Measurements of ^{222}Rn, ^{220}Rn, and CO_2 Emissions in Natural CO_2 Fields in Wyoming: MVA Techniques for Determining Gas Transport and Caprock Integrity

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U.S. Department of Energy

National Energy Technology Laboratory
Carbon Storage R&D Project Review Meeting
Developing the Technologies and
Infrastructure for CCS

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Presentation Outline

- Benefits to the Program
- Project Overview
- Technical Status
  - Field Work
  - Overview
  - Lab Work
- Accomplishments to Date
- Summary

Benefit to the Program

- Carbon Storage Program goals being addressed.
  - Develop and validate technologies to ensure 99 percent storage performance.
- Project Benefits Statement.
  - The purpose of this project is to evaluate methods, already used for natural volcanic systems, for assessing the integrity of caprock formations. The methods use quantitative measurements of CO_2 flux and naturally occurring Radon (Rn) isotopes to determine the source of the CO_2 (escaped from a reservoir or degassed from shallow soil).

Project Overview: Goals and Objectives

- To provide training opportunities for two graduate students and one undergraduate student in skills required for implementing and deploying CCS technologies.
- To perform fundamental research to advance the sciences of Monitoring, Verification, and Accounting (MVA).
- To take ^{222}Rn, ^{220}Rn, and CO_2 measurements at natural CO_2 analogues in Wyoming as well as in the laboratory to evaluate and calibrate the use of naturally occurring Rn isotopes for assessing the integrity of caprock formations.

Project Overview:
Goals and Objectives

- How the project goals and objectives relate to the program goals and objectives.
  - Measurements of Rn and CO_2 can constrain source and timescale of CO_2 fluxes. Supports the program goal of ensuring 99 percent storage performance.

Project Overview:
Goals and Objectives

- Success criteria for student training goals
  - Two graduate students complete advanced degrees
  - One BS student perform undergraduate research.

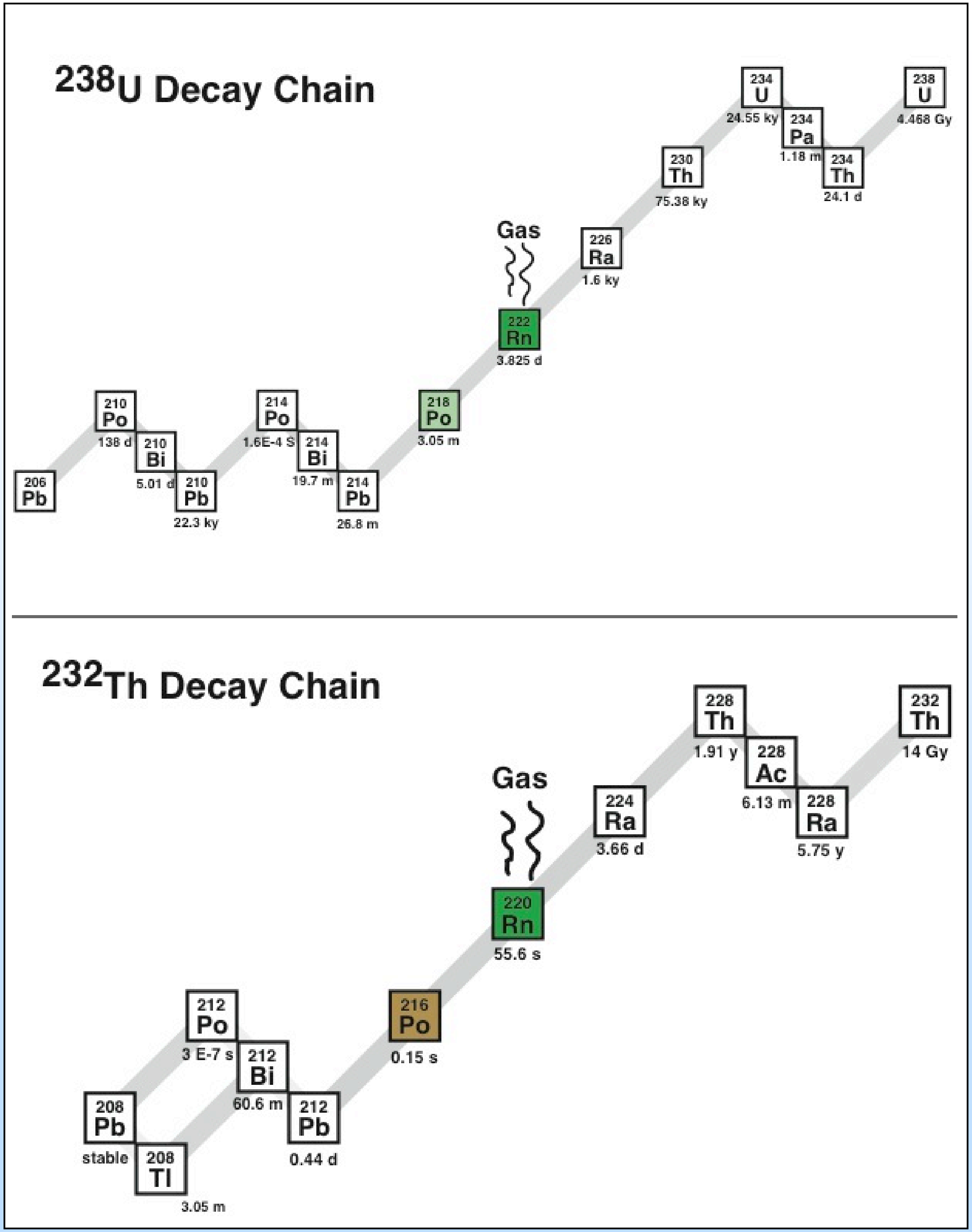
Project Overview: Goals and Objectives

- Success criteria for MVA research
  - Perform integrated Rn and CO_2 measurements at natural CO_2 analogues in Wyoming
  - Replicate field conditions in the laboratory to evaluate effects of reactions at depth on Rn degassing.
  - Determine the suitability of using measurements as MVA techniques for CCS.

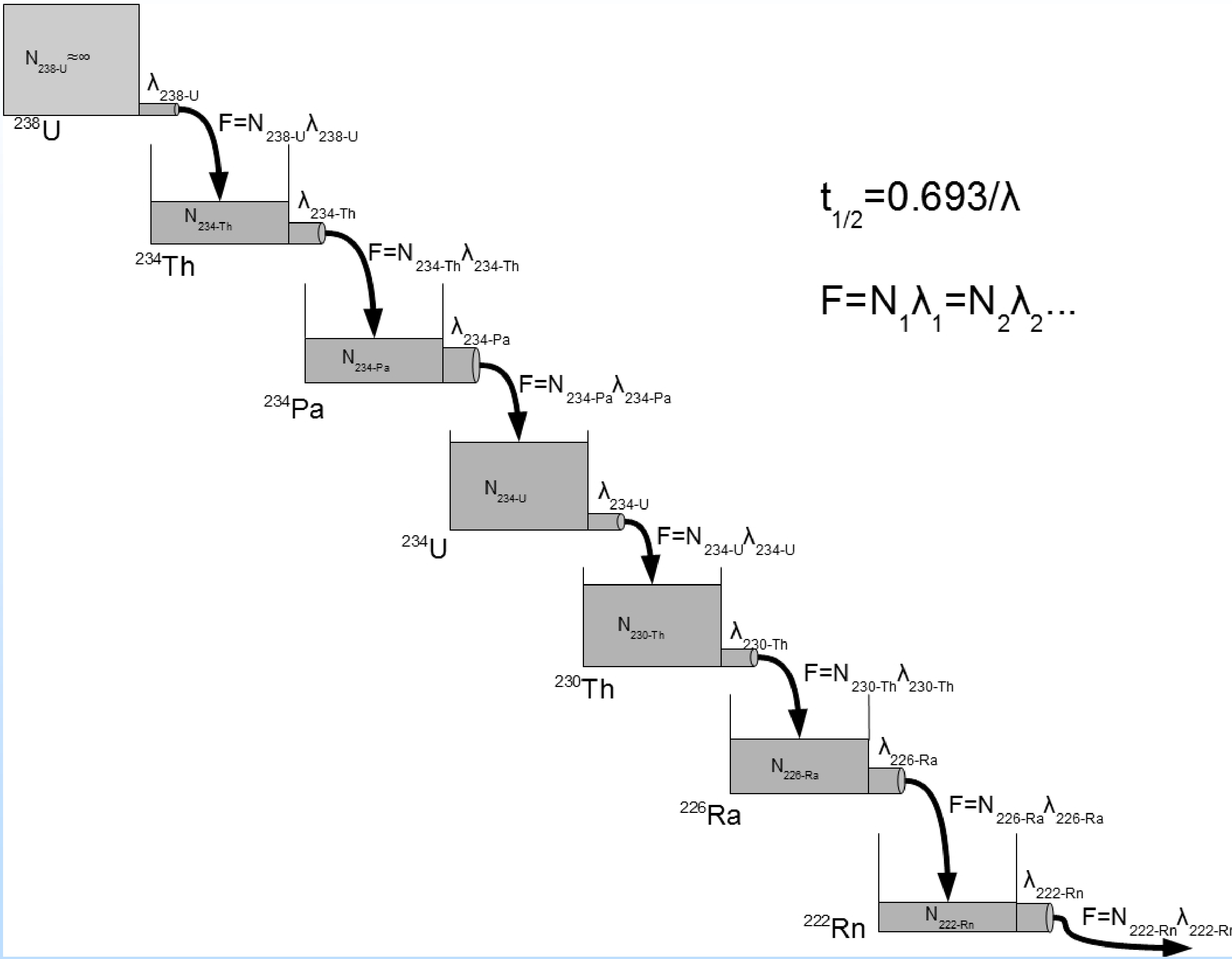
Technical Status

- Field Measurements in Wyoming
  - La Barge CO_2 Field (SW Wyoming)
  - Yellowstone (high pH, high temperature, challenging)
  - Thermopolis (similar to Yellowstone but less extreme)
- Overview
  - U-Series Basics
  - Secular Equilibrium
  - CO_2 and Rn
- Lab work
  - Rad7 CO_2 corrections (Lane-Smith and Sims, 2013)
    - Evaluate grain size control on alpha recoil

U Series Decay Chains

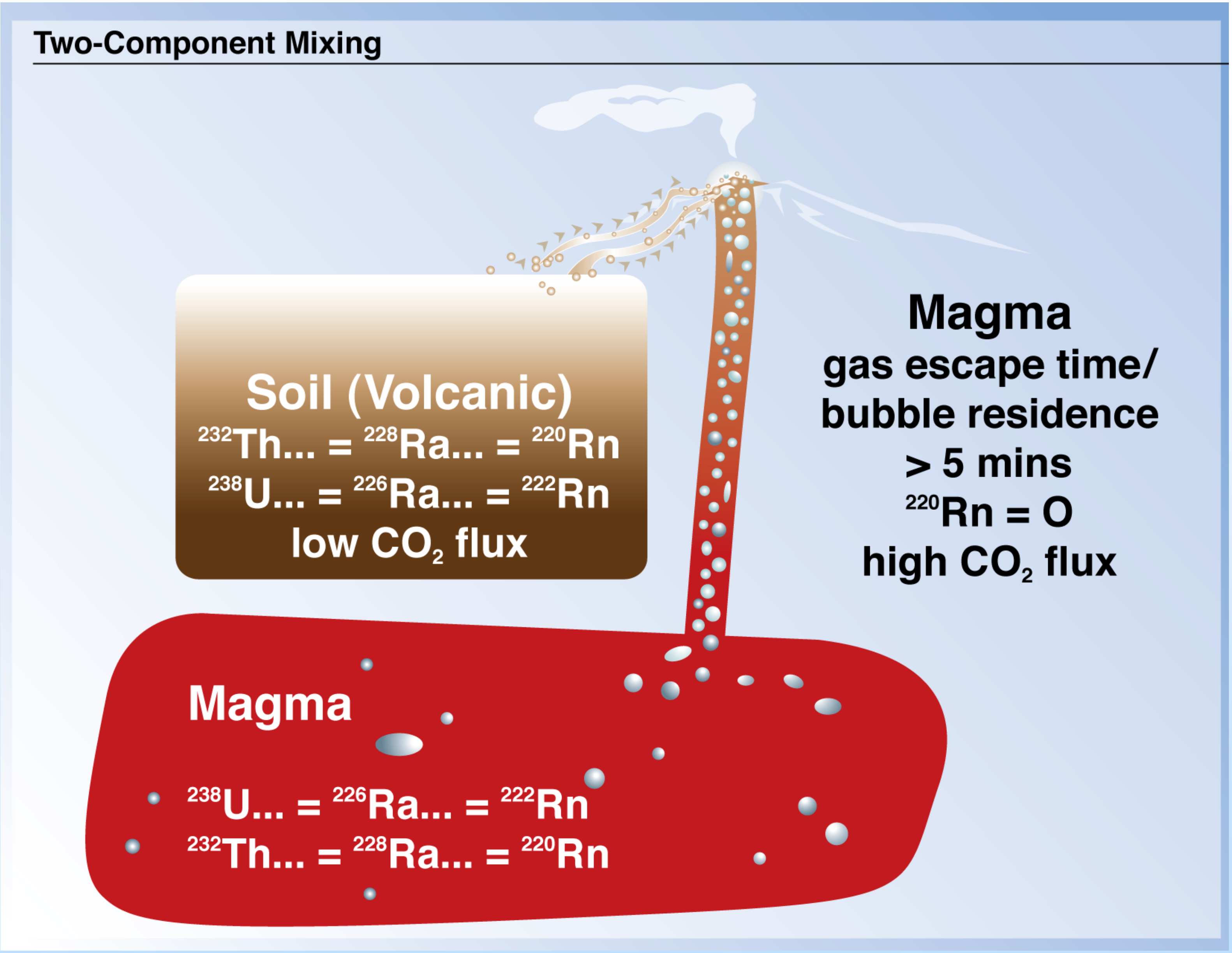


Secular Equilibrium

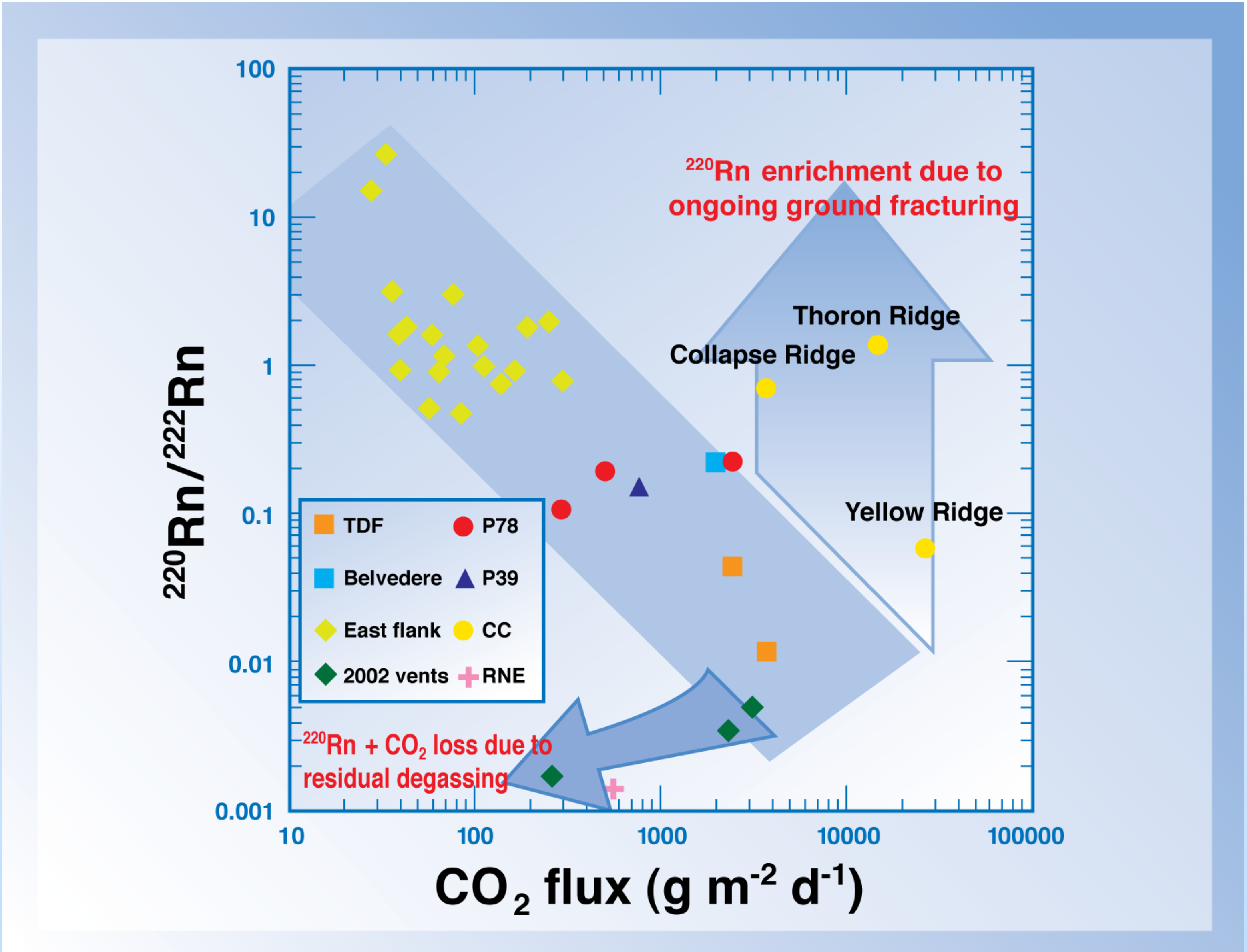


$$t_{1/2}=0.693/\lambda$$

$$F=N_1\lambda_1=N_2\lambda_2\ldots$$



Giammanco et al., 2007

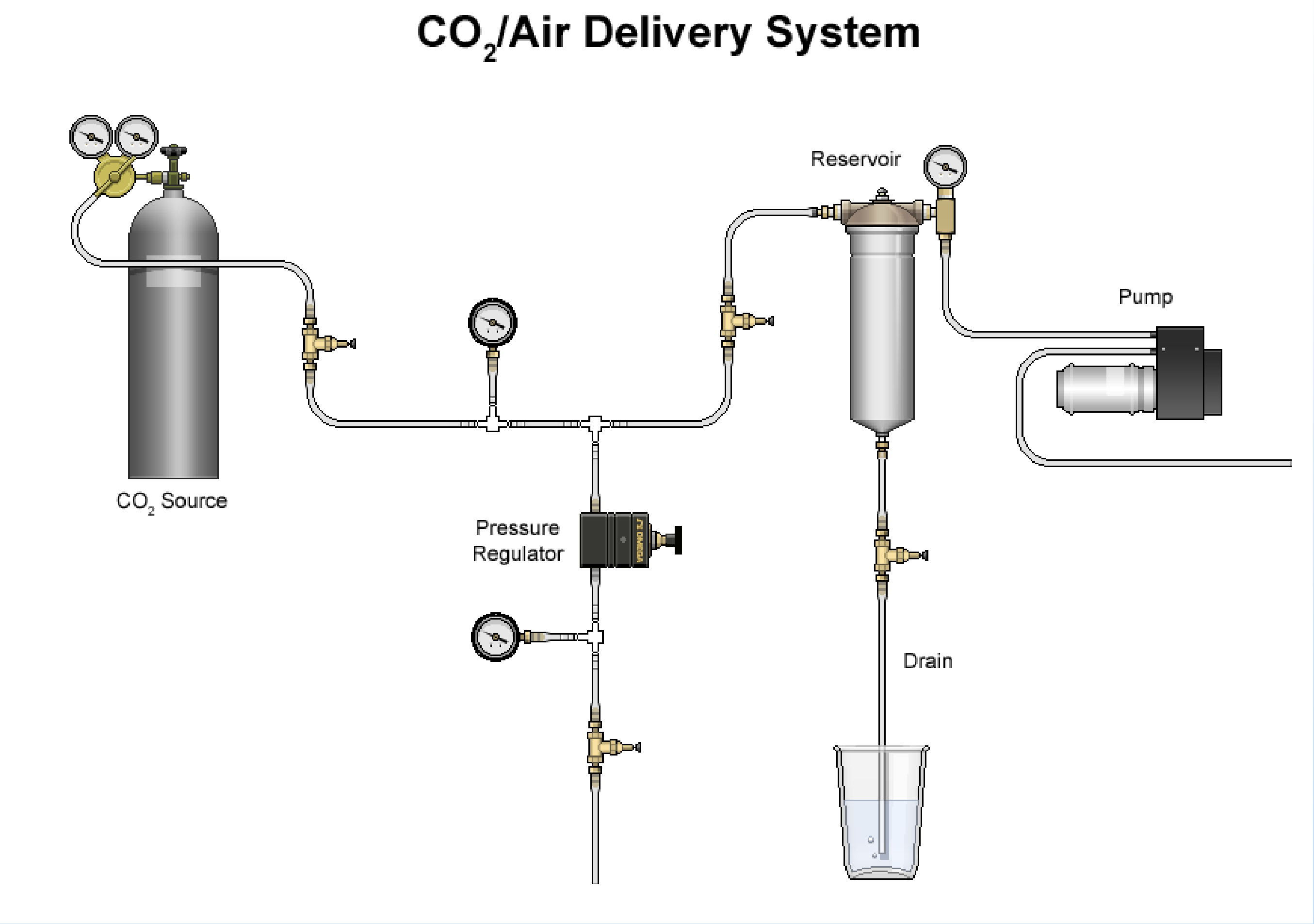


Giammanco et al., 2007

Technical Status

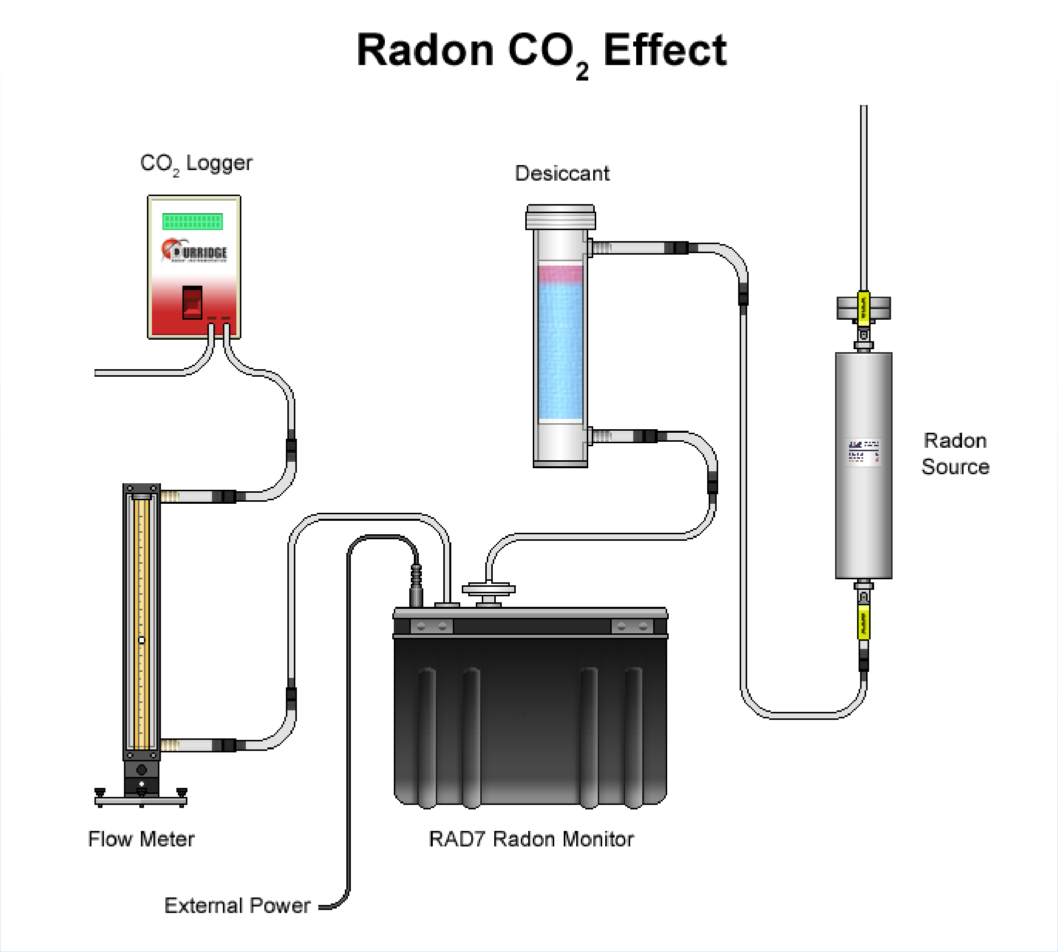
- Lab Work CO_2 vs. Rn
  - Hypothesis: Denser CO_2 slows down flight of particles to detector so more particles decay making a correction necessary.
  - The higher percentage of CO_2 the larger the correction.
  - Different corrections should be required for ^{220}Rn, ^{222}Rn and $^{220}Rn/^{222}Rn$.

Technical Status



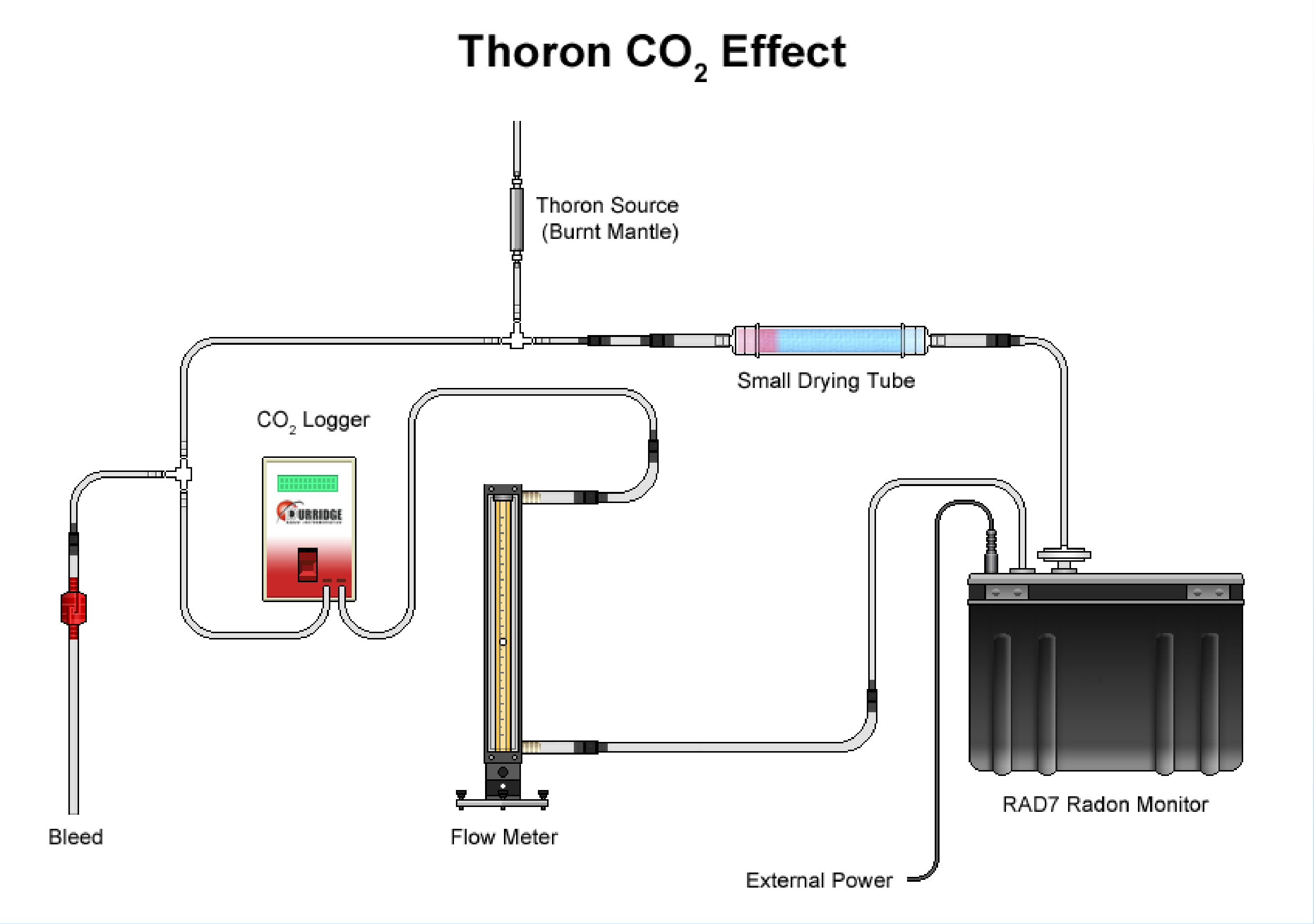
Lane-Smith and Sims, 2013

Technical Status



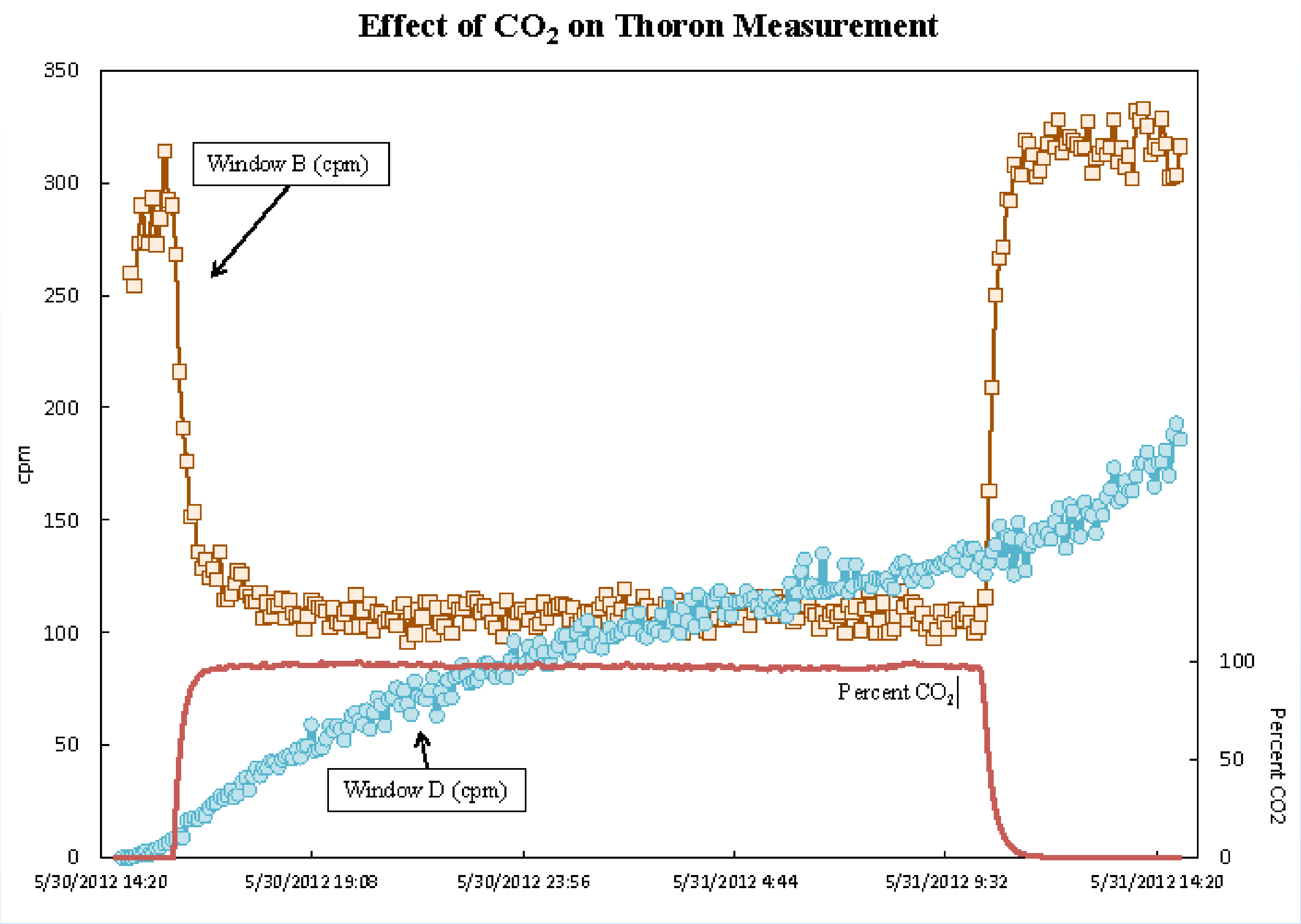
Lane-Smith and Sims, 2013

Technical Status



Lane-Smith and Sims, 2013

Lab Experiments



Lane-Smith and Sims, 2013

Experimental Summary

The ratio of $^{220}Rn/^{222}Rn$ is an important parameter.

In the presence of CO_2 the apparent ratio is decreased.

To correct for the presence of CO_2

the Radon reading should be multiplied by $1+(\%CO_2)\text{x } 0.003$

the Thoron reading should be multiplied by $1+(\%CO_2)\text{x } 0.019$

so

the $^{220}Rn/^{222}Rn$ ratio should be multiplied by $1+(\%CO_2)\text{x } 0.016$

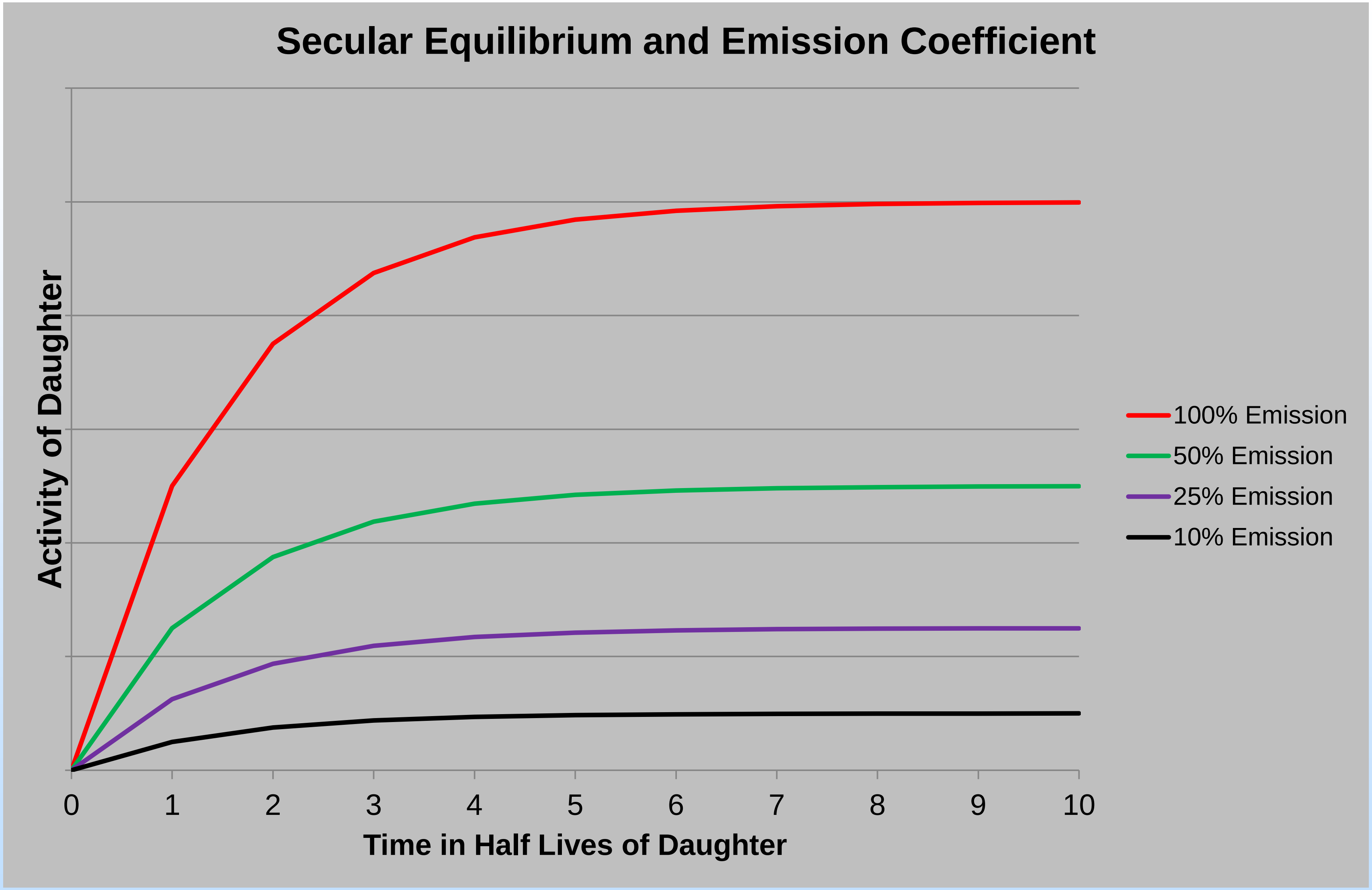
It appears that this correction will modify but not nullify the analysis and conclusions of Giammanco et al. 2007.

Technical Status

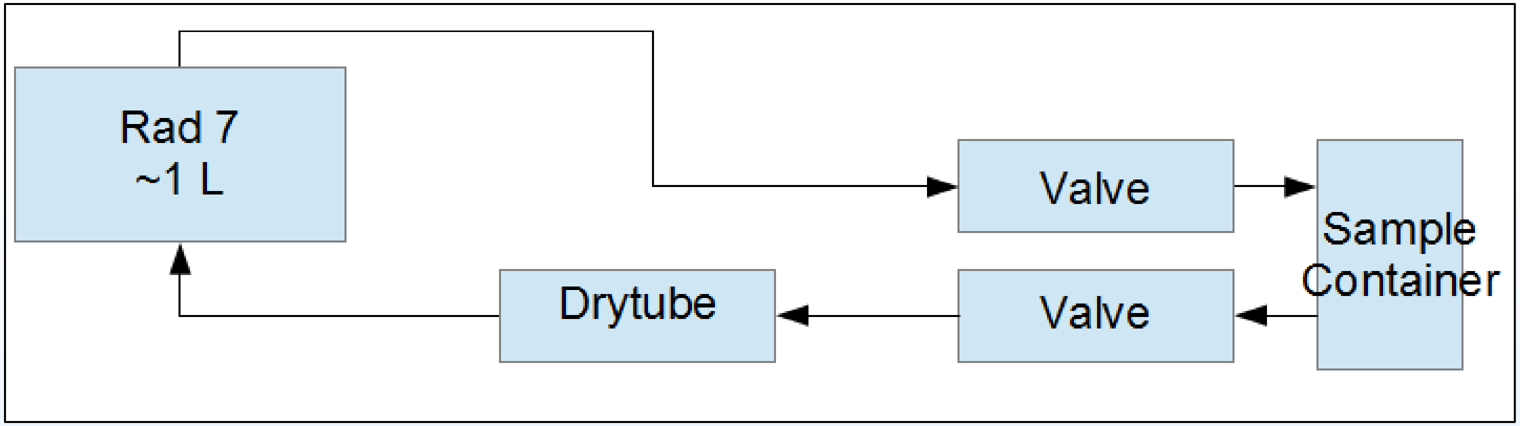
- Lab Work grain size (my project)
  - To understand shallow soil Rn degassing we must understand what controls the degassing.
  - Alpha recoil might produce enough kinetic energy for the Rn to escape the grain.
  - More surface area on a grain should increase amount of Rn that can escape through recoil. This makes Surface area to volume ratio an important .

Technical Status

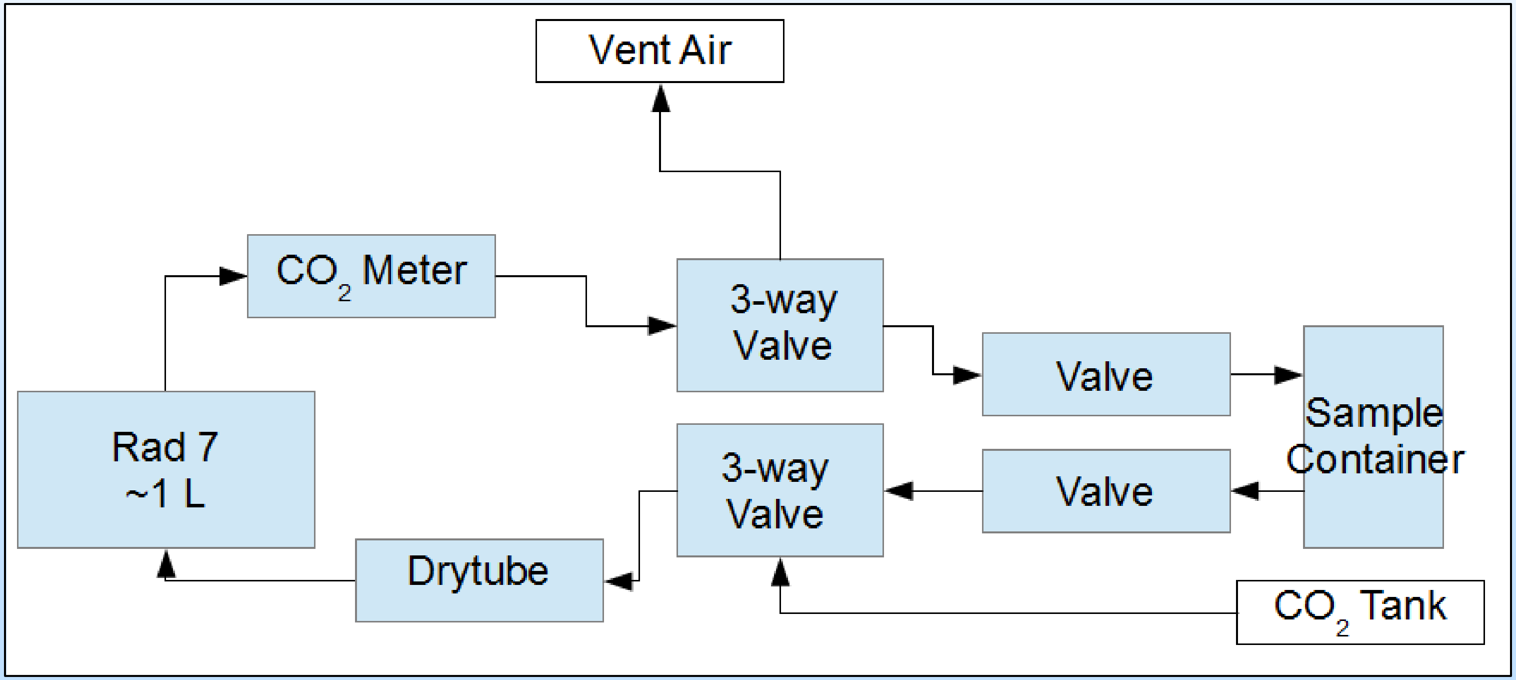
- Design
  - 3 Grains Sizes
    - 45-75 µm
    - 0.85-1 mm
    - 4-6 mm
  - 3 Lithologies
    - Banco Bonito Obsidian
      - U=5.7 ppm   Th=23 ppm
    - Cerros Del Rio Basalt
      - U=0.72 ppm Th=2 ppm
    - Guaje Pumice
      - U=11.8 ppm Th=40ppm

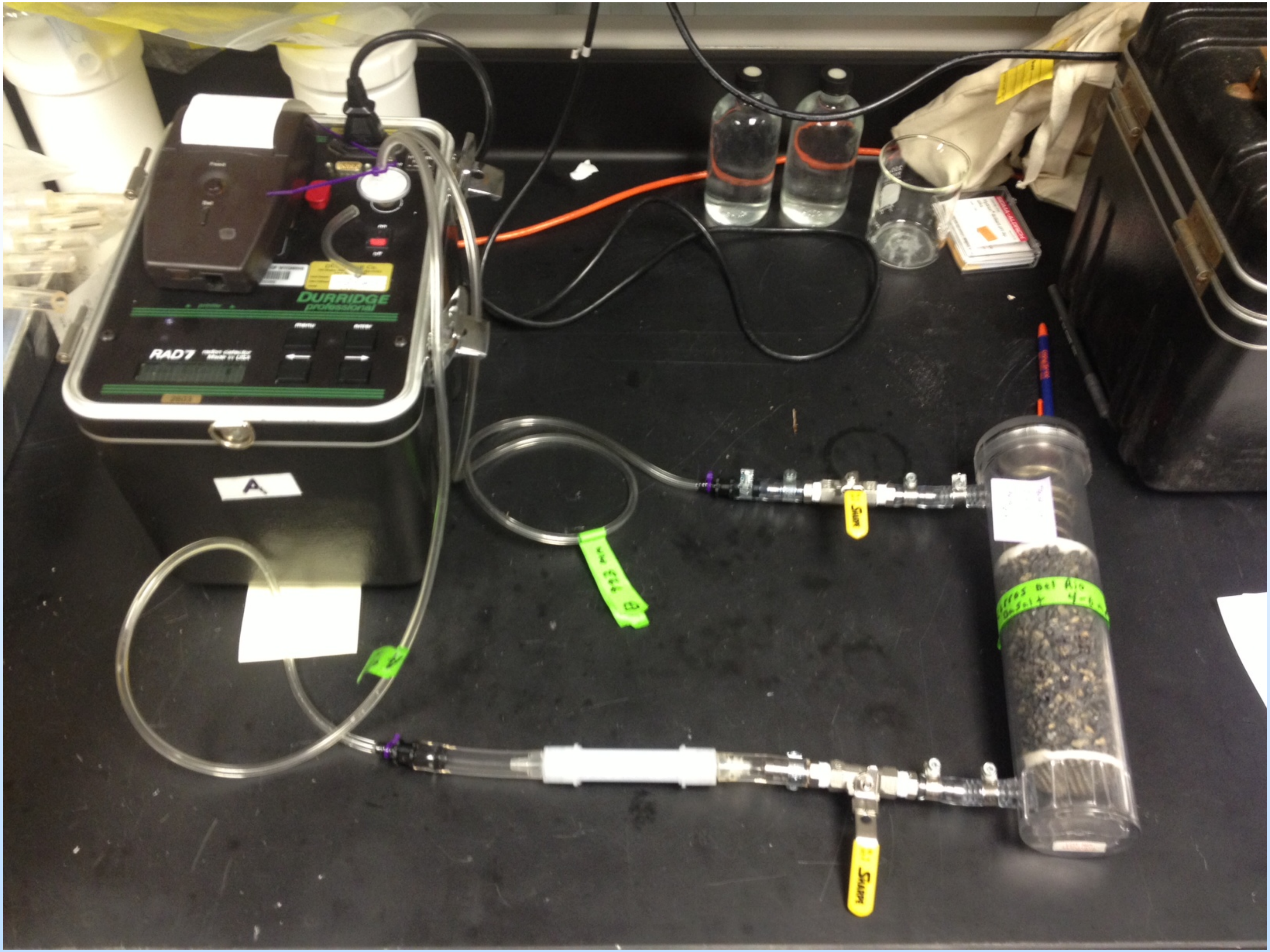

Secular Equilibrium and Emission Coefficient
Activity of Daughter
Time in Half Lives of Daughter
0
1
2
3
4
5
6
7
8
9
10
100% Emission
50% Emission
25% Emission
10% Emission

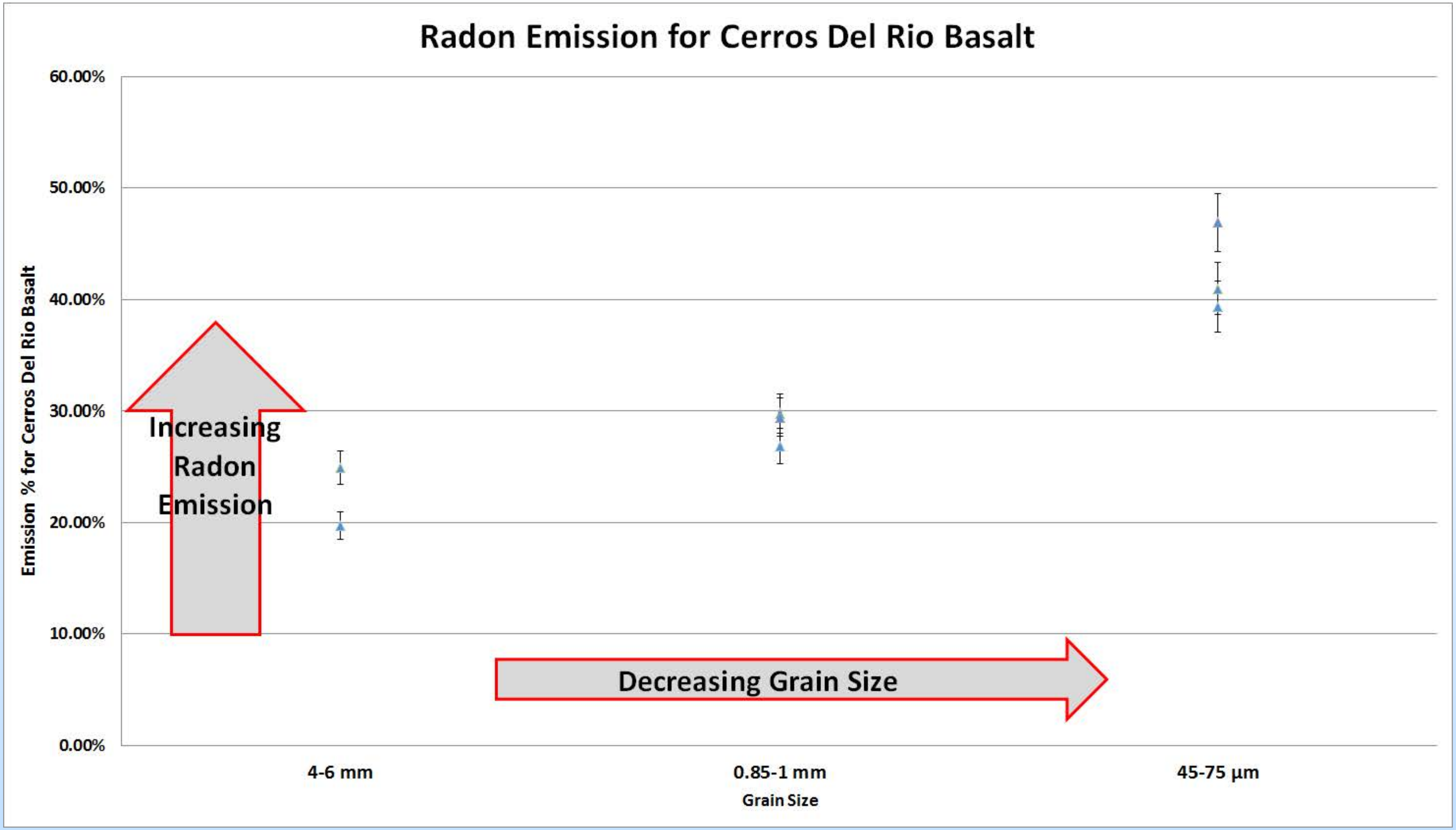
Ambient Air Schematic



CO_2 Schematic




DURRIDGE
professional
RAD7
A



Radon Emission for Cerros Del Rio Basalt
60.00%
50.00%
40.00%
30.00%
20.00%
10.00%
0.00%
Emission % for Cerros Del Rio Basalt
Increasing Radon Emission
Decreasing Grain Size
4-6 mm
0.85-1 mm
45-75 µm
Grain Size

Accomplishments to Date

- Two MS students performed research and successfully defended thesis
- Four BS students have performed undergraduate research
- Site characterization of natural analog (Moxa Arch, SW Wyoming; Thermopolis thermal area; Yellowstone) is complete
- Determined that standard method for measuring Rn needs correction in high CO_2 environments
- Rn versus grain size alpha recoil experiments are planned out and are close to 50% complete

Summary

- Key Findings
  - A measureable relationship between grain size and Radon exists but needs more information to sufficiently quantify
- Lessons Learned
  - In the presence of CO_2, Thoron and Radon must be corrected for the effect CO_2 has on the instrument
- Future Plans:
  - Continue alpha recoil experiments
  - Understanding the effect of water rock interaction on Rn and Ra in Yellowstone and Thermopolis, WY.
  - Start Laboratory Tests of Effects of CO_2-H_2O-Rock Reaction

Organization Chart

Dr. John Kaszuba
(co PI)

Dr. Ken Sims
(co PI)

BS Students
Virginia Marcon (graduated)
Matt Carberrry (graduated)
Evan Soderberg (graduated)
Mike Schedel

MS Students
Tim Moloney (defended)
Allison Pluda (defended)

External Collaborators
Dr. Salvatore Giammanco (Istituto Nazionale di Geofisica e Vulcanologia)
Dr. Matt Charette (WHOI)
Dr. Derek Lane-Smith (Durridge Company, Inc.)

Gantt Chart

Subtask	Milestone					
1	HQ Milestone: Project Kick-off Meeting					
1	HQ Milestone: Educational Program Instituted					
1	HQ Milestone: Semi-Annual Progress Report					
1	HQ Milestone: Yearly Review Meeting					
1	HQ Milestone: Yearly Review Meeting					
1.1	Update Project Management Plan					
1.3	Annual progress report submitted					
1.3	Annual progress report submitted					
2.1 & 2.2	Obtain & calibrate radon and CO2 analyzers					
2.3	Begin 1st set of Field Measurements					
2.4	Complete 75% of Field Measurements					
3.1	Begin 1st set of Laboratory Tests of Rn and Grain Size					
3.2	Submit report for Laboratory Tests of Rn and Grain Size					
3.3	Begin 1st set of Laboratory Tests of Effects of CO2-H2O-Rock Reaction					
3.4	Submit report for Laboratory Tests of Effects of CO2-H2O-Rock Reaction					
4.1	Develop initial model					
4.2	Final report submitted					
		2010	2011	2012	2013	2014

Bibliography

- Lane-Smith, D., & Sims, K. W. W., 2013. The effect of CO_2 on the measurement of ^{220}Rn and ^{222}Rn, with instruments utilizing electrostatic precipitation. *Acta Geophysica*. V. 61, p. 822-830, available at http://agp.igf.edu.pl/
- Giammanco, S., K. W. W. Sims, and M. Neri, 2007, Measurements of ^{220}Rn and ^{222}Rn and CO_2 emissions in soil and fumarole gases on Mt. Etna volcano (Italy): Implications for gas transport and shallow ground fracture,. Geochemistry, Geophysics, Geosystems, v. 8, Q10001, doi:10.1029/2007GC001644, available at: http://onlinelibrary.wiley.com/journal/10.1002/(ISSN)1525-2027

Notes

- 1 Becquerel = 1 decay/second = 27.027027 picocuries
- Otowi Member Guaje Member = 1.4 Ma (Guaje Pumice)
- Cerros Del Rio Basalt Fields = 1.96 to 4.62 Ma
- Banco Bonito = Still debated most likely <140 ka

Notes cont.

- The DURRIDGE RAD7 uses a solid state alpha detector. A solid state detector is a semiconductor material (usually silicon) that converts alpha radiation directly to an electrical signal. One important advantage of solid state devices is ruggedness. Another advantage is the ability to electronically determine the energy of each alpha particle. This makes it possible to tell exactly which isotope (polonium-218, polonium-214, etc.) produced the radiation, so that you can immediately distinguish old radon from new radon, radon from thoron, and signal from noise. This technique, known as alpha spectrometry
- When the radon and thoron daughters, deposited on the surface of the detector, decay, they emit alpha particles of characteristic energy directly into the solid state detector. The detector produces an electrical signal. Electronic circuits amplify and condition the signal, then convert it to digital form. The RAD7 's microprocessor picks up the signal and stores it in a special place in its memory according to the energy of the particle. The accumulations of many signals results in a spectrum.