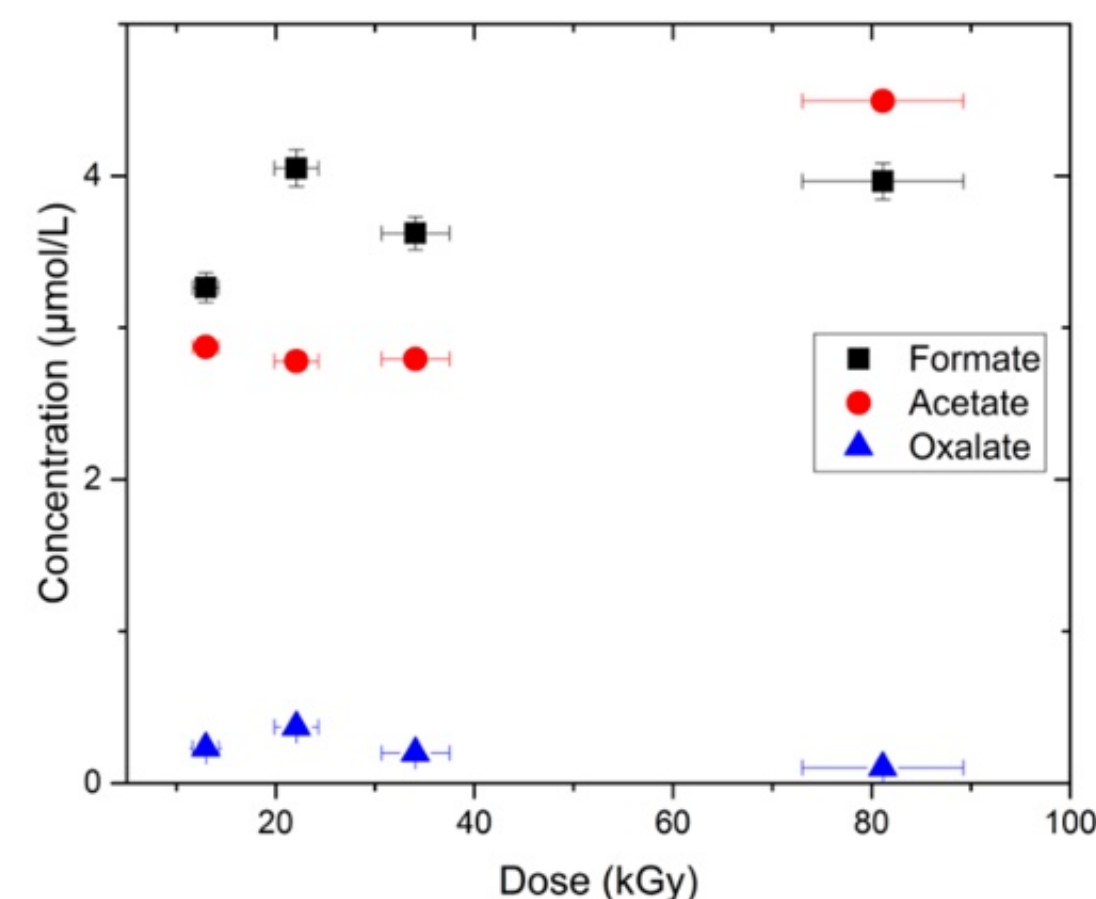
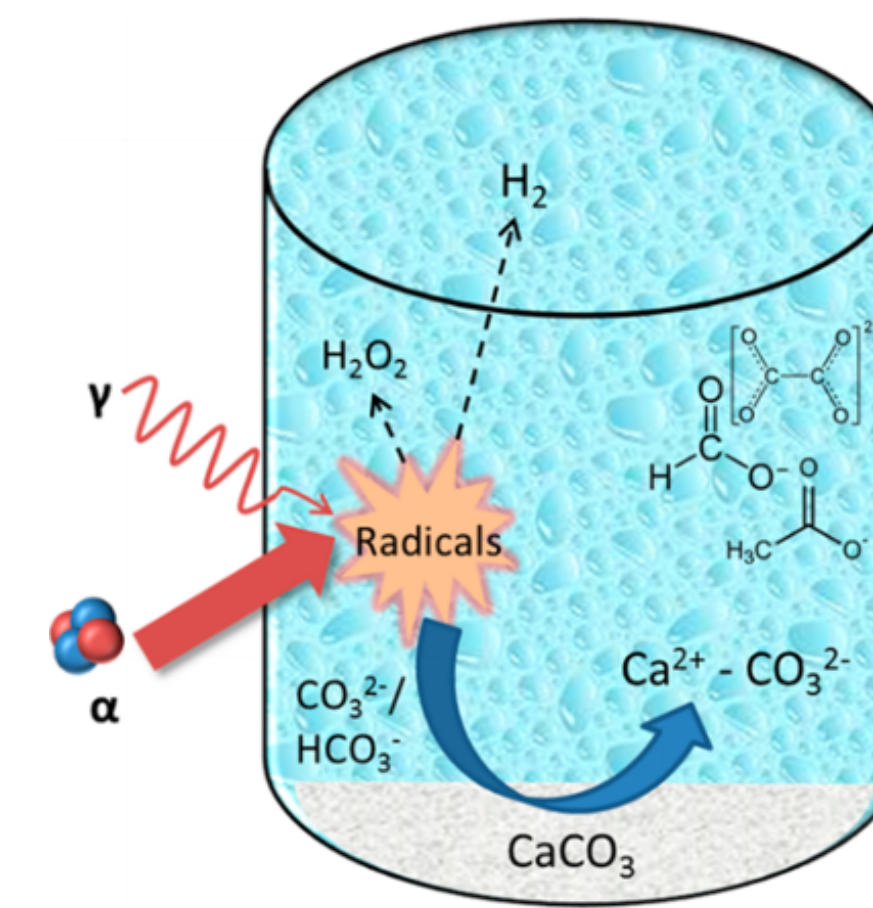


Figure 11. Formation of organic ions after the γ irradiation of calcite powder (D31) (atmosphere: Ar).



Radiolysis — Upcoming experiments at TRIUMF

- Irradiate **“mock” water samples** that mimic the complex geochemistry of the ancient **hypersaline brines from Kidd Creek Mine** (vary sample compositions and pH, brine water-rock slurries, blanks, standards, etc) with:

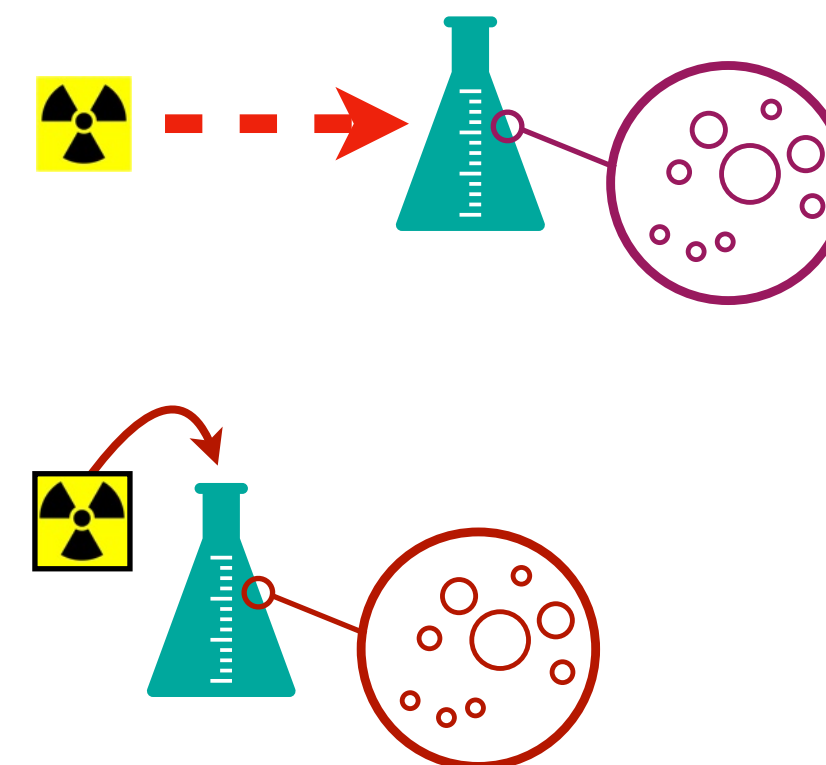
(a) — **Accelerated $^4\text{He}^{+2}$** with **ASPIRE**.
Up to ~50-60 MeV/u (**ISAC-II energies**).

(b) — **Radioisotopes** including alpha-emitters **^{212}Pb (10.6 h)** and **^{225}Ac (9.9 d)** added to samples (**approved expt. L-169**).
(Radiochemistry Hot-Labs, Life Sciences Division).

(c) — **γ -rays** at up to 10 MeV.
(**ARIEL e-linac**).

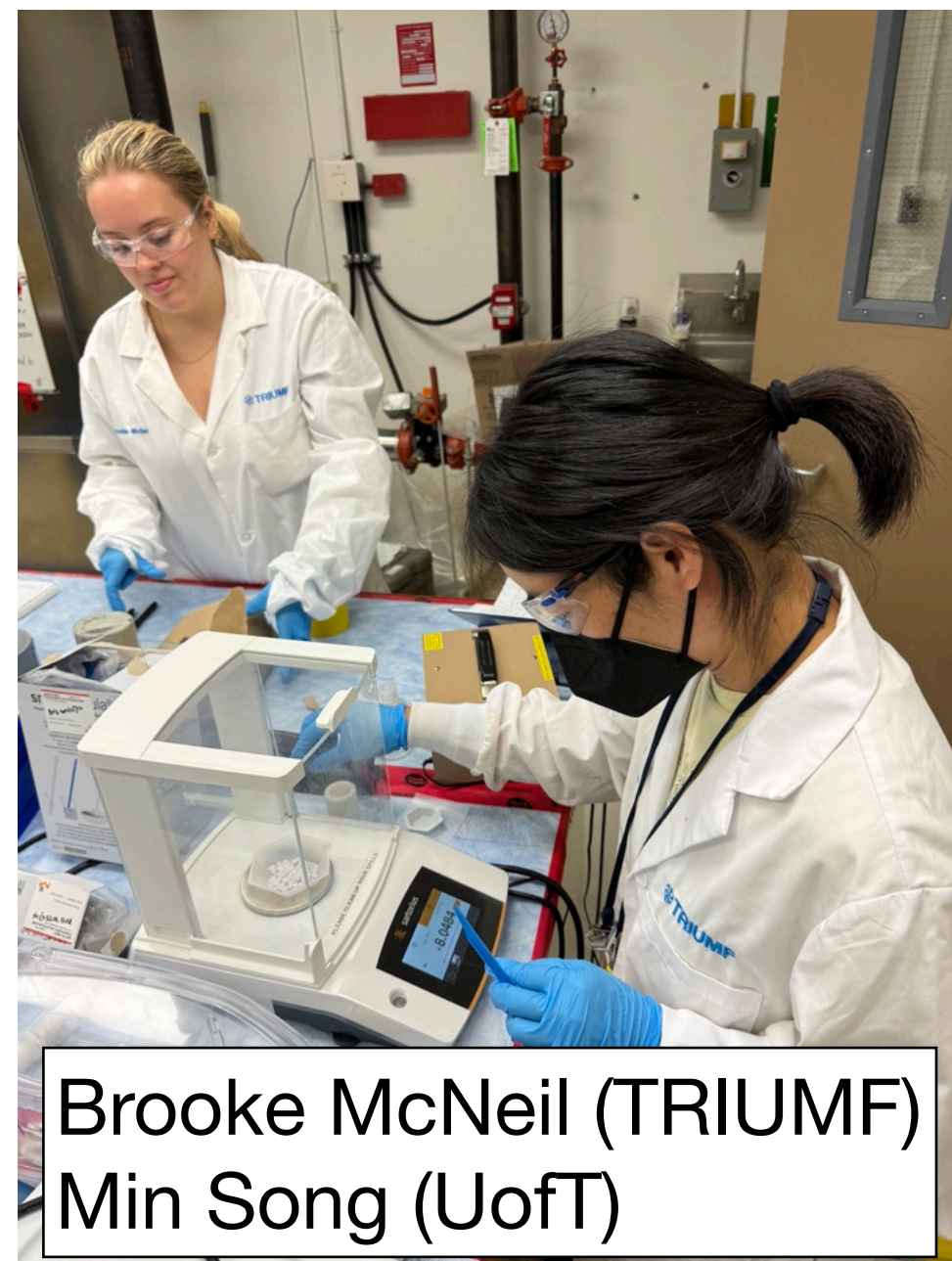
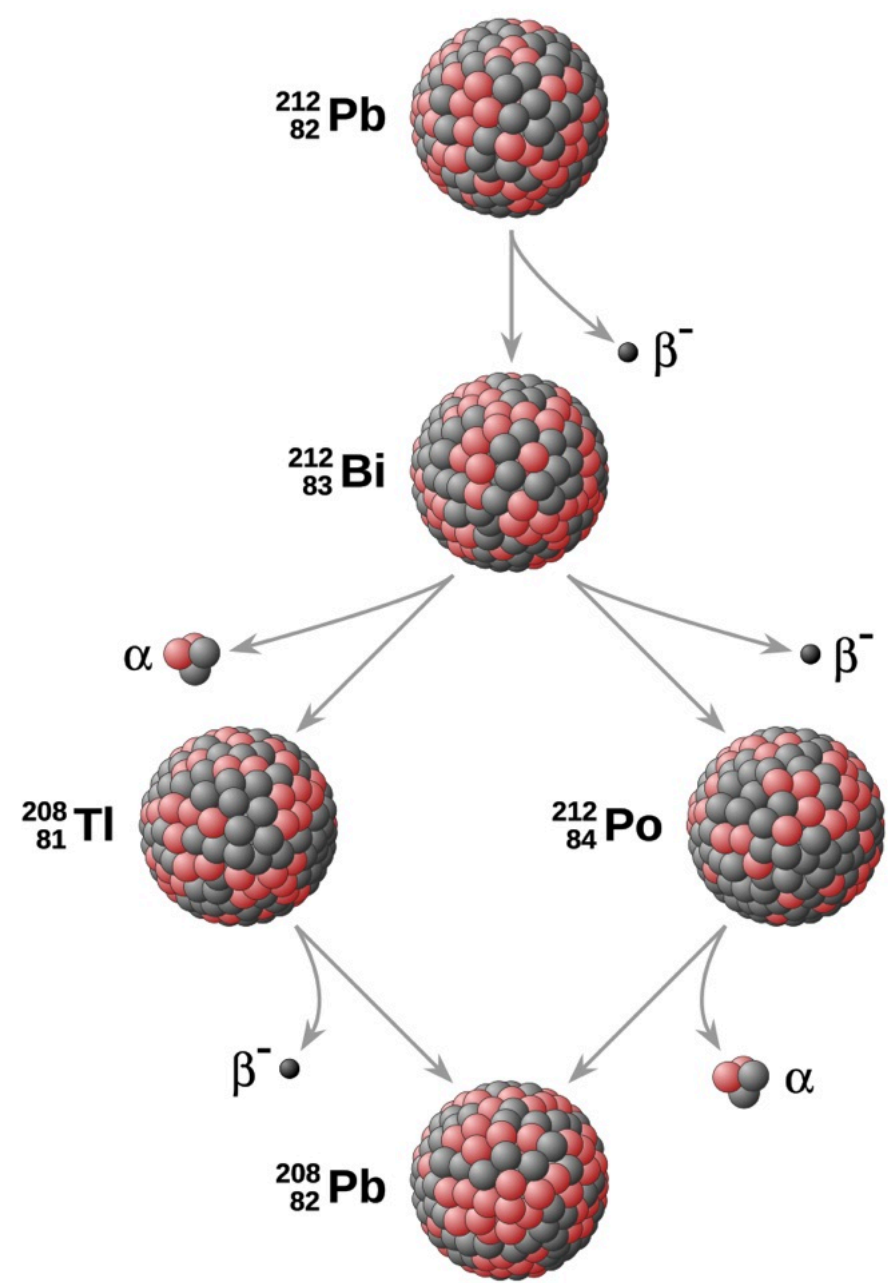
Systematically study possible radiolytic formation of **organic molecules** (i.e., **acetate, formate, other VFAs**) **from abiotic starting materials**.

- Collaboration: C. Charles (PI), Nigel Smith, Aurelia Laxdal (TRIUMF), M. Song (UofT), B. Sherwood-Lollar (co-I, UofT), B. McNeil (TRIUMF), Oliver Warr (uOttawa), Bianca Currie (student, UNB)



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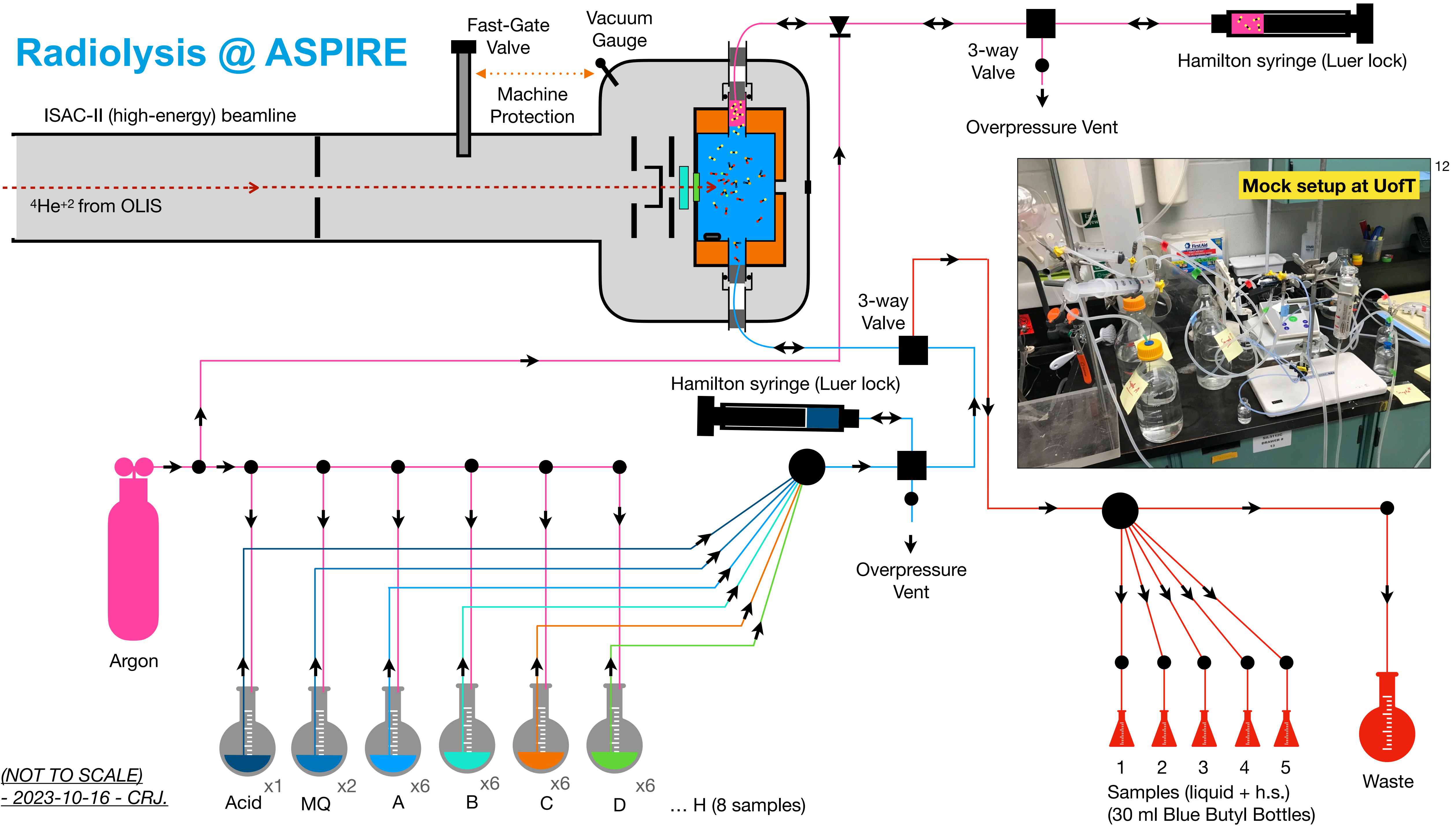
- Radiolytic mechanisms, and production rates as f'n of radiation remain unknown.**
- Stable isotope fractionation factors (i.e. $\delta^{13}\text{C}$) completely unknown.**
- Nuclear/radioactive inputs/outputs very unclear.**
- Conditions necessary to create/destroy complex molecules in extreme isolated planetary environments?**



- **Expt. L-169** is now underway in the Life Sciences Radiochem labs (RCR2).
- Completed 1st round of ^{212}Pb injections into mock-samples about 10 days ago.
- **First results of organic molecule production, or not, expected soon!**



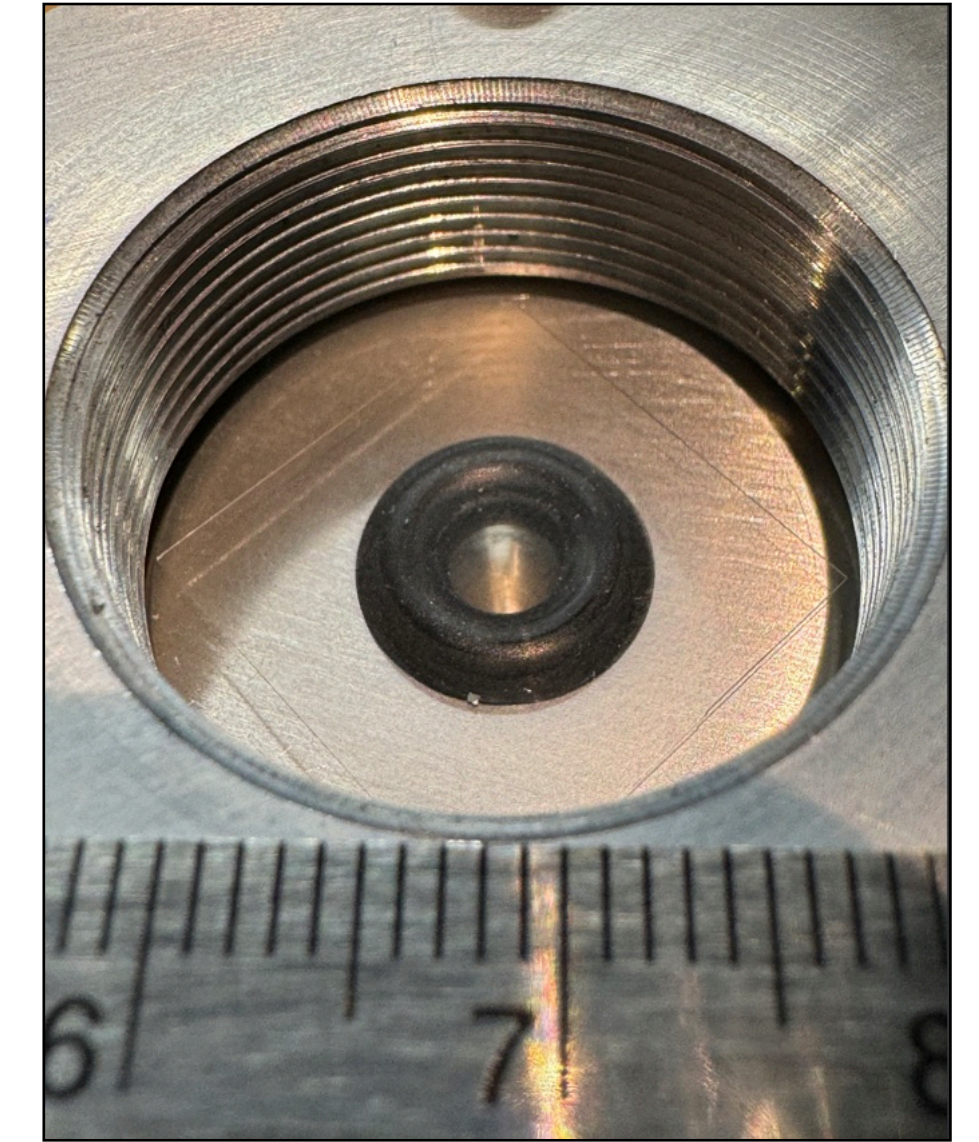
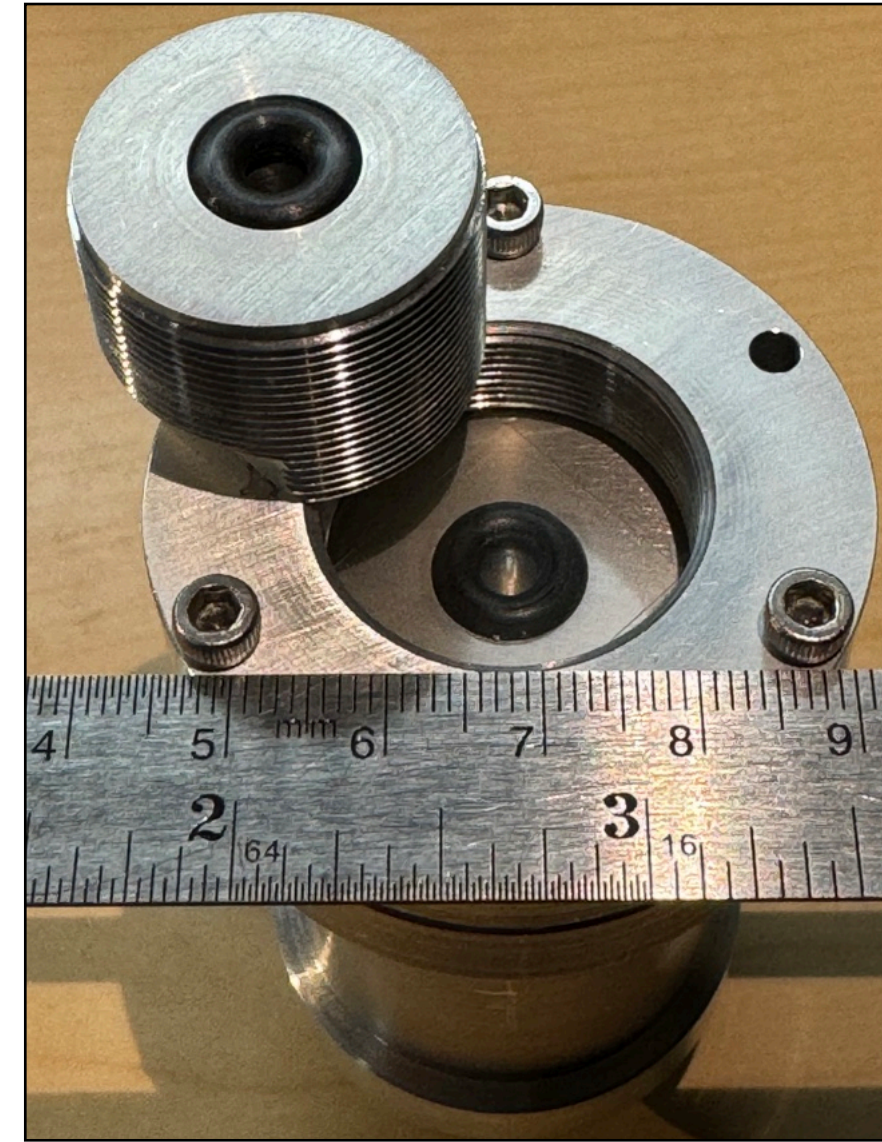
Radiolysis @ ASPIRE



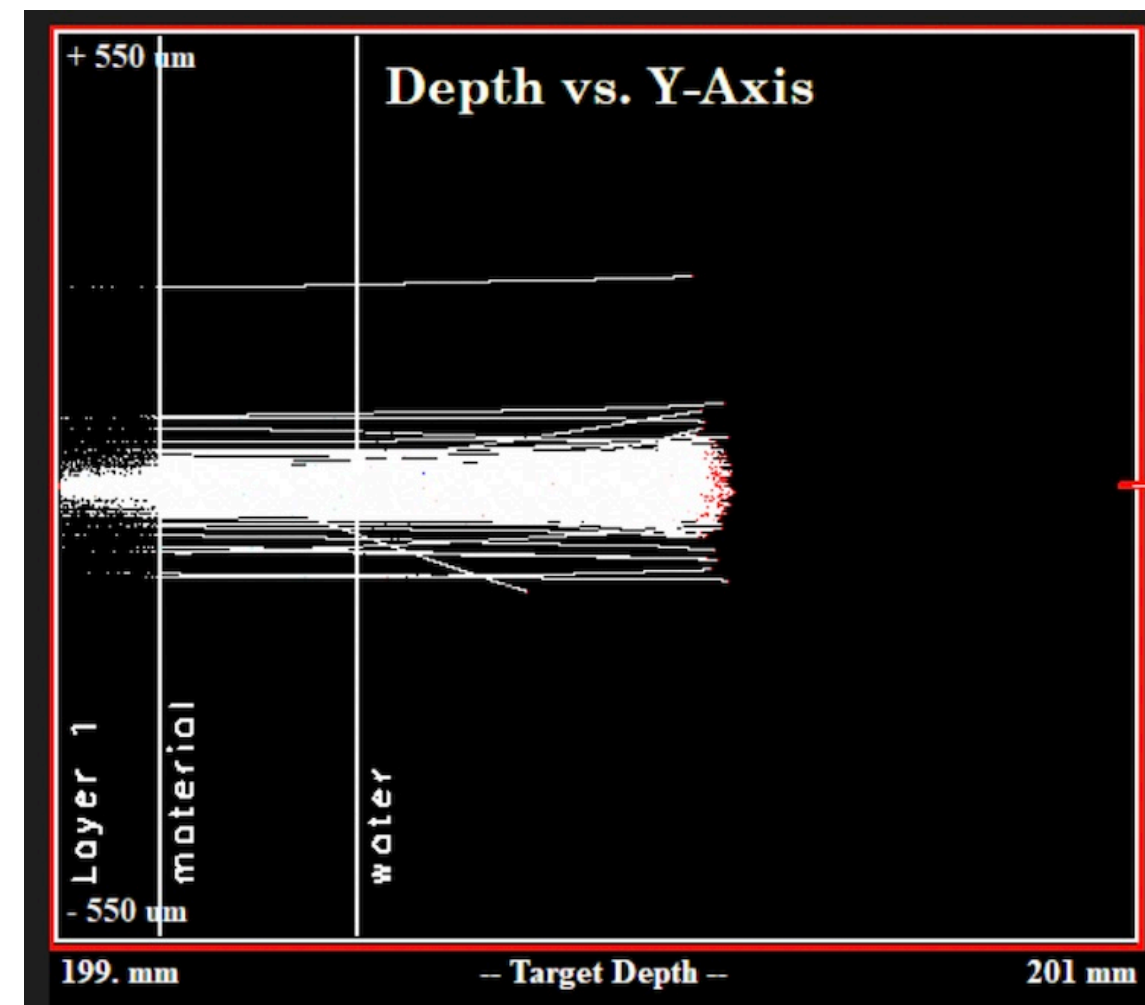
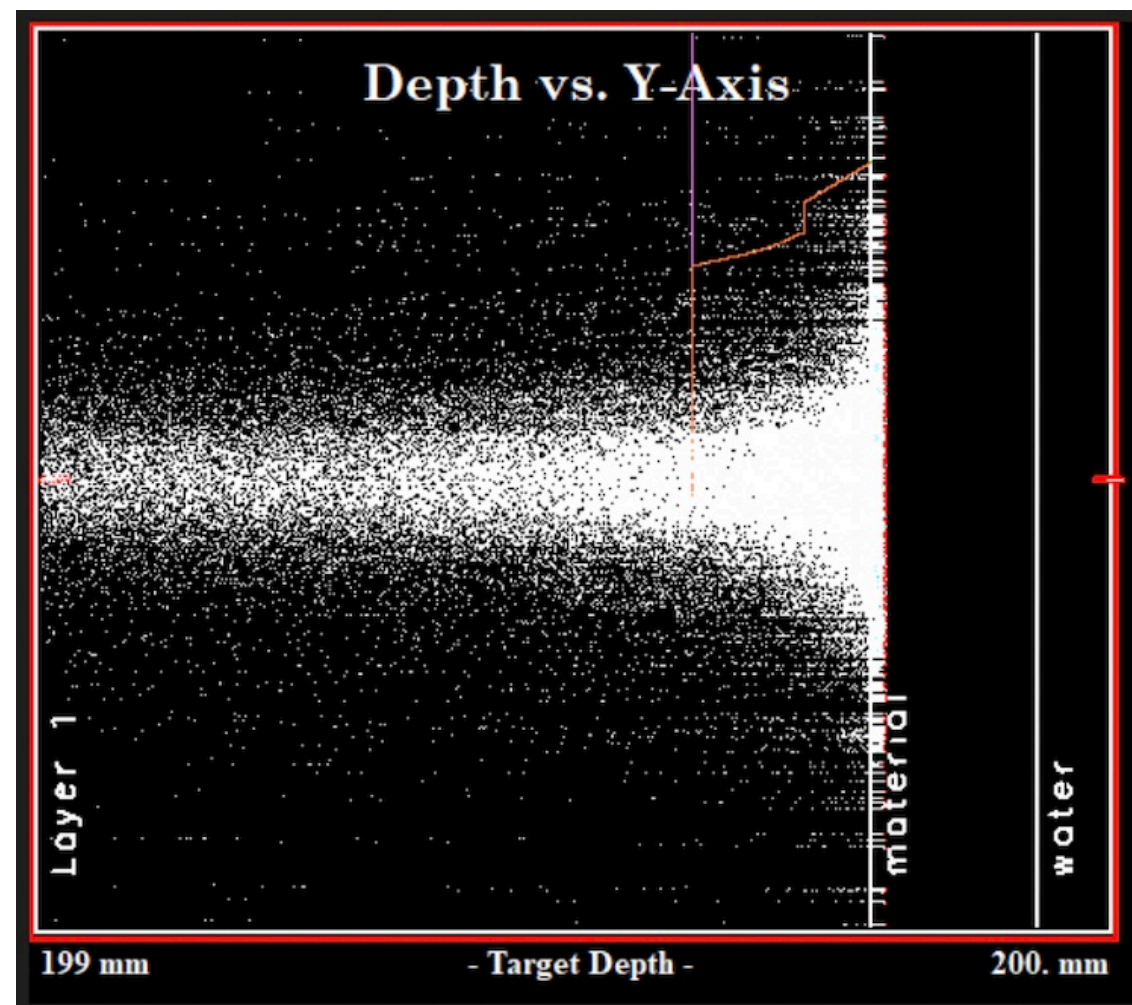
(NOT TO SCALE)
- 2023-10-16 - CRJ.

Radiolysis @ ASPIRE

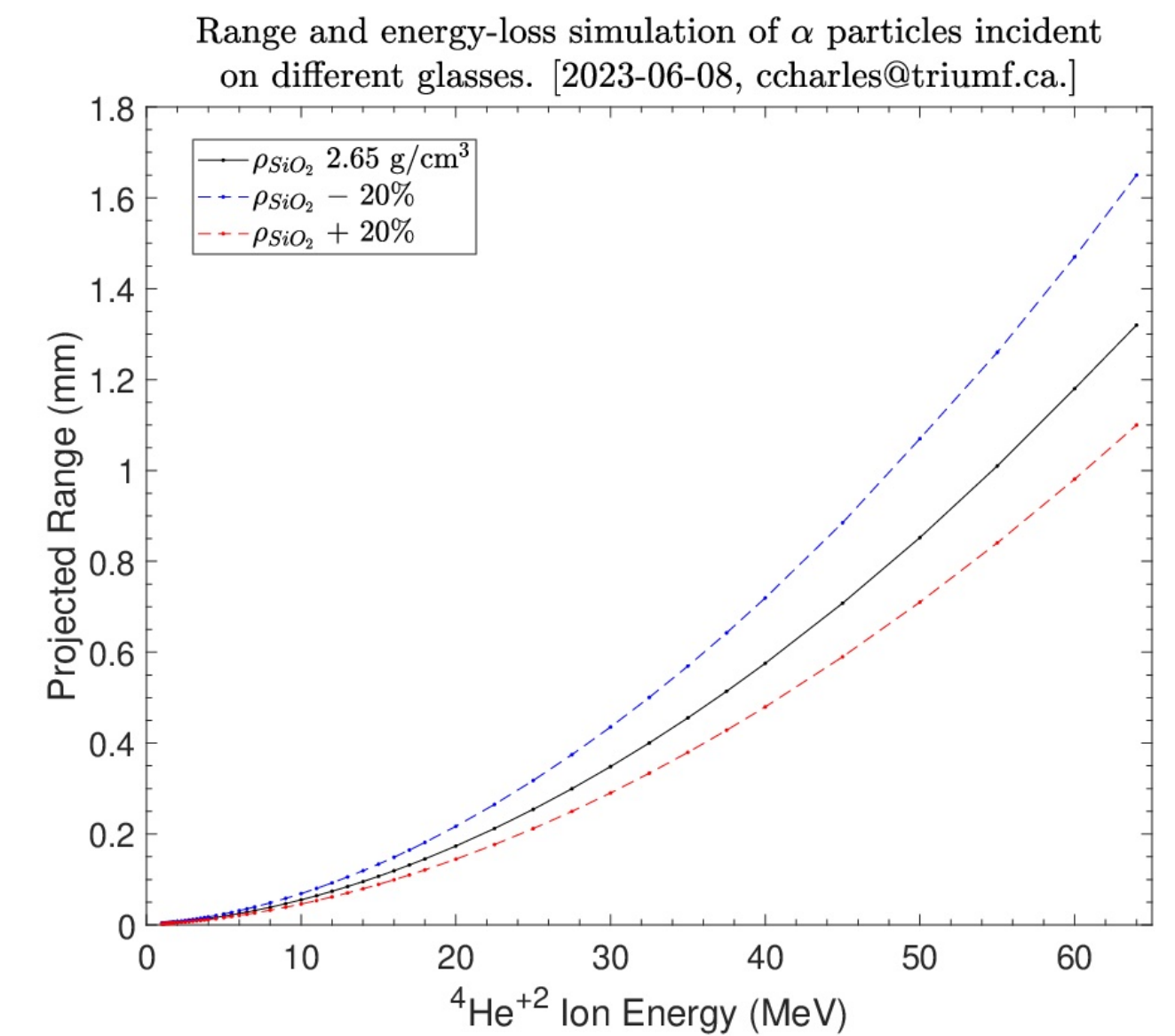
Water irradiation chamber v2.2; Custom machined by **Marco Lovera & Brian Minato** (TRIUMF)



SRIM/TRIM simulations of windows (re: **Dave Prevost & Peter Kunz**, TRIUMF)



Ariana Pearson (UWO Astrophysics, B.Sc. student)



Interstellar Ice Astrochemistry

- **Icy dusty grains** → site of the most complex chemistry in the interstellar medium (ISM).
- Bombardment of **ice** with cosmic radiation leads to low-temperature radical chemistry on grain surfaces.
 - ✓ Organic molecules formed are potential precursors to biological molecules.
- These basic low-temperature physiochemical processes are complex and poorly understood, poorly unconstrained.
- Understanding ISM molecular **formation**, **destruction**, and **abundance** on **ices**, provides insight into galactic chemical evolution, with implications to development on life on Earth and beyond.

Cosmic Ray-Driven Radiation Chemistry in Astrochemical Models

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² Max-Planck-Institut für extraterrestrische Physik, D-85748 Garching, Germany
³ Institute for Theoretical Chemistry, University of Stuttgart, Pfaffenwaldring 55, 70569 Stuttgart, Germany
⁴ Harvard-Smithsonian Center for Astrophysics, 60 Garden Street, Cambridge, MA 02138, USA
⁵ Department of Astronomy, University of Virginia, Charlottesville, VA 22904, USA

Abstract

Cosmic rays are widely known to have significant physiochemical impact on interstellar sources. In addition, laboratory astrophysics experiments have indicated that cosmic ray interactions with dust grain ice mantles could lead to astrochemically relevant species, including complex organic and prebiotic molecules [1].

In spite of the growing body of experimental work on interstellar radiation chemistry, incorporating cosmic ray-driven reactions and processes into astrochemical models has proven challenging, in part because of a lack of relevant data for many species now included in chemical networks. Recently [2], we have developed a general method of estimating radiochemical yields (G-values), rate coefficients, and decomposition pathways for species that have not been studied in detail in the laboratory in this context. Here, we will describe the derivation and application of our method, as well as point to much needed areas for future development in astrochemical radiation chemistry modeling.

Suprathermal Species

Example Network

$A \rightarrow A^* + e^- \rightarrow B^* + C^*$
 $A + B^* \rightarrow \text{Products}$

$\Rightarrow \frac{dn(B^*)}{dt} = k_{ion2}n(A) - v_0^B n(B^*) - k_{sr}n(A)n(B^*)$

The Fate of Suprathermal Species in Our Network

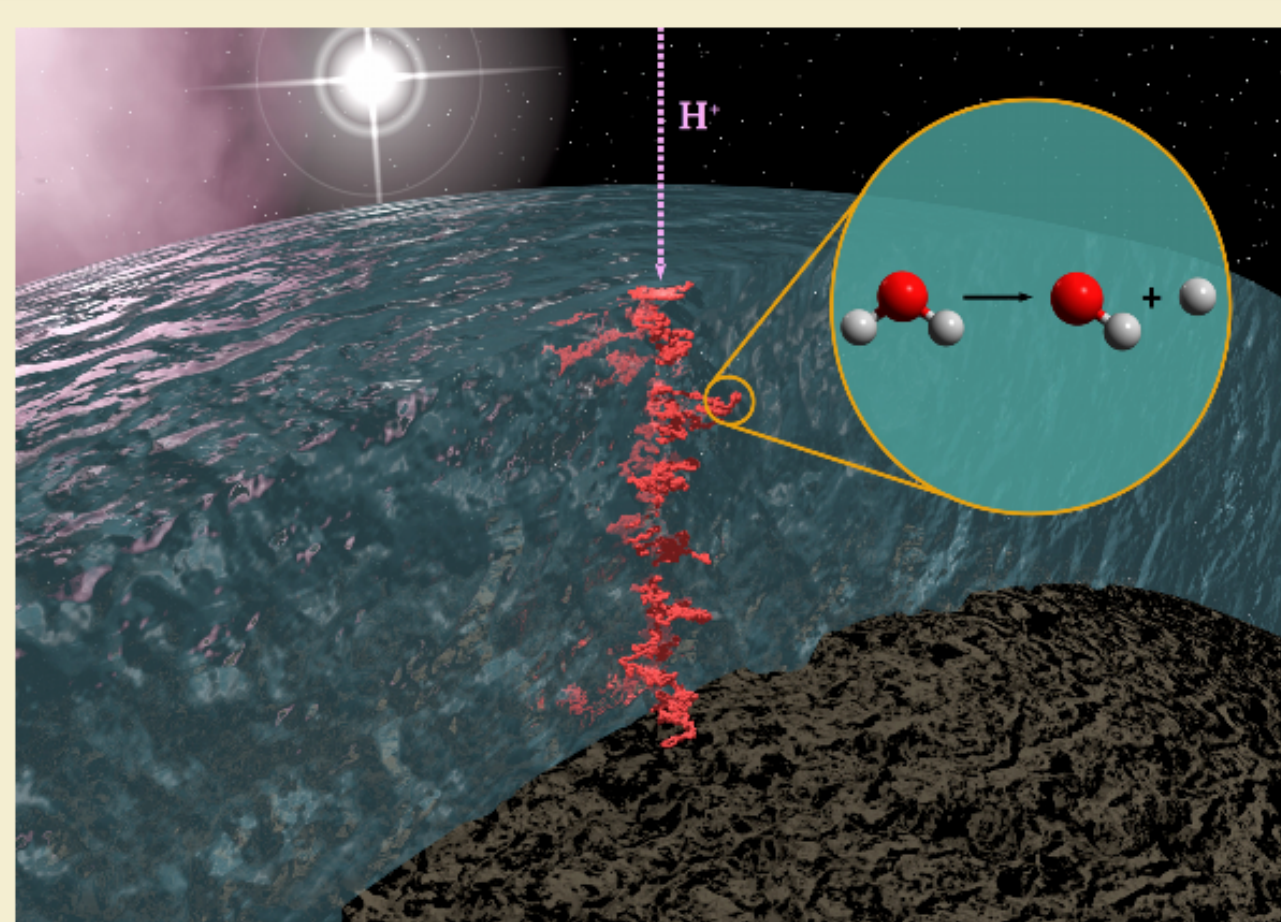
Formation of B^*

Possibility 1: Quenching

$$v_0^B = \left[\frac{2 N_{\text{site}} E_b^B}{\pi^2 m_B} \right]^{1/2}$$

Possibility 2: Reaction

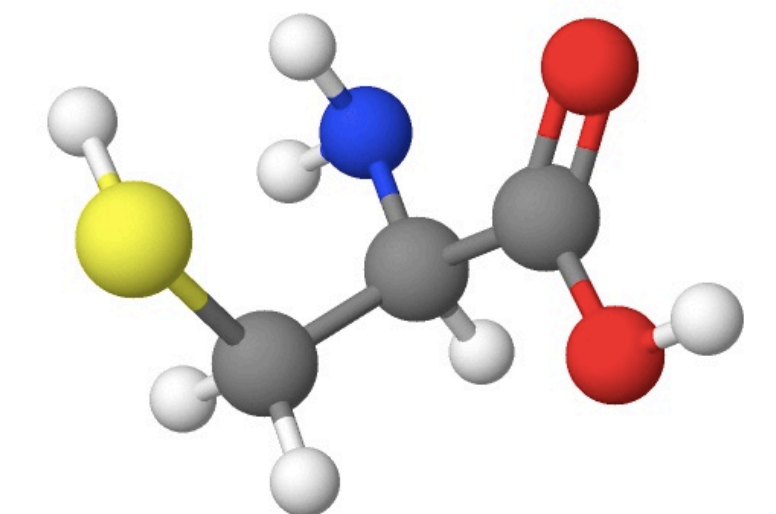
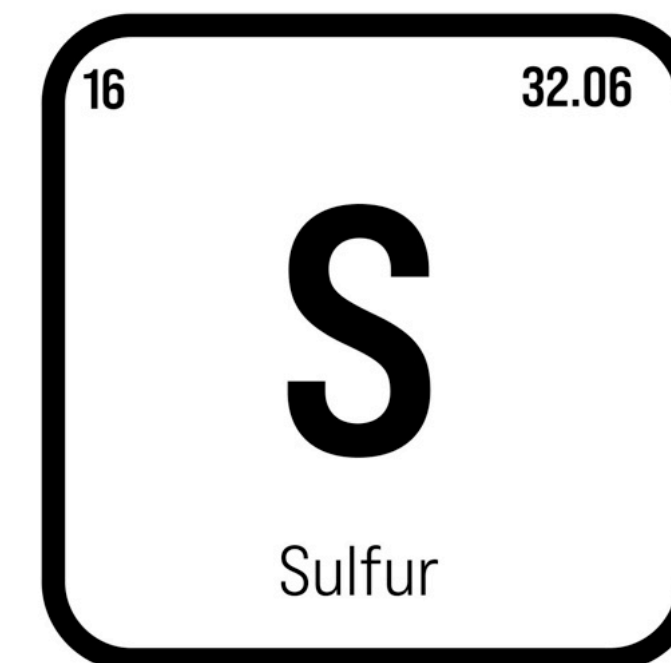
$$k_{sr} = f_{br} \left[\frac{v_a^A + v_a^B}{N_{\text{site}} n_{\text{dust}}} \right]$$



Interstellar Ice Astrochemistry

— Sulphur Depletion Problem (SDP)

- SDP: most **sulphur** in the observable ISM is missing or as yet undetected (only ~1% is accounted for).
- ***Does exposing icy mineral samples to energetic cosmic ray analogues cause interactions between ices and minerals, and partition sulphur?***
 - ***If S-bearing species are formed in ices during irradiation, this supports a longstanding astrochemical hypothesis that S is locked away in icy dusty ISM grains.***
- Implications to understanding the origins of biological **S** in the universe — e.g. in amino acids (cysteine, or vitamins like thiamine) + delivery to the early Earth.



Interstellar Ice Astrochemistry

— Sulphur Depletion Problem (SDP)

Dartois et al — Astronomy & Astrophysics 2023, 671, A156.

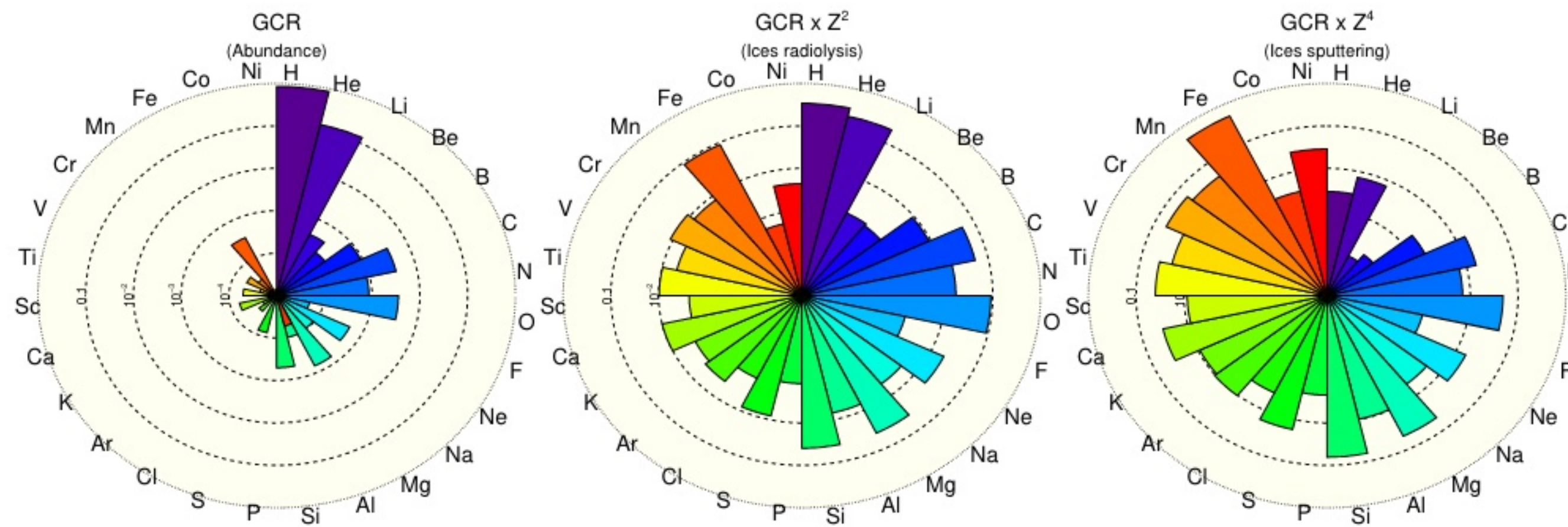
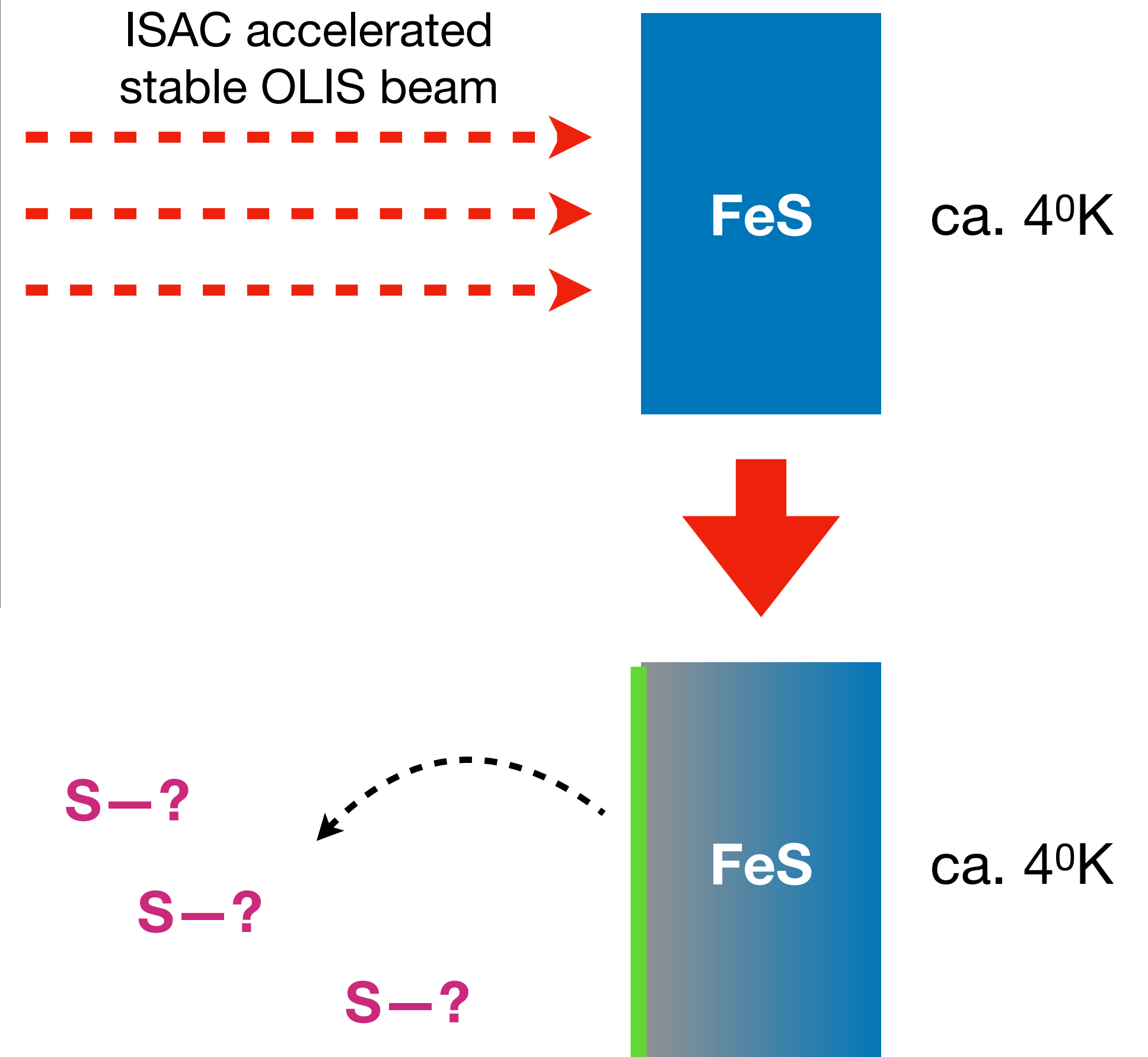


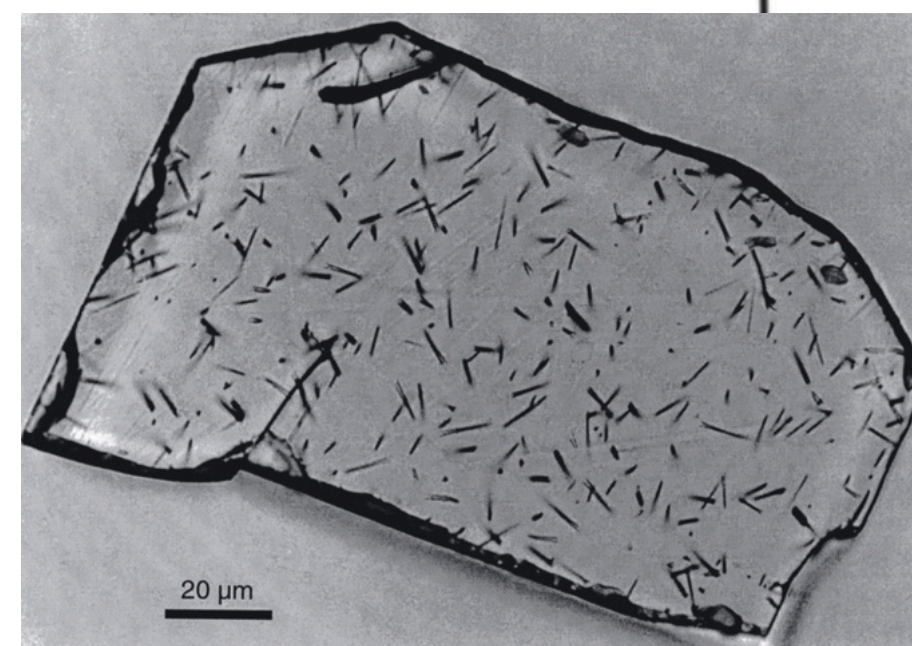
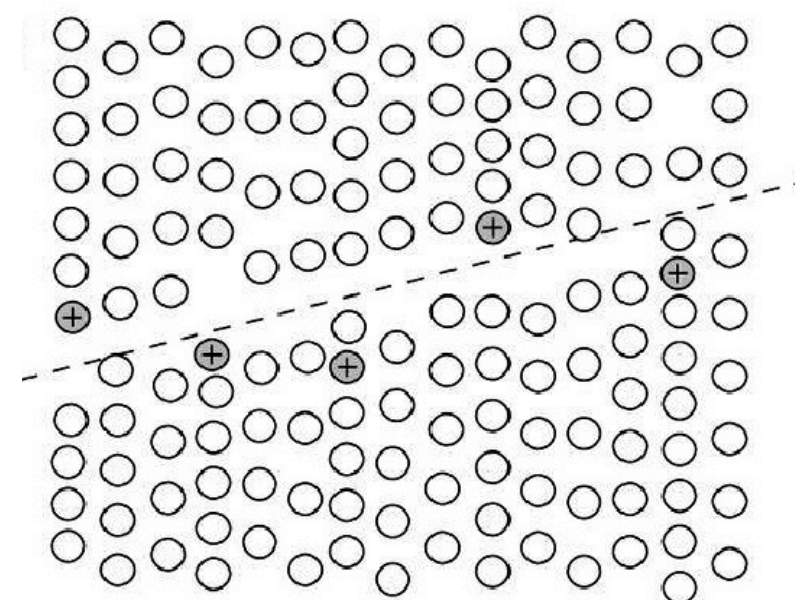
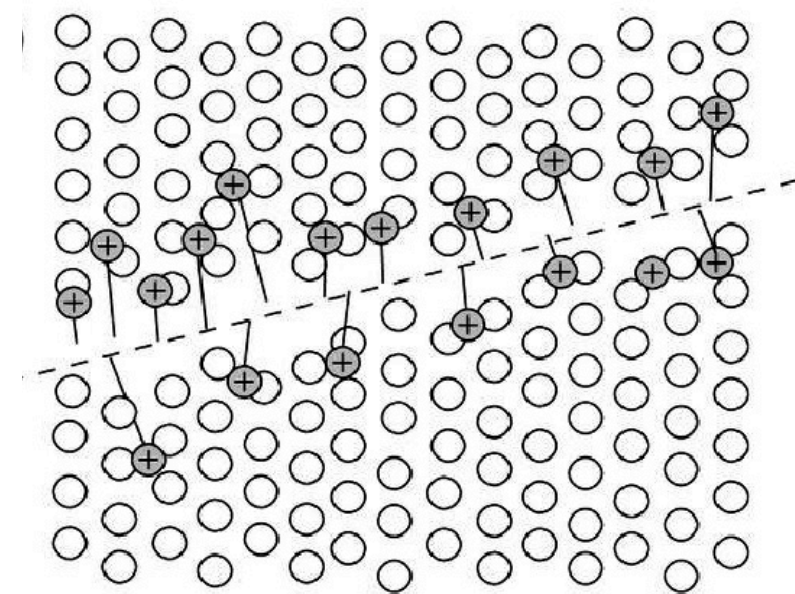
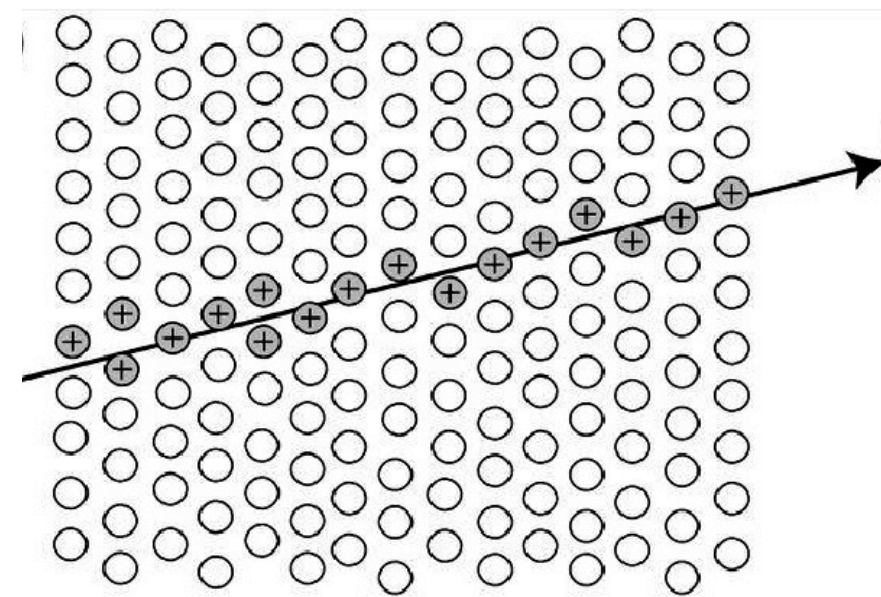
Fig. 1. Influences of the different elements in processes associated with cosmic rays represented as fractional proportions on a log scale. Left: Galactic cosmic-ray abundance. Middle: relevance of the cosmic-ray elements in processes proportional to Z^2 , i.e. typically the radiolytic destruction of ices constituents. Right: relevance of the cosmic-ray elements in processes proportional to Z^4 , i.e. typically associated with the electronic sputtering mechanisms discussed in this article.

ASPIRE — cryogenic ISM analog sample

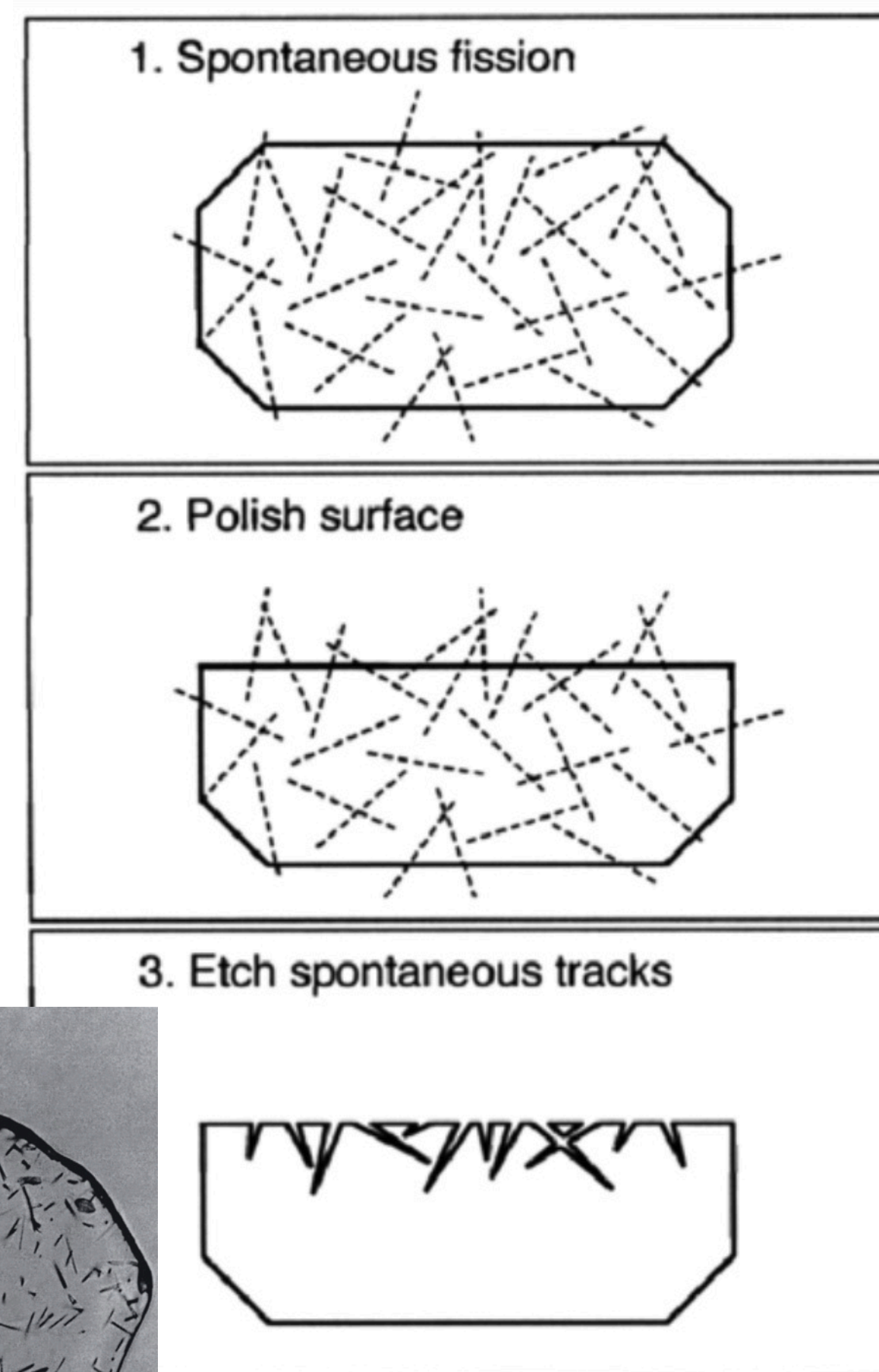


- Collaboration: C. Charles (TRIUMF) + UBC Chemistry: Ilsa Cooke (PI), Elsa Yuan (Ph.D. student), Arie Irving-Hughes (B.Sc. student).

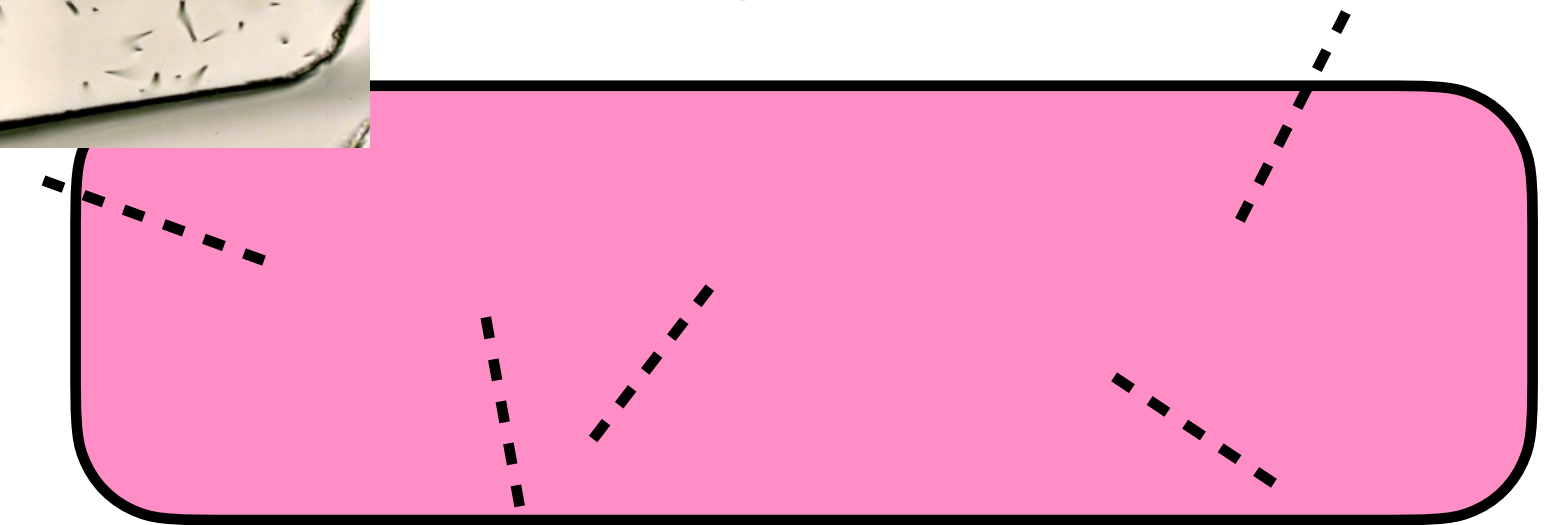
Fission-Track Age-Dating Enhancement



Old Sample

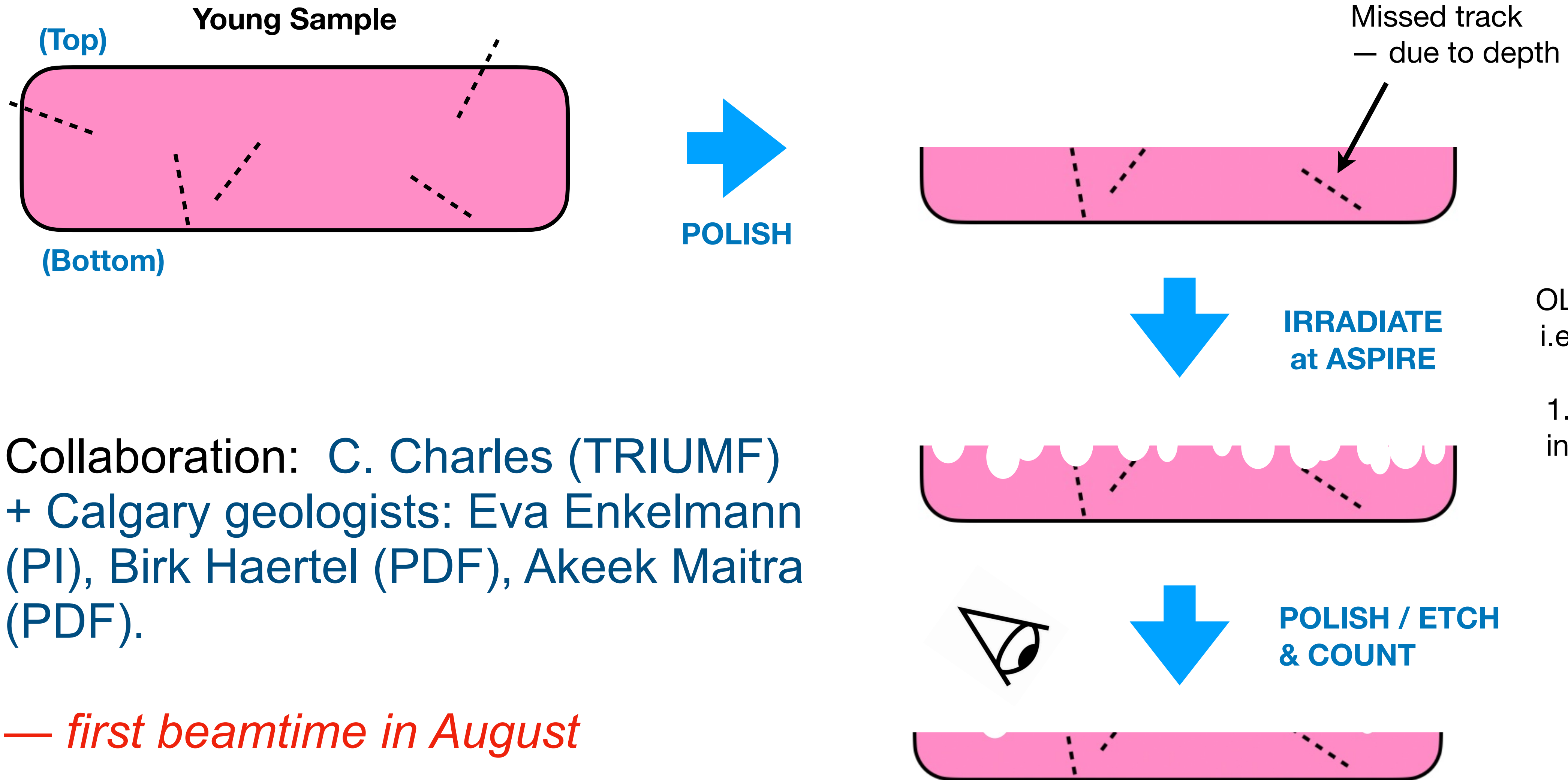


Young Sample



- **Apatite** — $\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{Cl}, \text{OH})$ is usually well suited to FT-age dating because it contains ^{238}U , ^{235}U when the crystal forms from its magma.
- However, young samples (< 2 Myr) are much more difficult or sometimes impossible to date because:
 - ✓ **Less time for U damage tracks to accumulate (sparse, weakly observable).**
 - ✓ **Poor counting statistics of sparse tracks from chemical etching.**
 - ✓ **Poor precision & poor accuracy ages.**
- **Enhancing sparse/weak tracks would provide better counting statistics = higher precision and accuracy ages for younger minerals.**

Fission-Track Age-Dating Enhancement



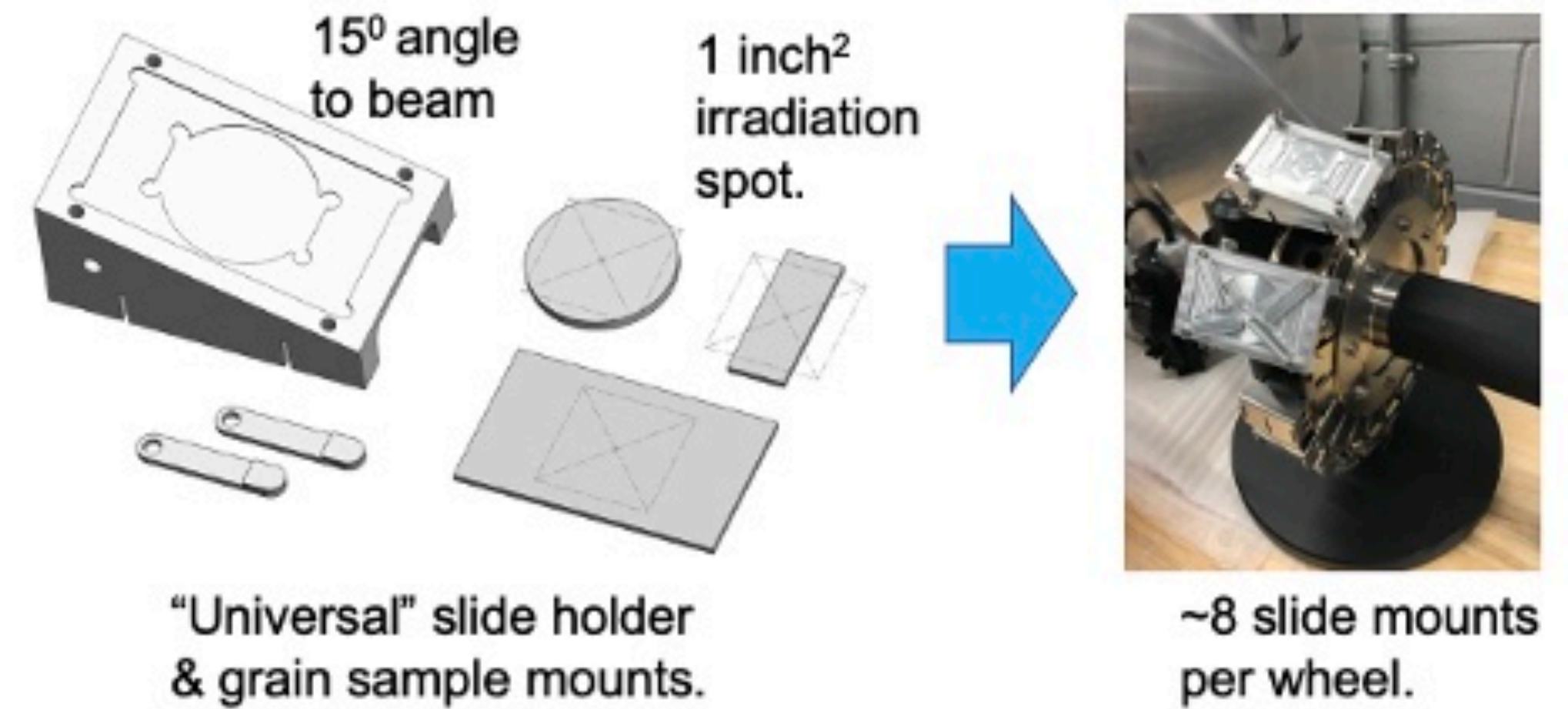
- Collaboration: C. Charles (TRIUMF) + Calgary geologists: Eva Enkelmann (PI), Birk Haertel (PDF), Akeek Maitra (PDF).

— *first beamtime in August*

Fission-Track Age-Dating Enhancement



(a)



(b)

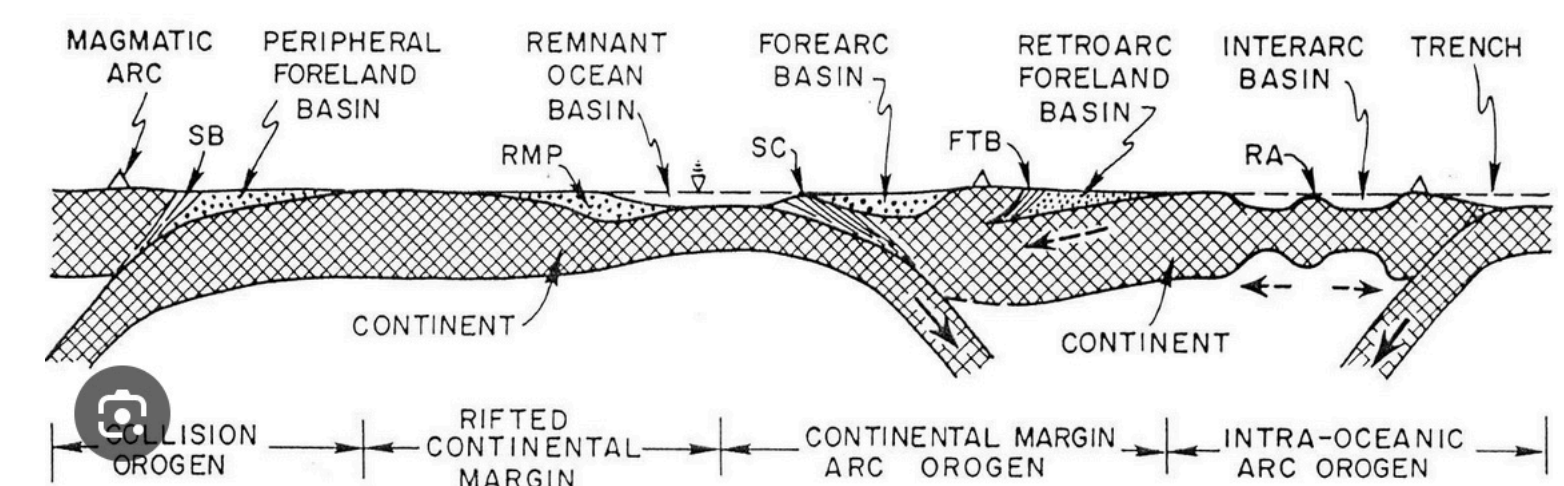
FIGURE 2 – (a) microscope (46 x 27 x 1.2 mm) slide showing mounted and flat-polished apatite grains in epoxy; (b) custom “universal” sample holder, which hold the microscope slides in (a) at 15° to the incident beam as well as two additional slide sizes being either (i). 25 x 2 mm disk, or (ii). 50.8 x 76.2 x 1.2 mm rectangle. The universal holder(s) are shown mounted to the sample wheel in (b), which are then inserted into the ASPIRE vacuum chamber in Fig. 1(e).

Fission-Track Age-Dating Enhancement

- Aim to obtain improved FT ages for many **geologic processes**, i.e.:
 - ✓ Evolution of mountain belts.
 - ✓ Erosional processes & sedimentation.
 - ✓ Thermal history/evolution of the continental crust.
 - ✓ Timing of volcanic events.
 - ✓ Tracing of archaeological artifacts.
 - ✓ Evolution of lakes & basins.
 - ✓ Age dating strata (difficult).
 - ✓ Applications to other minerals (sphene, zircon, micas, glasses, garnet, epidotes).



Various stolen web images



Conclusions

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- ASPIRE = new experimental facility at TRIUMF-ISAC.
- Irradiating wide varieties of “planetary material samples” (solids, liquids, gases) to study leading-edge problems in planetary sciences, astrochemistry, early solar system cosmochemistry, geology, origins of life,, requiring light or heavy accelerated beams.
- 3 upcoming ASPIRE projects — to increase the breadth & depth of TRIUMF science with accelerated beams & radioisotopes, to address novel, exciting, leading-edge problems in other fields:
 - ✓ Fission-track dating enhancement of geologic mineral grains.
New “regime” for fission-track age-dating in geology (1st beam time in August).
 - ✓ Astrochemistry on icy interstellar grains.
Production/degradation of complex molecules on grain surfaces in space.
 - ✓ Radiolysis of water & origins of life.
Implications to natural production of organic molecules by radiation.
- TRIUMF’s unique accelerated beam infrastructure at ISAC, together with “wet chemistry” experiments in the Life Sciences radiochemistry labs, offers a perfect world-class combination of possibilities for novel interdisciplinary projects across fields.

