Issue 24, 2021 | Previous Article | Next Article

From the journal:
Physical Chemistry Chemical Physics

https://arxiv.org/pdf/1807.09253.pdf

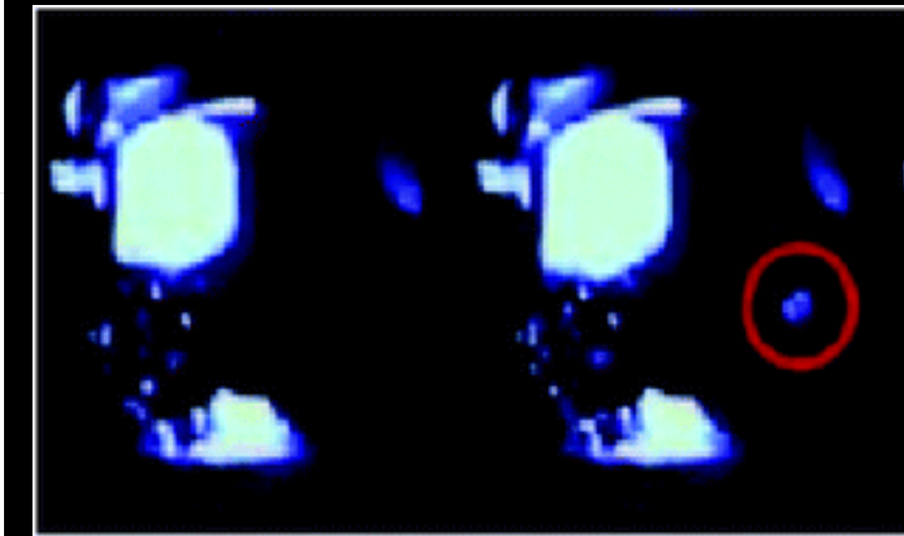
Demonstration of neutron radiation-induced nucleation of supercooled water†

Check for updates

Matthew Szydagis, *[a] Cecilia Levy, *[a] Yujia Huang,[a] Alvine C. Kamaha,[a] Corwin C. Knight,[a] Gregory R. C. Rischbieter [a] and Peter W. Wilson *[bc]

Author affiliations

https://pubs.rsc.org/en/content/articlelanding/2021/cp/d1cp01083b



The Snowball Chamber: Supercooled Water for Dark Matter, Neutrinos, and General Particle Detection

University at Albany, State University of New York

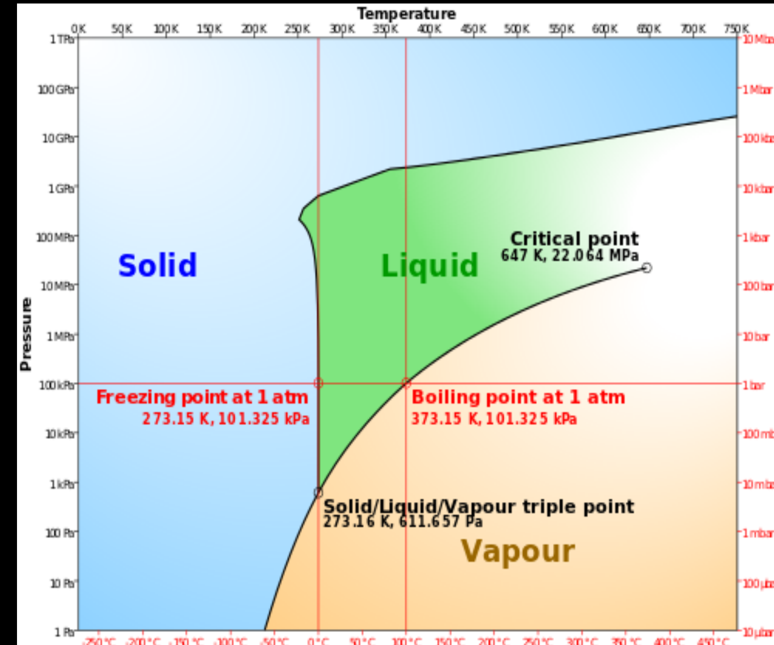
Matthew Szydagis

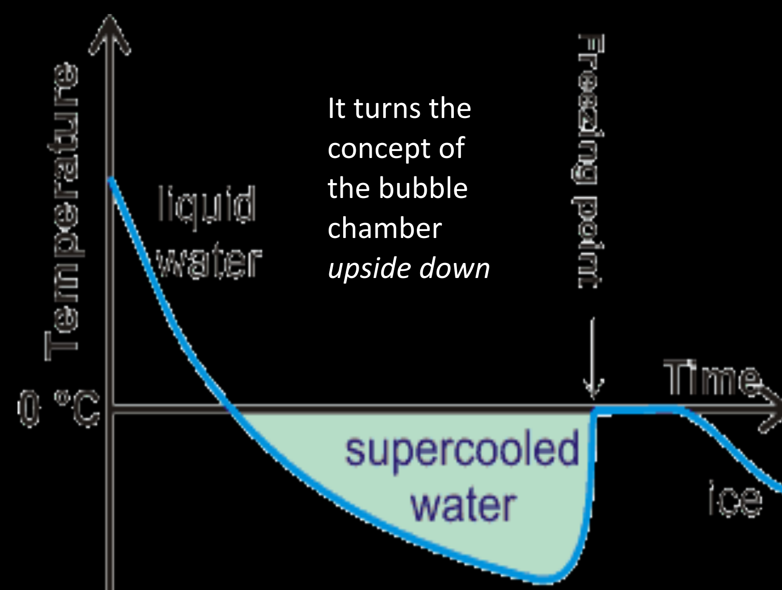
SUNY RF The Research Foundation for The State University of New York

UCLA DM Conference April 1st, 2023

What is Supercooling?

- A liquid is cooled below its normal freezing point, not using freezing point depression (e.g., salting the sidewalk)
  - Metastable
  - Requires high purity and a clean, smooth container, just like with superheating liquid (heating above the boiling point)
- Freezing occurs when the liquid finds a nucleation site, or it has otherwise been "disturbed" (sound, electric fields)
  - One cannot stop nucleation: it **snowballs**
  - The process is highly exothermic: see the cartoon at the right
- Smaller samples are easier to cool
  - Min temperature depends on radius of sample (Bigg 1953, Mossop 1955)
- Unexplored phase transition in physics!
  - Cloud & bubble chambers both done





It turns the concept of the bubble chamber *upside down*

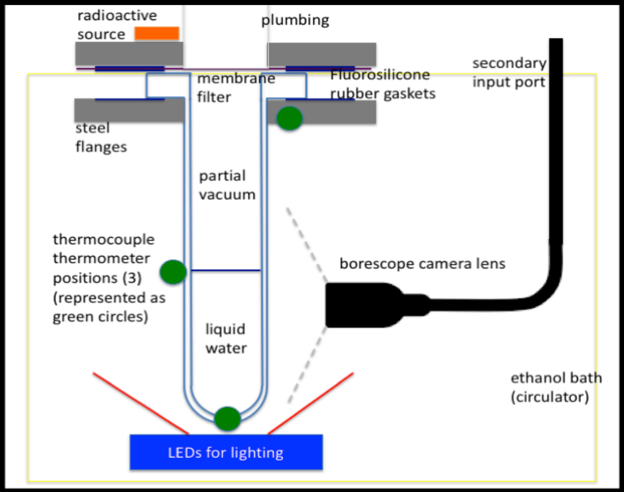
Challenges Using Supercooled Water

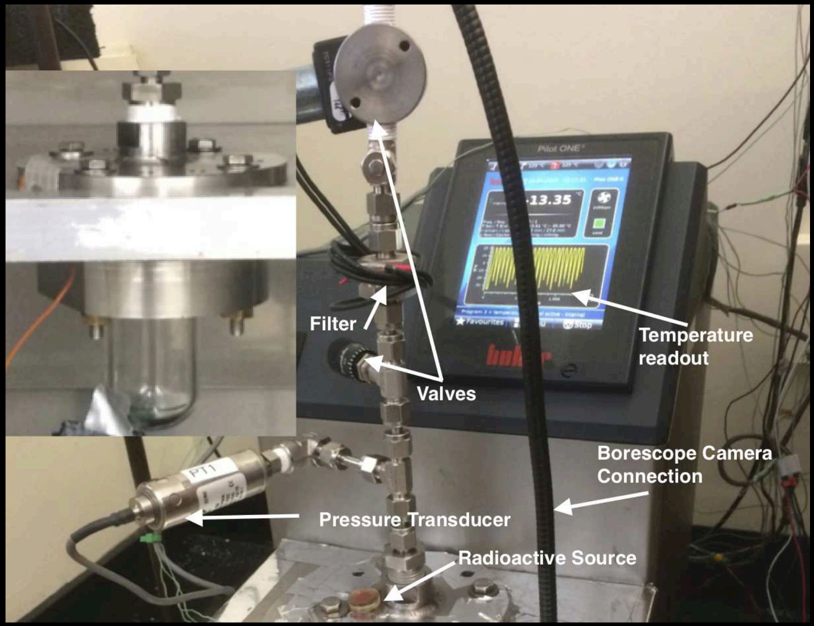
done before, but only with beta and gammas, most recently by Varshneya (Nature, 1971)

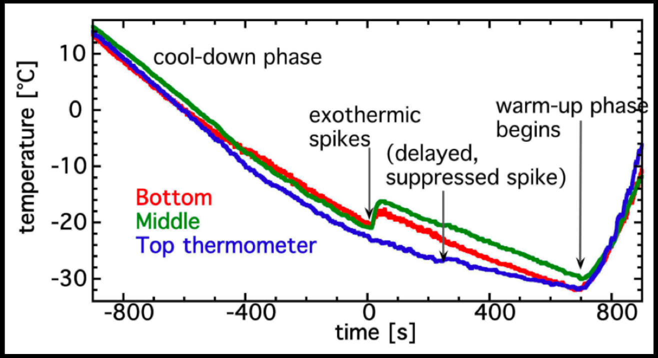
Physics Department, University of Roorkee, India

- Getting as cold as feasible, sans unwanted nucleation as a background
  - If like a bubble chamber except in reverse, colder should be **better**, because it should mean lower energy threshold
  - Must not just avoid particulates (heterogeneous nucleation) but the homogenous nucleation limit too (this may imply the existence of a low-threshold asymptote)
- Finding the ideal rate of cooling
  - Too slow means low live-time and/or more opportunity for an unwanted nucleation (from vibration, background radiation, etc.)
  - But too fast means thermal lag/gradient, which encourages nucleation
- The scientific method in its purest form: “let’s try it and see” approach
  - Hypothesis: radiation, specifically neutrons, is/are able to freeze supercooled water

Prototype Detector Setup

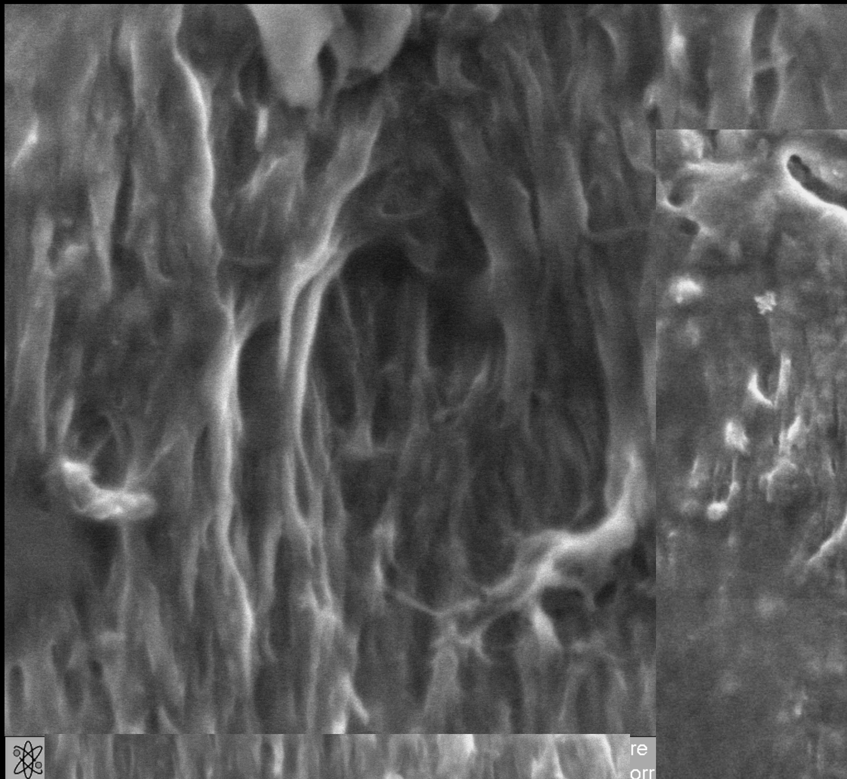


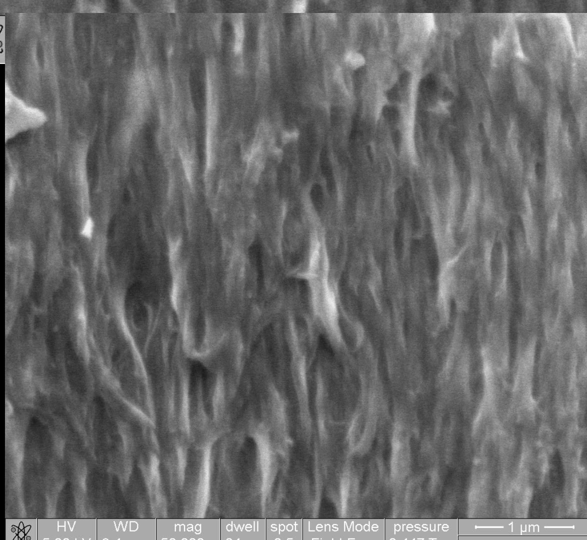


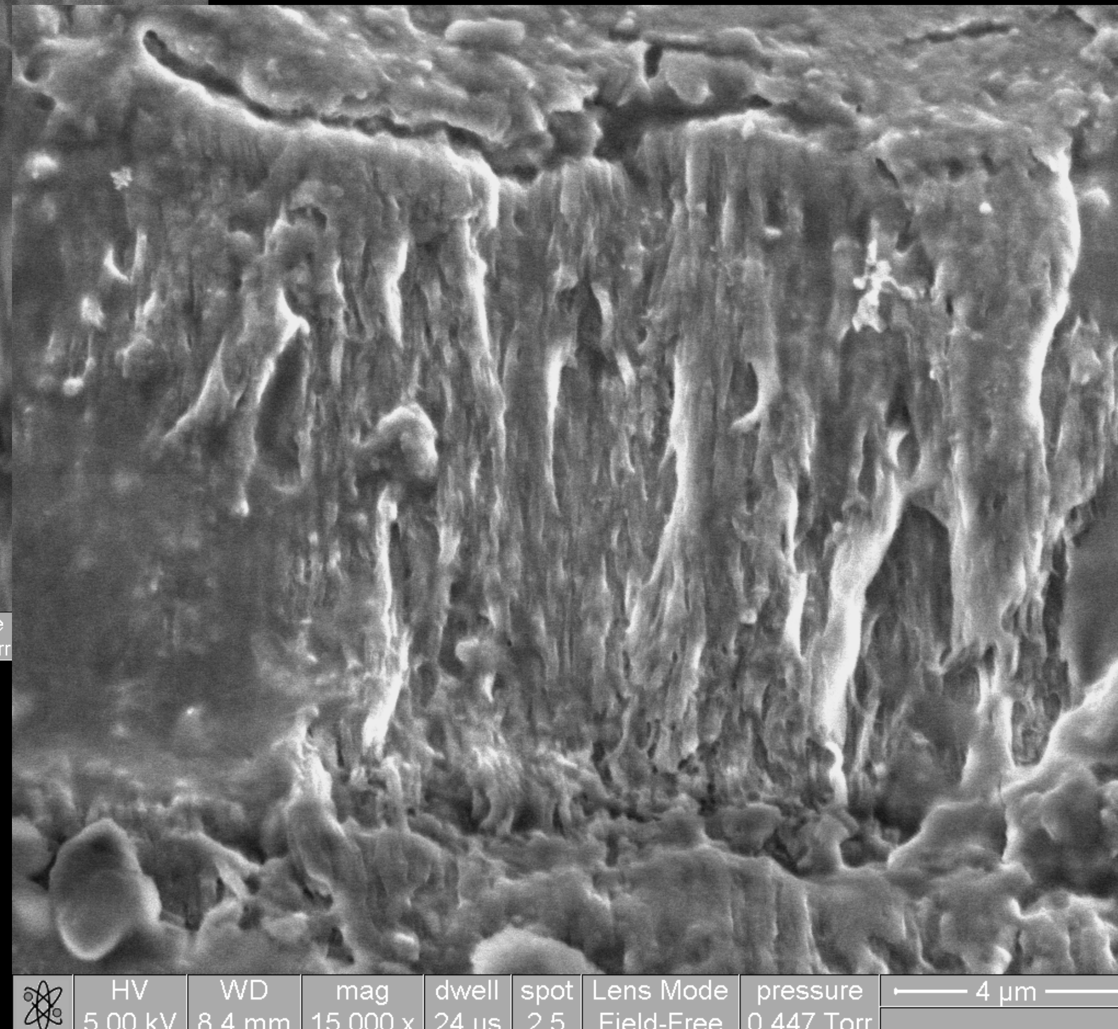


- 20 g (20 mL) of purified water contained in a smooth, cleaned fused quartz vessel
  - The water is processed through multiple filters, deionized, and ultimately distilled through a 20-nm flat-sheet non-linear membrane (only gas can pass through)
- Thermocouple thermometers (all used)
  - 3: top, middle, and bottom -- to see that "exothermic spike" <=
- Borescope camera for image acquisition
  - Only 1, so no 3-D info, but counted # of scatters
- Coincident counter under vessel, aligned
  - Plastic scintillator with attached SiPM

Electron Microscope Images of a Membrane Filter (Novamem)



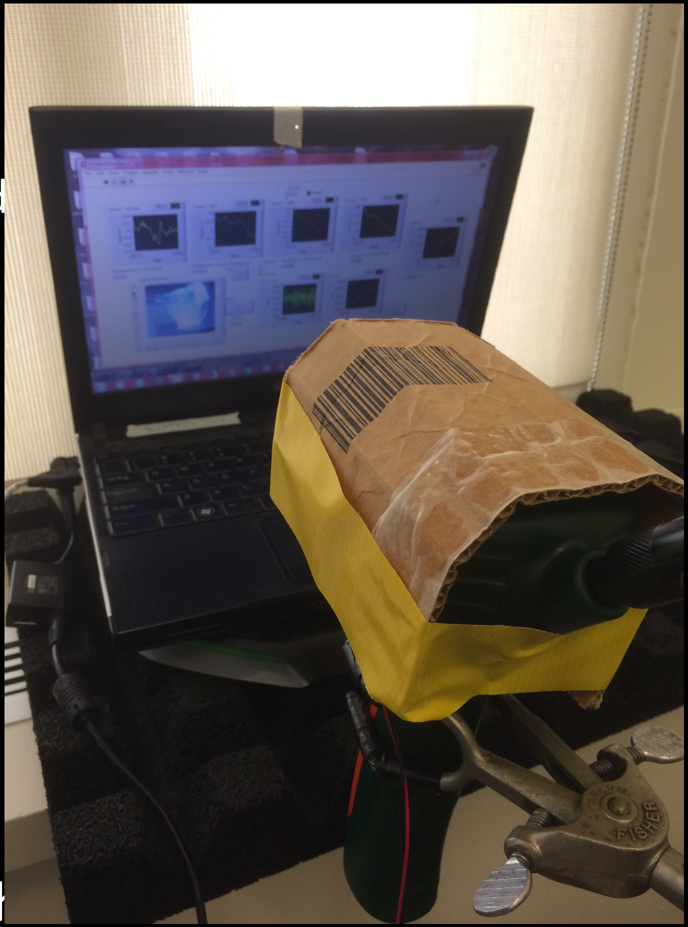




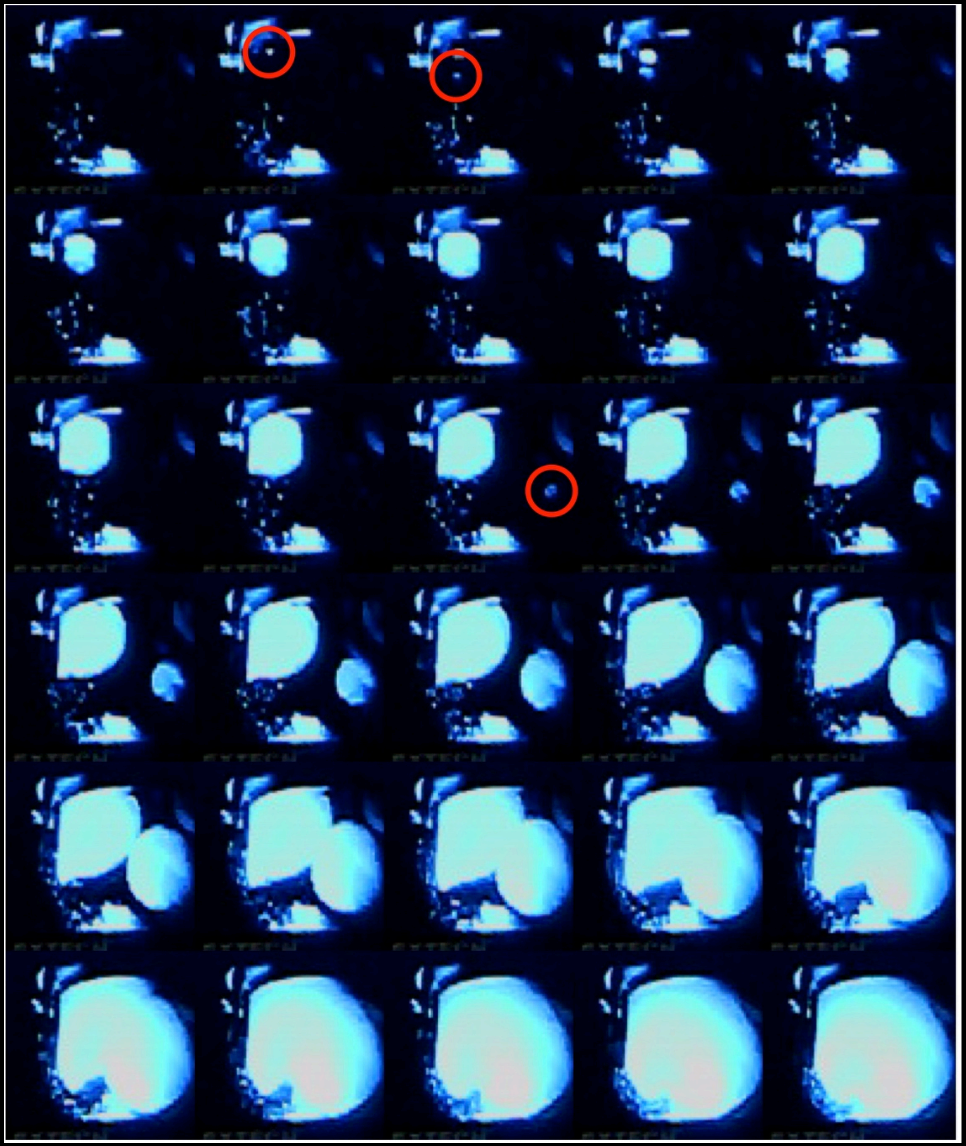
photos courtesy of Prof. Kathy Dunn, SUNY Poly CNSE

Detector Operation

- About -20 °C and lower achieved, at a maximum cooling rate of -2°C per minute
  - Water may be able to go as cold as -40°C (world record: Goy, 2011)
- Partial vacuum of ~8-9 psia (water vapor, after earlier evacuation of the air)
- 1-hour cooling and heating (melting) full cycle, with ~50% time spent < 0°C ("live")
- Multiple run conditions / calibrations
  - Control (no radioactive source)
  - 200 n/s AmBe (with, w/o lead shielding)
  - 10 μCi ^{137}Cs gamma-ray source
  - 3,000 n/s ^{252}Cf (with Pb shielding)
- Shielding stops gammas from interfering with the thermocouples' operation
  - Also makes more n's, alters their E-spec

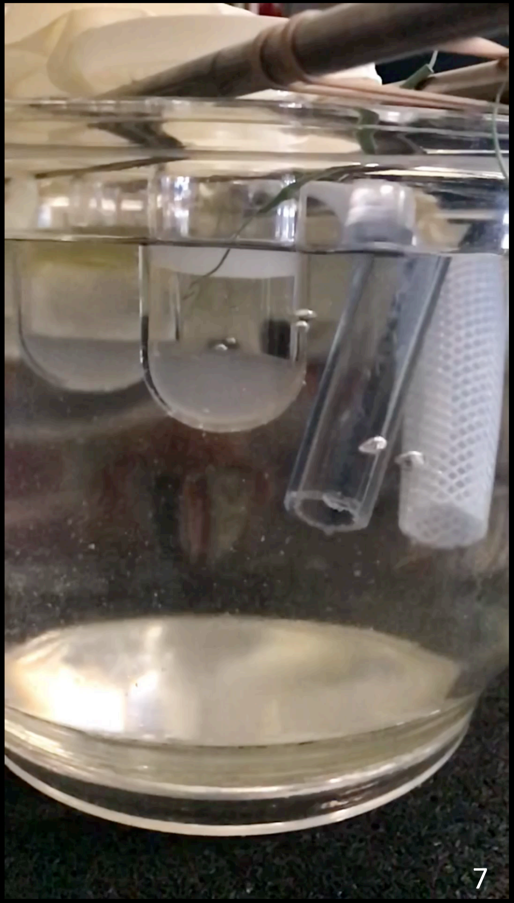


Some Example Events

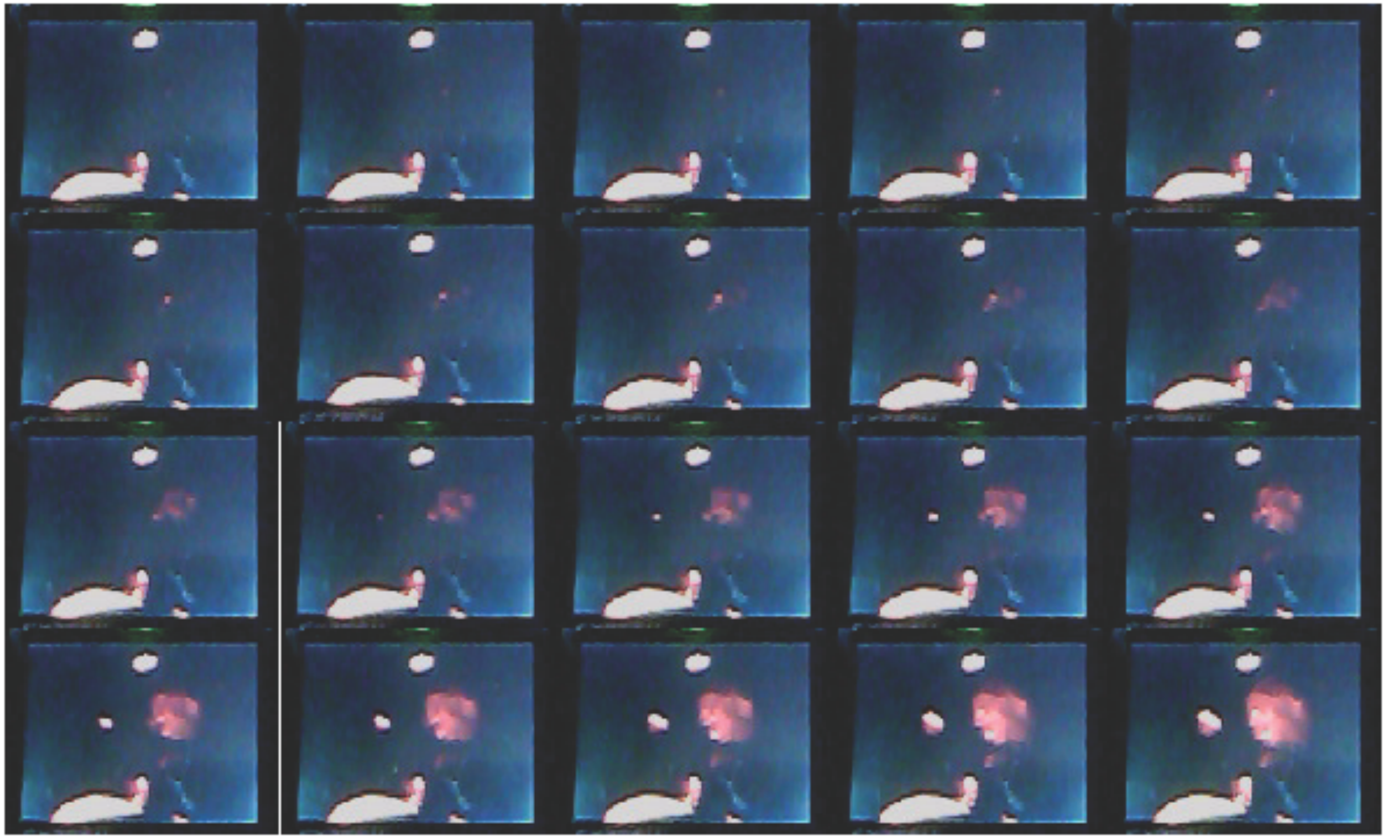


Pause for: superCool.mp4

<= triple (2+1) nucleation



Double Crystal Slide Show



Snowmass LOI Submitted

Metastable Water: Breakthrough Technology for Dark Matter & Neutrinos

M. Szydagis[1], C. Levy[1], P. S. Barbeau[2], A. E. Bolotnikov[3], E. Brown[4], M. C. Carmona-Benitez[5], G. Cox[5], L. de Viveiros[5], M. Diwan[3], T. Guile[1], G. Homenides[1], Y. Huang[1], A. Kamaha[1], D. Kodroff[5], I. Magliocca[1], G.R.C. Rischbieter[1], D. Woodward[5], and M. Yeh[3]

[1] *The University at Albany, State University of New York (UAlbany)*
[2] *Duke University / Triangle Universities Nuclear Laboratory (TUNL)*
[3] *Brookhaven National Laboratory (BNL)*
[4] *Rensselaer Polytechnic Institute (RPI)*
[5] *Pennsylvania State University, University Park, PA*

August 2020

Cosmic Frontier Topical Groups:

- (CF1) Cosmic Frontier: Dark Matter: Particle-like

Neutrino Frontier Topical Groups:

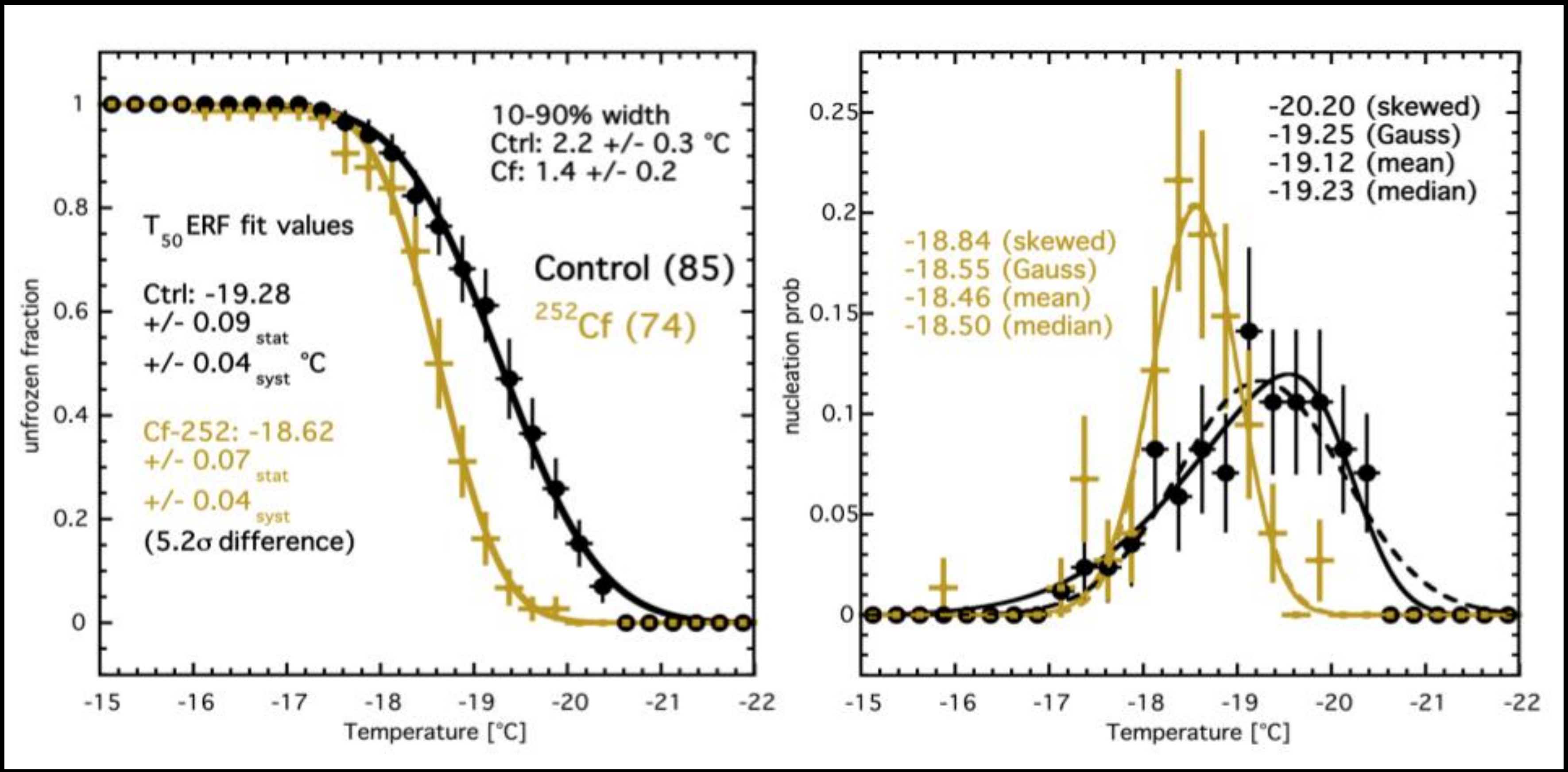
- (NF04) Neutrino Physics Frontier: Neutrinos from Natural Sources
- (NF05) Neutrino Physics Frontier: Neutrino Properties
- (NF06) Neutrino Physics Frontier: Neutrino Interaction Cross-Sections
- (NF07) Neutrino Physics Frontier: Applications
- (NF10) Neutrino Physics Frontier: Neutrino Detectors

Intensity Frontier Topical Groups:

- (IF6) Instrumentation: Calorimetry
- (IF8) Instrumentation: Noble Elements

also gave “Community Voices” talk

Our Most Important First Results



KS test p-values: 6.64 x 10^-5 comparing times
Conservatively using only "local" control

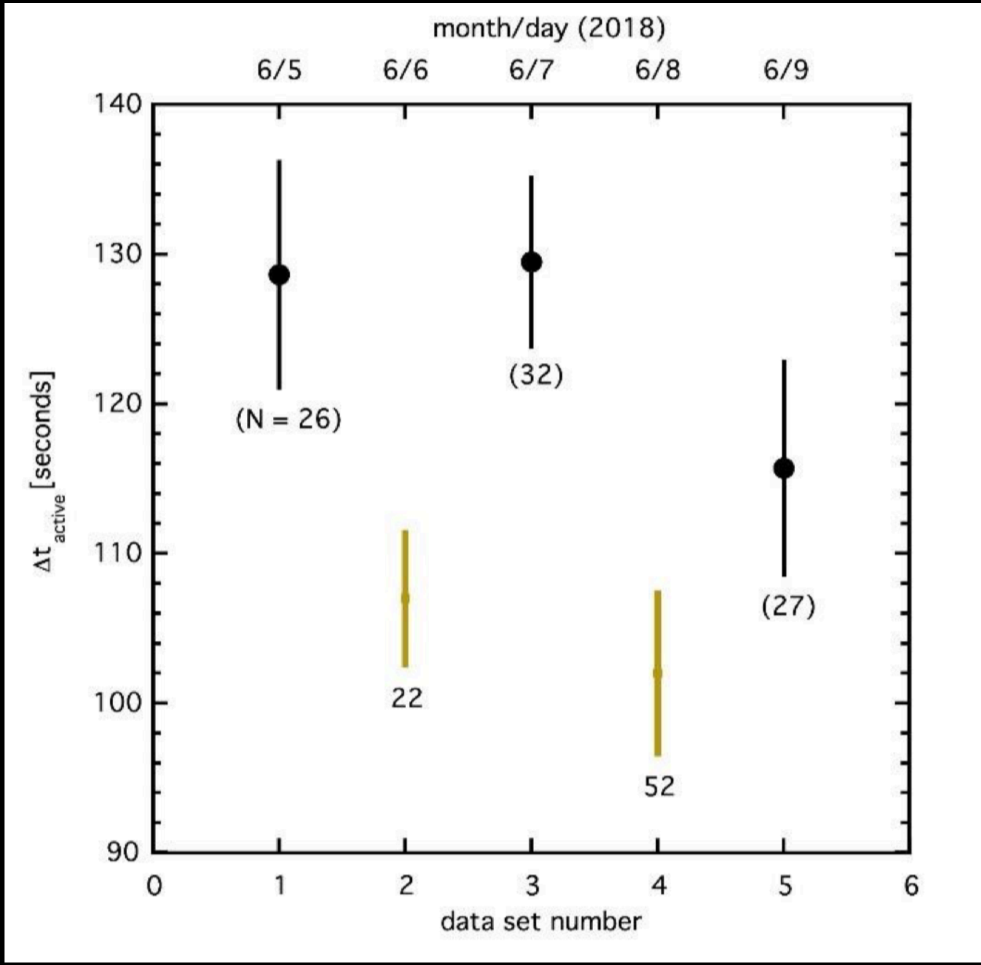
p = 3.09 x 10^-8
for temperatures!

Further Analyses (Cf)

- Reduction in supercooled time in presence of neutron sources
  - Effect enhanced with lead shielding
  - Bigger effect with stronger source

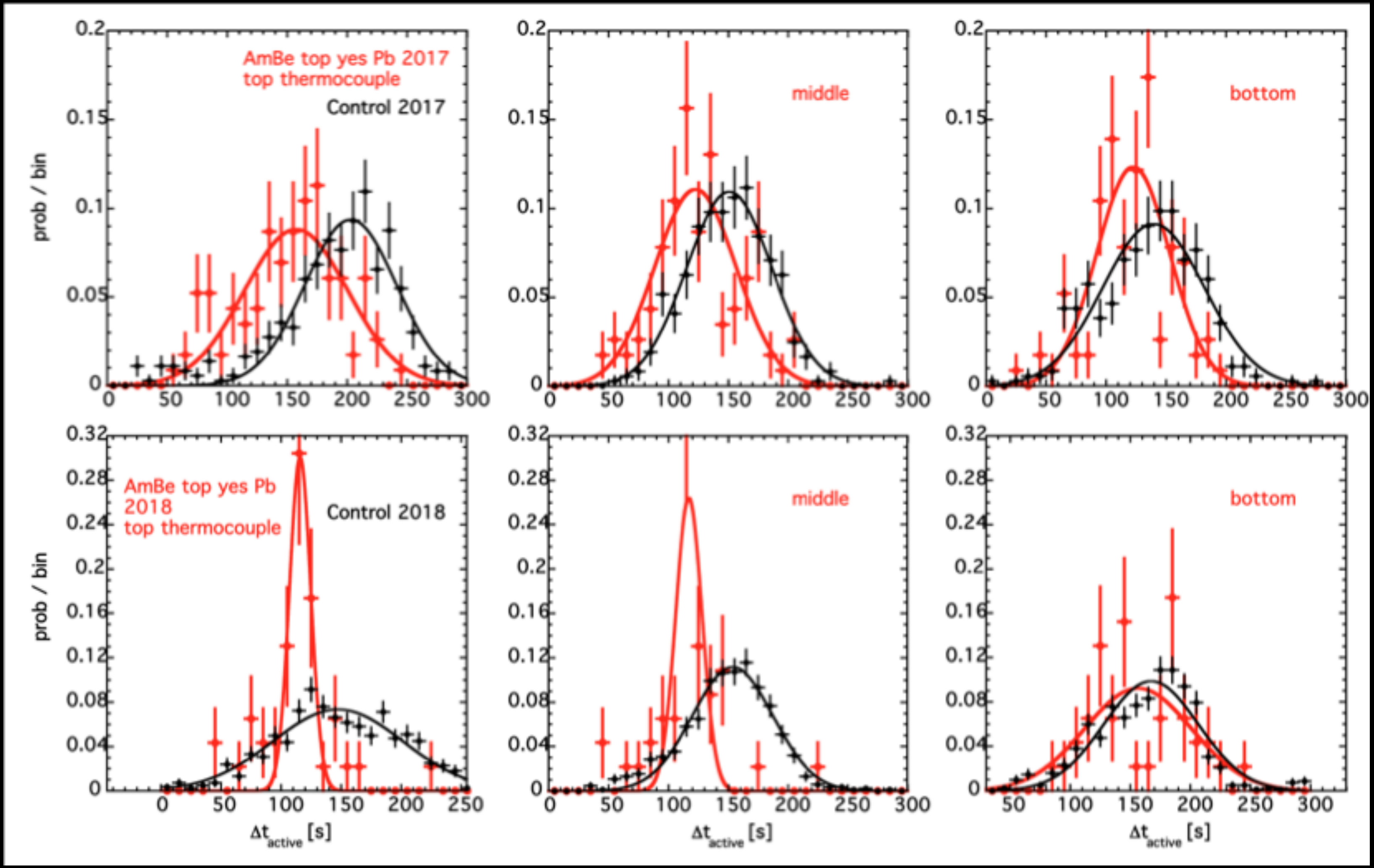
* We conclude that neutrons can *freeze* H_2O (1^{st} observation)

  - Alternated the source and BG runs
  - Checked room temp as a systematic

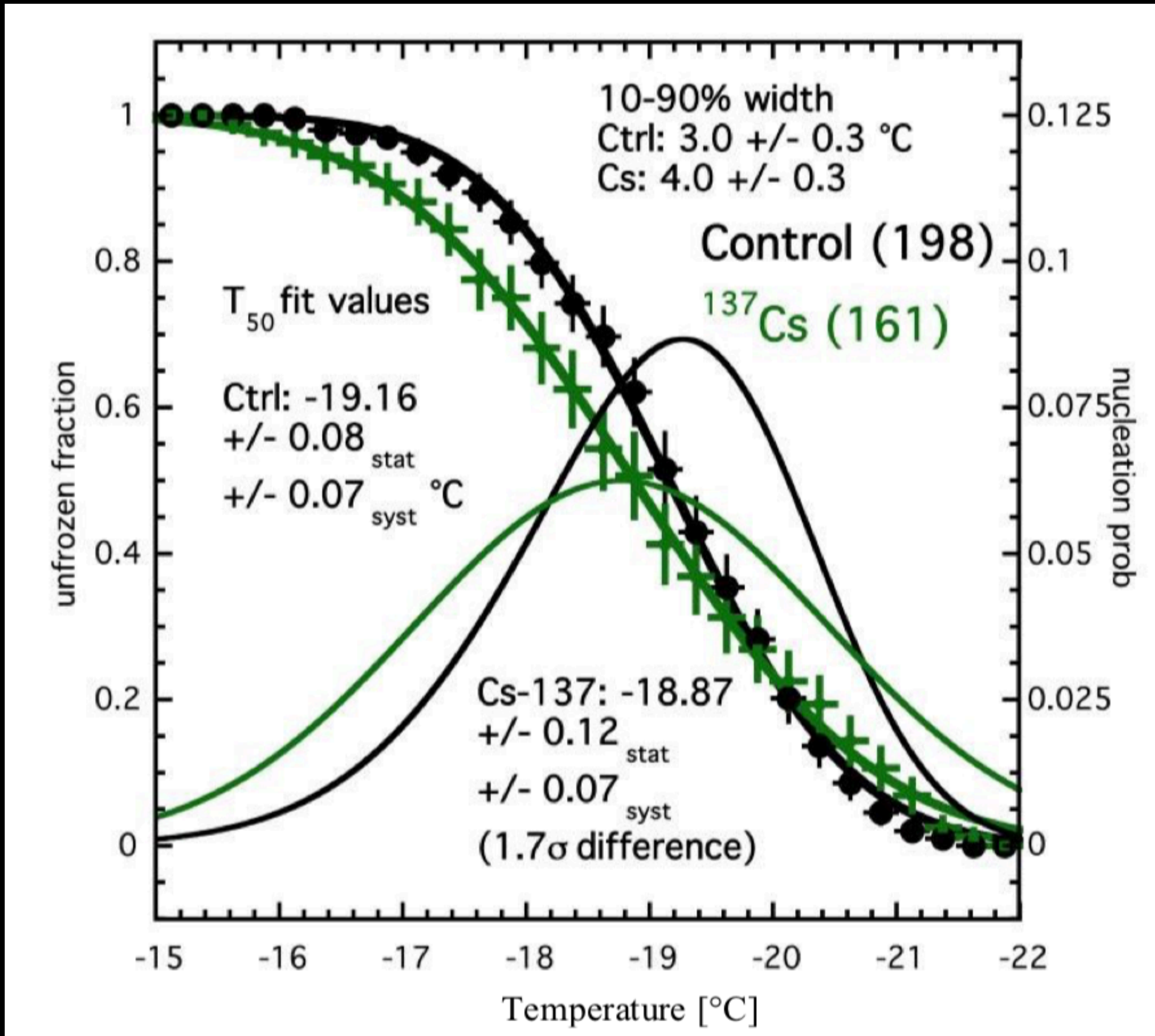


Systematics: AmBe in 2017 and 2018

Across three different thermometers (Why not Cf? Similar study not possible for it)

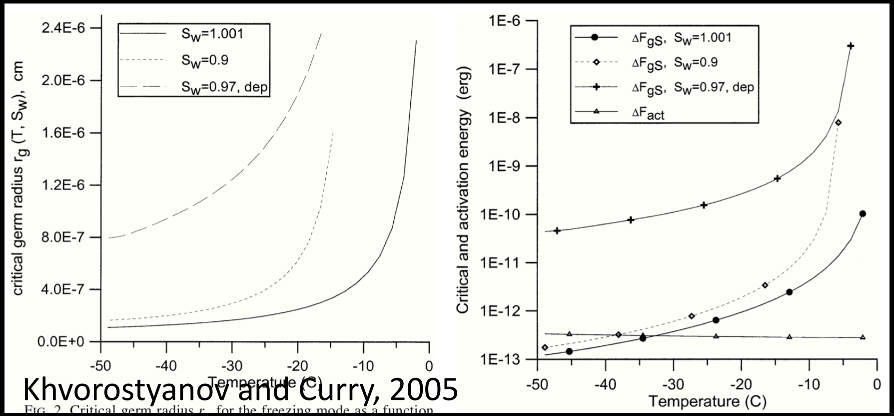


Gamma-Ray Calibration (^{137}Cs 662 keV)



- No statistically significant effect so far from gammas (0.662 MeV energy)
  - May be a sign of SOME e- recoil rejection?

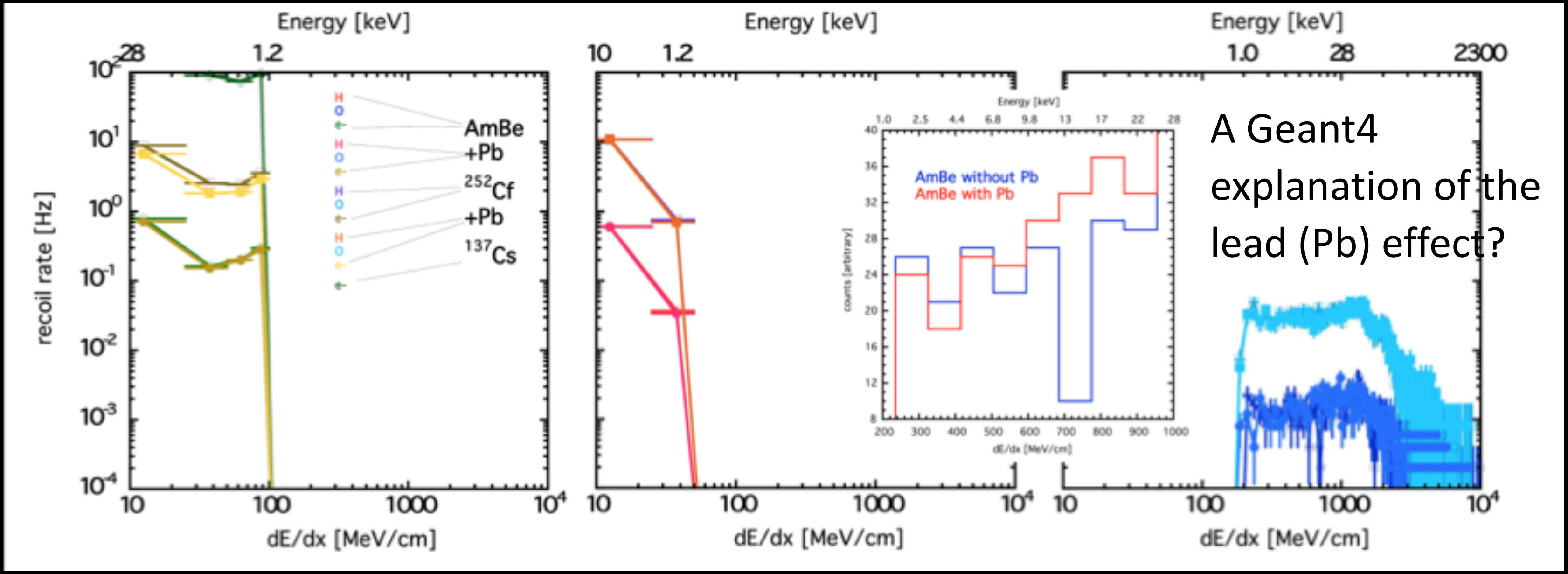
Geant4 Sims of these Data



Khvorostyanov and Curry, 2005

- Our initial data APPEAR to be following "worst-case scenario" for threshold, but even then extrapolates to O(10 eV) at ~ -30 °C. O(1 eV) across most of lit
- Our **snowball chamber** appears to have pair of tunable thresholds, just like bubble chamber: one for *E* and one for stopping power or *dE/dx* (or the LET)

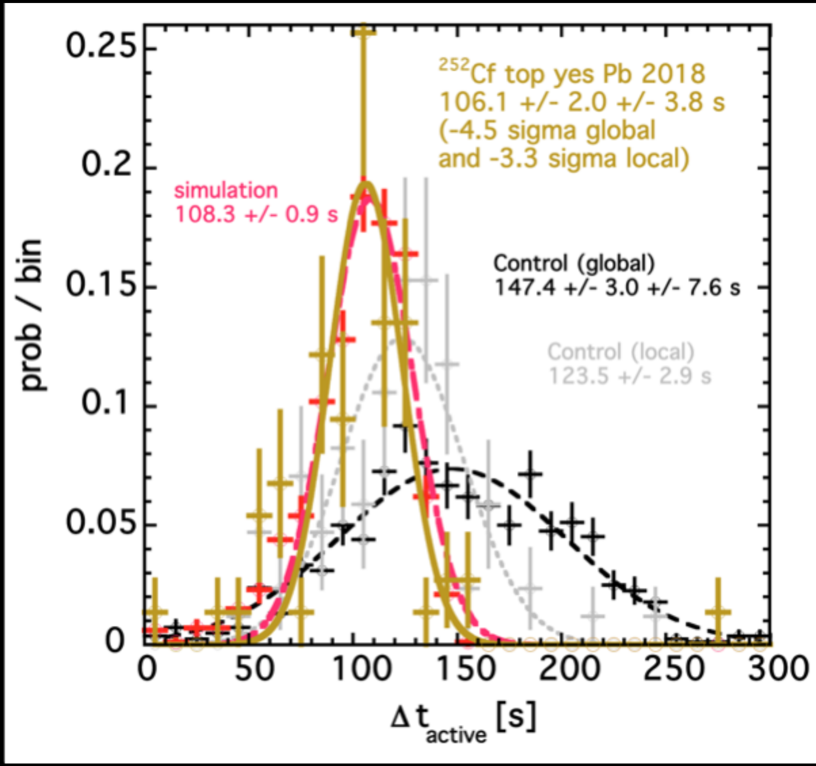
Electrons *Protons (Hydrogen)* *Oxygen*

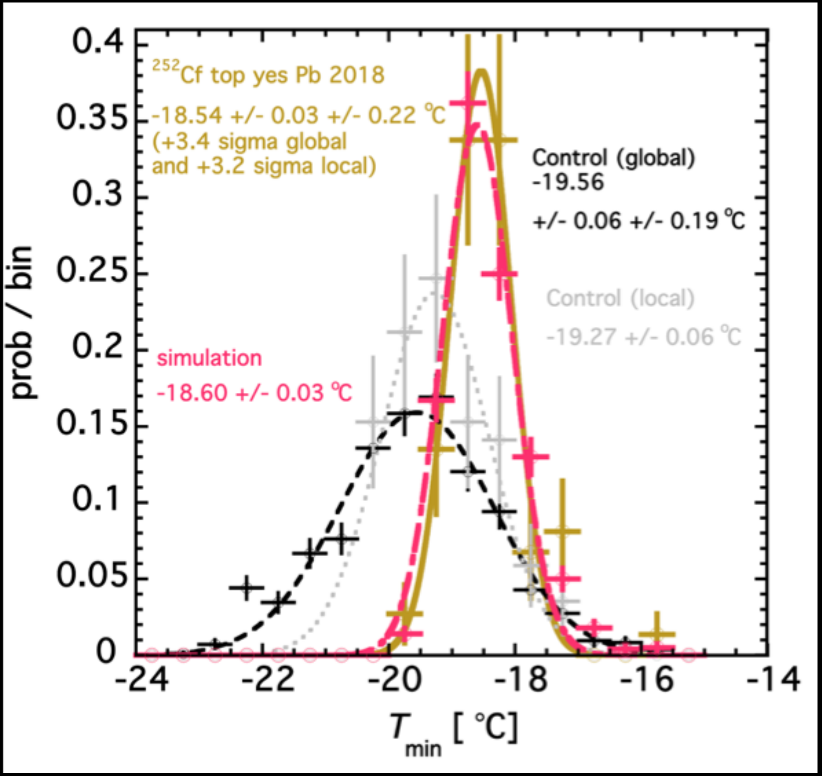


A Geant4 explanation of the lead (Pb) effect?

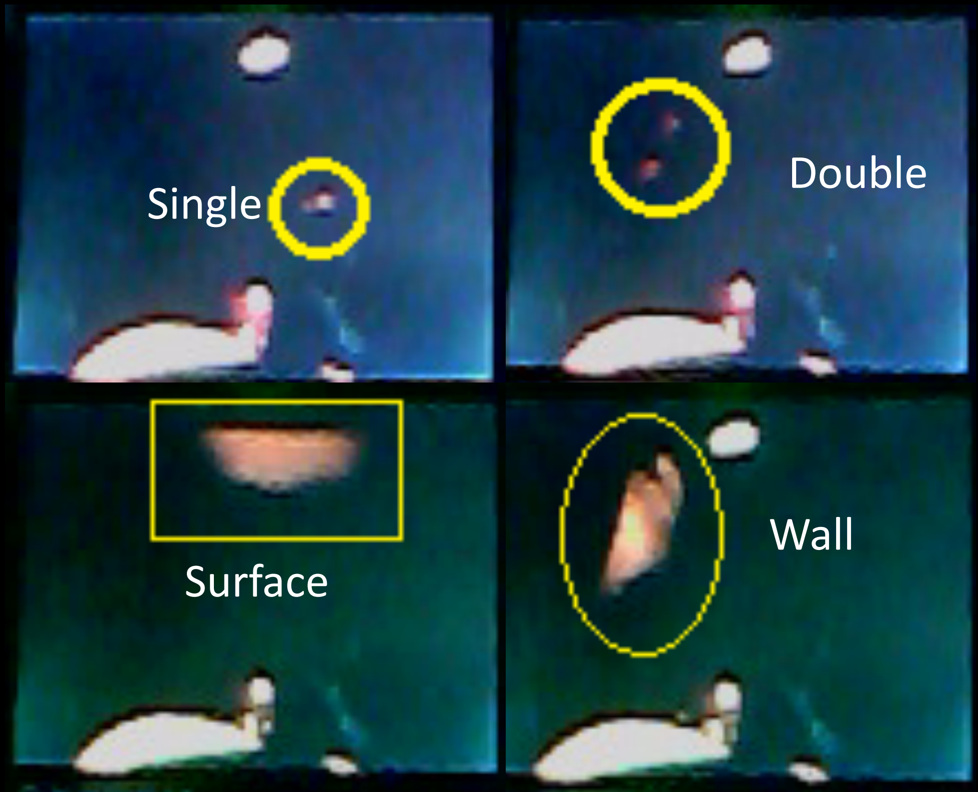
Additional MC post-G4

Open question: does neutron MFP match data?



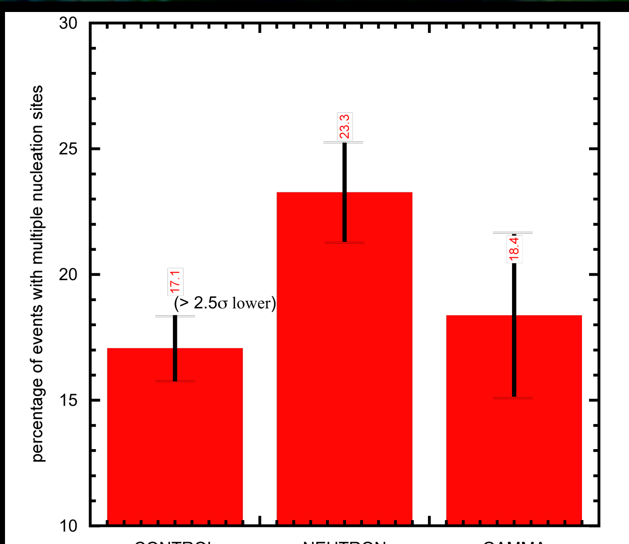


types of events



A Preliminary Image Analysis

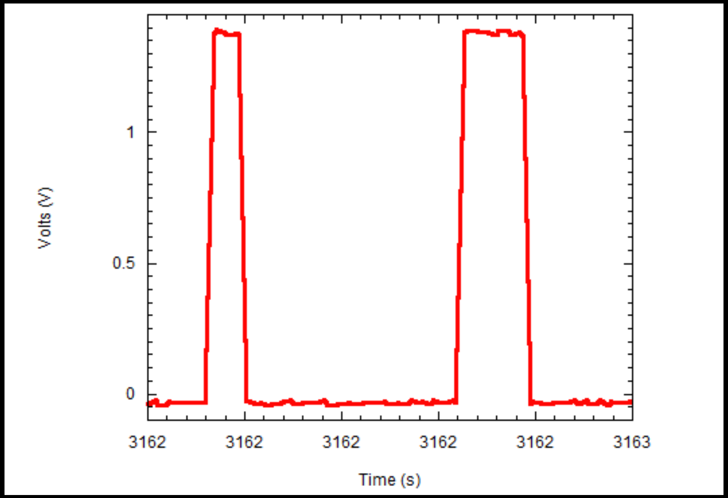
- Even without a second camera or mirror, can kind of tell wall/surface events
  - Most common, especially in control results
- Still far from perfect by eye
  - So, focus only on counting
- More multiple scatters by a lot in neutron data
  - Confirmation neutrons can cause crystallization
  - Triples, quad seen even



blind analysis performed, employing large team of undergraduate students scanning photographs

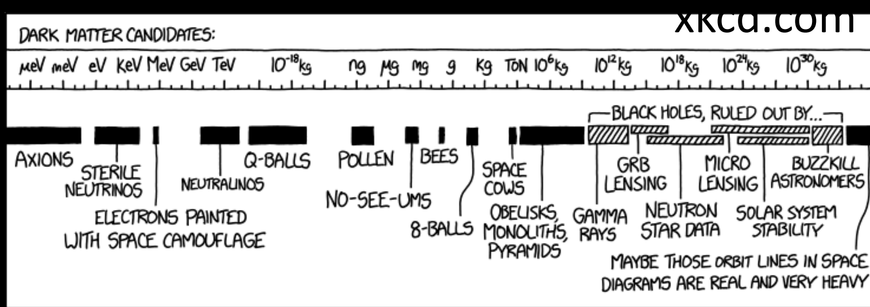
Coincidence Counter Analysis

- Looking for any peak above accidental coincidence probability level
  - Done with images
- In progress, but looking promising at least for Cf-252
- Interdisciplinary: of enormous interest to atmospheric science

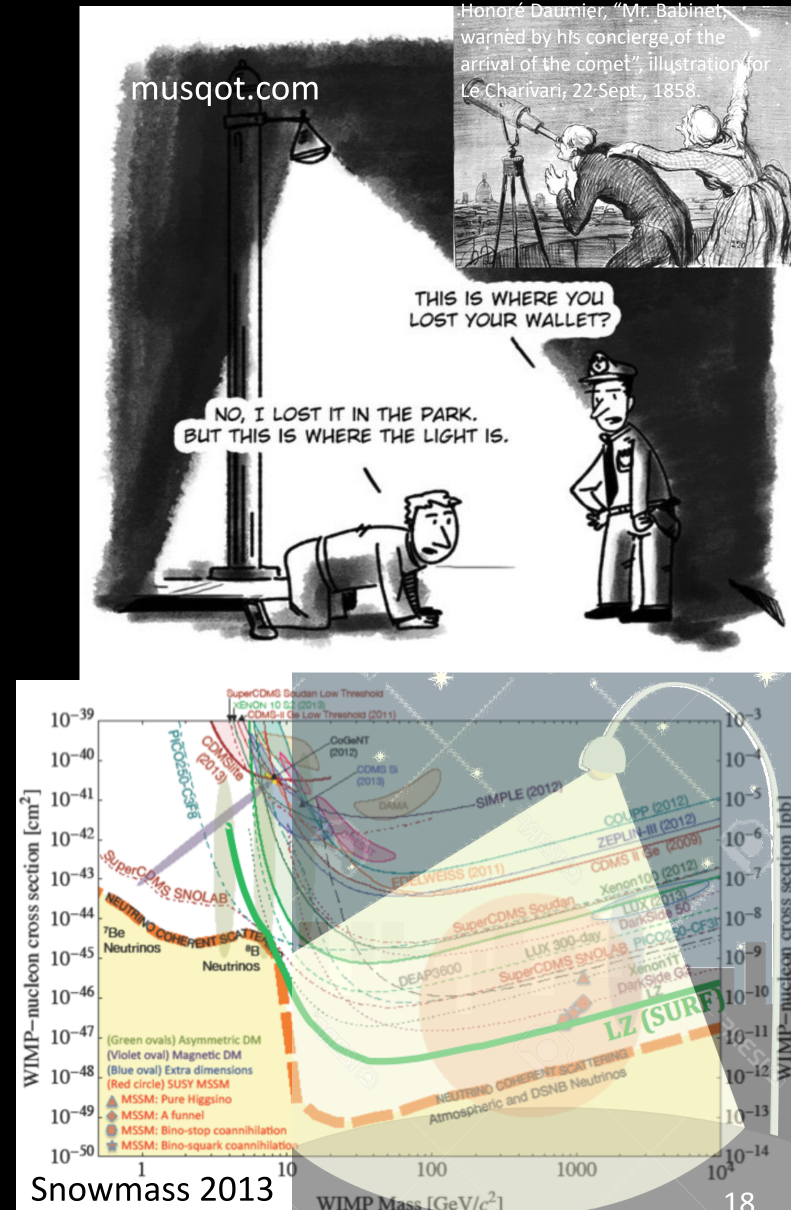


http://cosmicwatch.lns.mit.edu

Bigger Motivation: Dark Matter



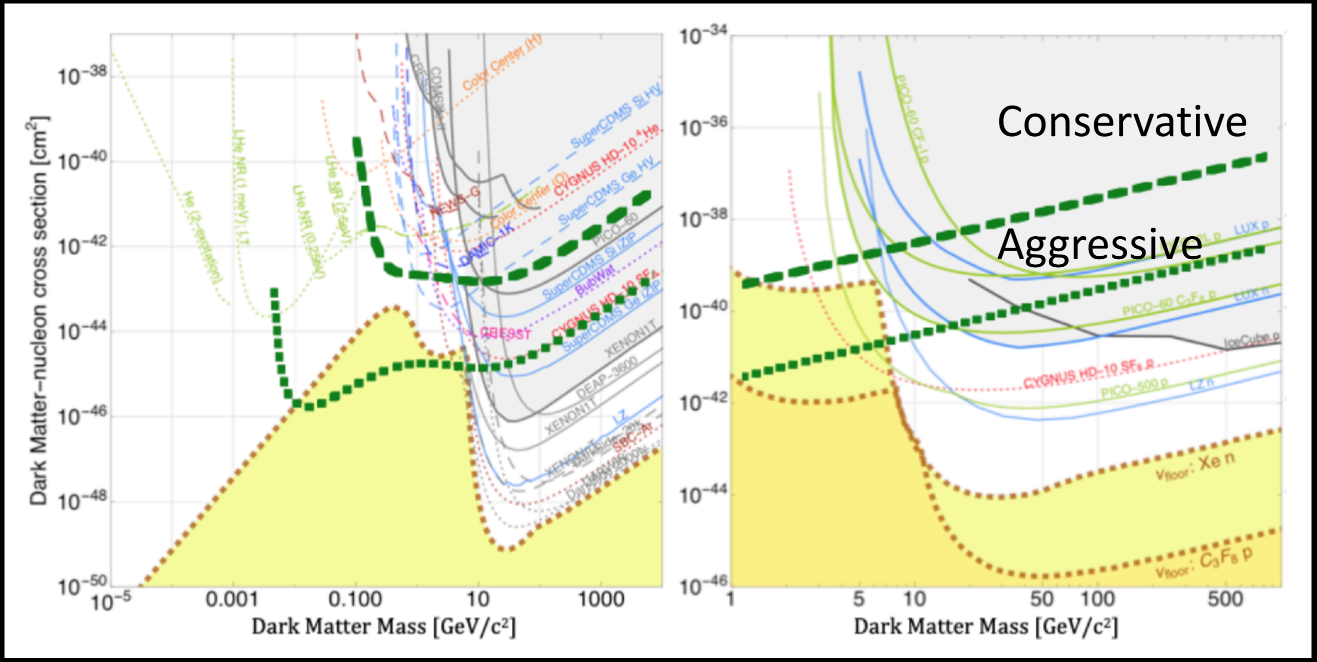
- Continued lack of discovery of dark matter as ~50-100 GeV/c^2 mass WIMP
  - Motivates looking elsewhere
- What is better target for lower-energy recoils, than the lightest possible target element, hydrogen?
  - Hydrogen bubble chamber would be great, but less practical
  - Other ideas exist already, so far from only game in town, even at sub-GeV
- Water is inexpensive and relatively easy to purify even on large scales (SNO, SuperK) while great at moderating n's
  - Cheap and scalable particle detection technology used in past already



Sensitivity to Vanilla Default WIMPs

borrowed plot from the DoE Cosmic Visions Report (arXiv:1707.04591) and overlaid our own curves

- Spin-independent (SI) and spin-dependent (SD)
  - Approaching the (lower for H) neutrino “floor”
- Dark photons and axions through e- scattering?



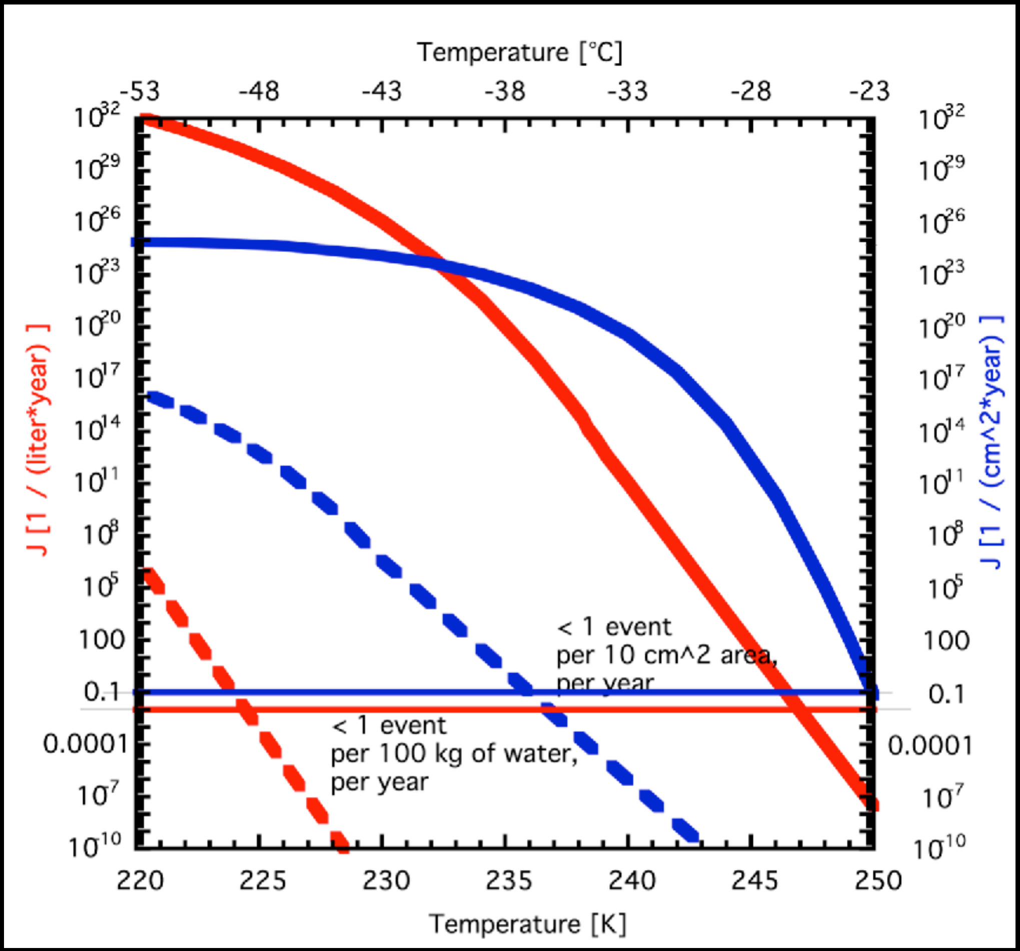
1 kg-year live exposure, at 12 eV energy threshold w/ low BG, underground
->e.g. only 1 kg for 1 yr!
100 kg-years, 16 meV is the lower curve

SI (left)
SD-proton (right)

fraction of the cost (and complications) of competing experiments at 100 MeV to 10 GeV! Potentially self-confirming

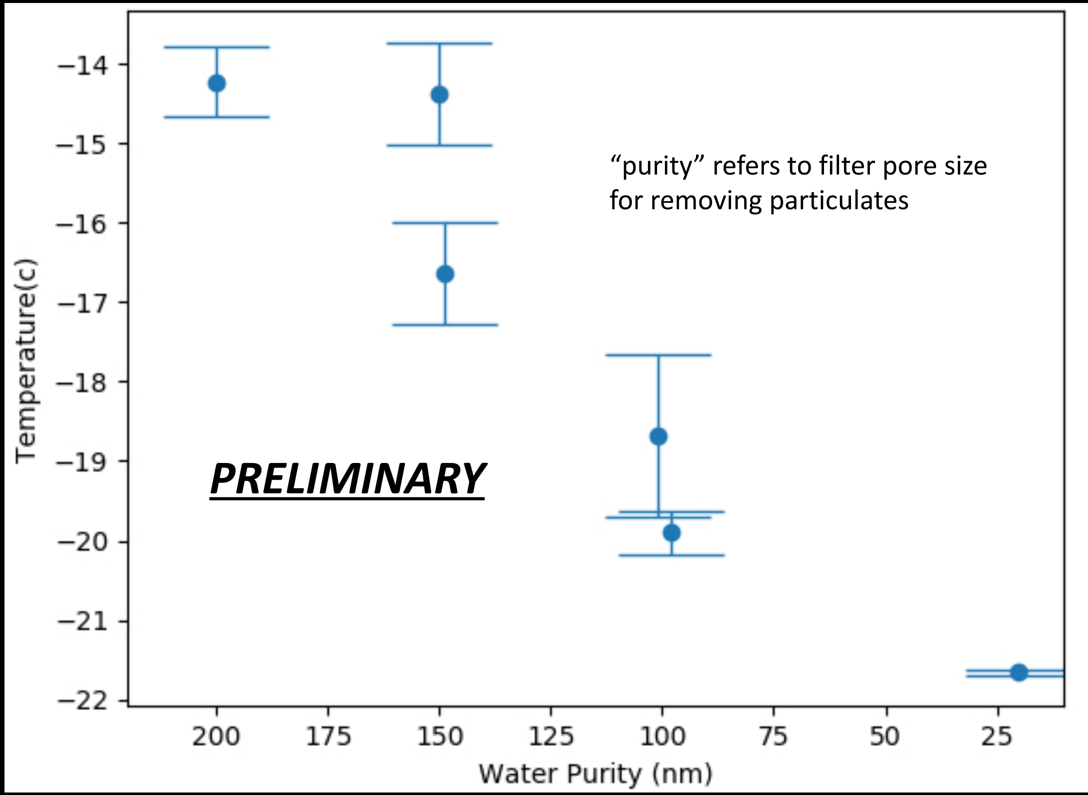
How to Make DM Discovery Possible

D. Barahona, "Thermodynamic derivation of the activation energy for ice nucleation," *Atmospheric Chemistry & Physics*, vol. 15, pp. 13,819–13,831, Dec. 2015.
D. Barahona, "On the thermodynamic and kinetic aspects of immersion ice nucleation," *Atmospheric Chemistry and Physics*, vol. 18, no. 23, pp. 17,119–17,141, 2018. [Online]. Available: https://www.atmos-chem-phys.net/18/17119/2018/
V. I. Khvorostyanov and J. A. Curry, "The theory of ice nucleation by heterogeneous freezing of deliquescent mixed CCN. Part II: Parcel model simulation," *Journal of the Atmospheric Sciences*, vol. 62, no. 2, pp. 261–285, 2005. [Online]. Available: https://doi.org/10.1175/JAS-3367.1



- Unclear whether to use homogeneous or heterogeneous nucleation energy thresholds
  - In either case, sub-keV threshold possible, even sub-eV
- Around ~240 K or -30 °C there appears to be a "sweet spot" of low threshold and 0 BG (from spontaneous nucleation)
  - Spontaneous rates drop precipitously with higher temperature
  - Analysis considers both the area and the volume

Measurement of Filtration Effect

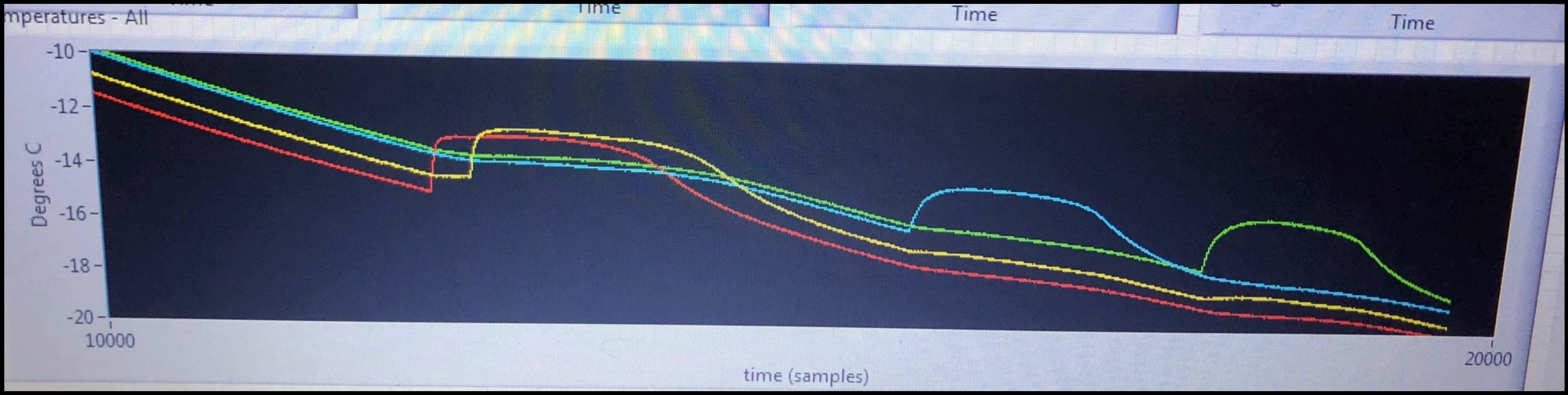


Min temperature achieved while supercooling before sample freezes

Four 1-mL water samples tested 6 times: each point on plot is set of 24 measurements, complete with statistical and systematic errors bars

Lowest point at right different: that is from the results published in PCCP

How to Optimize the Energy, dE/dx Thresholds: An Optimal Temperature!



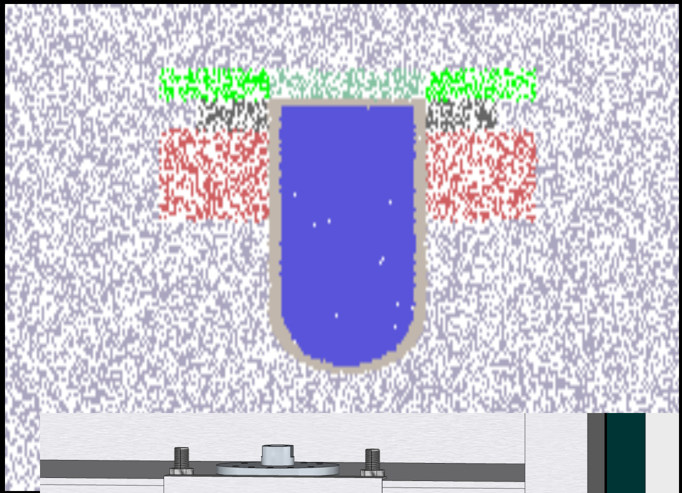
- Multi-dimensional search for lowest T's and longest supercool control (non-source) times
  - Across multiple small samples
- Buffer fluids top and bot, max volume, cool-down rate, initial temp, bath level, container

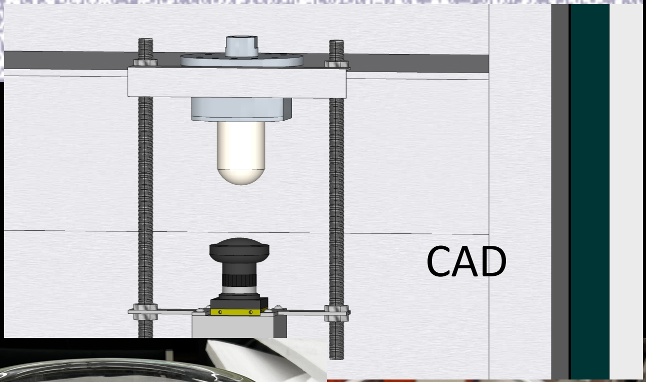
(Near-)Future Work

With colleague and collaborator Prof. Cecilia Levy

- More cameras (higher FPS)/mirror for 3-D recon
  - Automatic, including event type; snow ***directionality***?
- Lower threshold with lower T, hydrophobicity
  - Volume optimization, of water, and environment
- Increase the livetime (big current drawback, as it is too low). How to melt, then re-freeze?
  - ***Modular*** detector?
  - Extreme heat, lasers, microwaves, agitation
  - Supercooled droplet detector (ScDD)
- Full Geant4 sim, not just n & γ rates: #vertices
  - Molecular dynamics in more distant future
- The exhaustive characterizations of energy threshold
  - Possibly P too not just T, and more source types
- Hard: secure some $, start global program (Australia on board: Prof. Peter Wilson)

Geant4





new outer vessel
(Shruti De)

hydrophobic coat
(SUNY Poly)
for inner vessel

Conclusions, Challenges

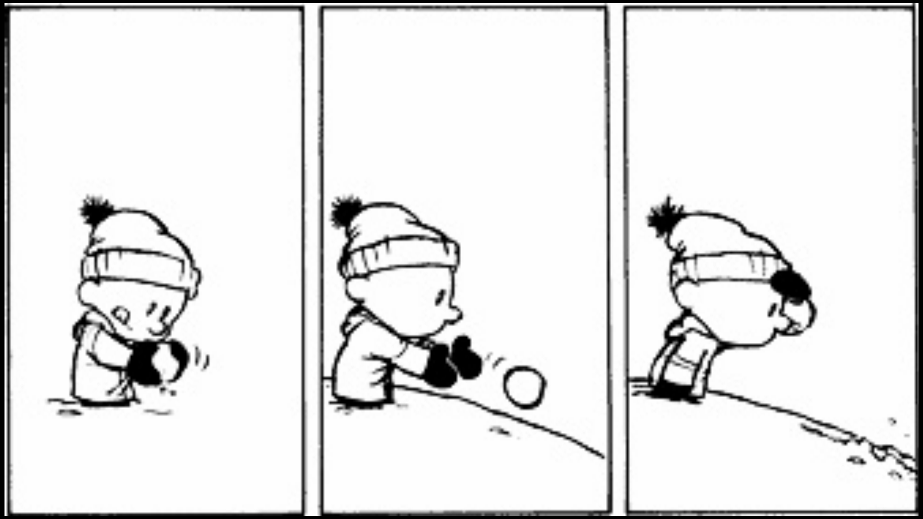
- Neutrons can make supercooled water freeze: a new discovery
- They can even multiply scatter, as they do in a bubble chamber!
- At least some types of events are coincident with a scintillator
- There is at least some degree of electron recoil (gamma) discrimination
- What are the actual backgrounds, from random nucleation, alphas,...?
- Energy threshold is not known, but likely sub-keV already at -20°C
- Need to calibrate it better. But looks good for low-mass DM & CEvNS
- Possible tangential relationship to other fields (CLOUD @CERN)
- All in all, this is a very promising start to a RE-discovered technology
- So much more we can do: CEvNS with a low-mass, even-even nucleus?
- D_2O for normalizing low-E neutrino fluxes from stopped pion beams?

Multiple Thanks

https://www.nps.gov/dena/learn/nature/arcticgroundsquirrel.htm

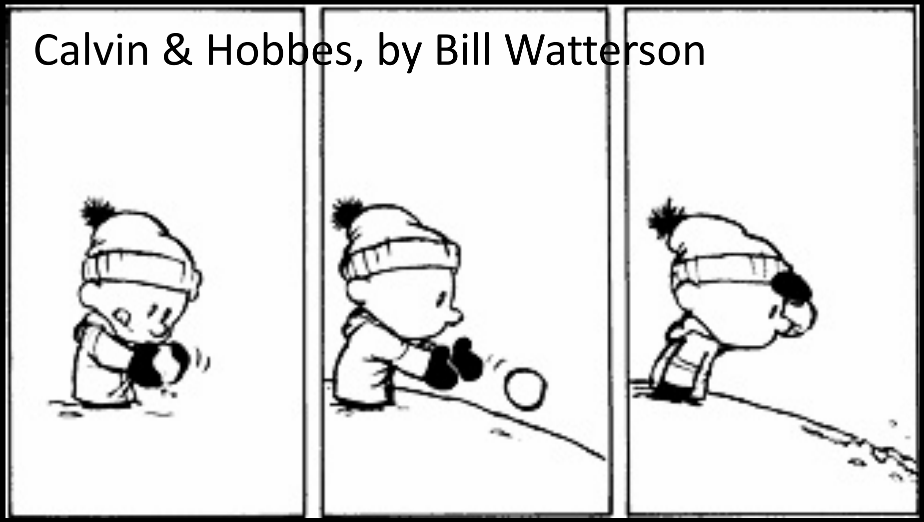
- I wish to also thank my collaborators at UCLA, BNL, RPI, Penn State, and Duke/TUNL, all trying to start a large program with UAlbany

- Questions today??



This research was funded by a UAlbany NSF SAGES/WISH award to Prof. Cecilia Levy, SUNY/RF PIFRS & FRAP-A awards for Prof. Matthew Szydagis, and startup

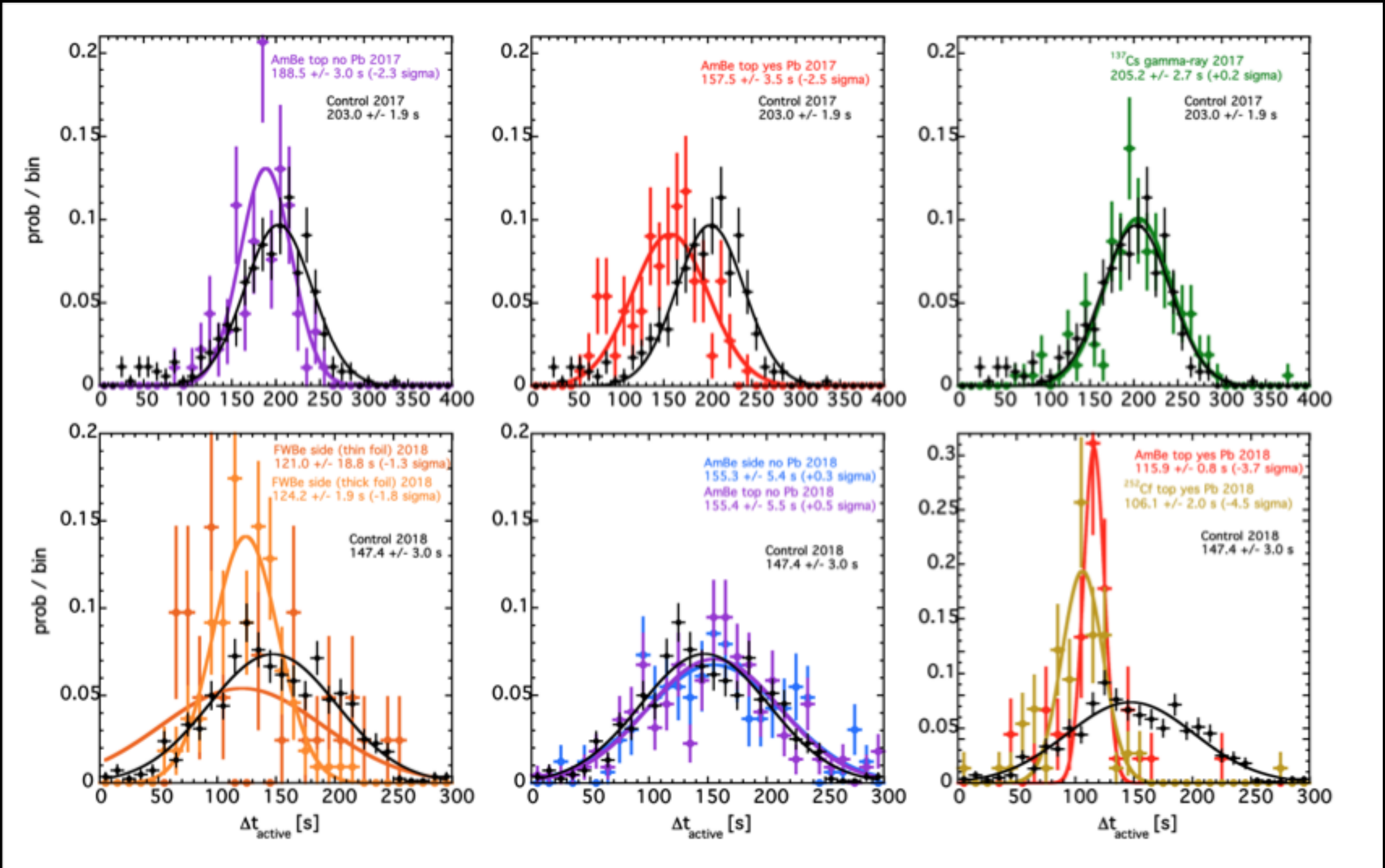
A Few Backup Slides

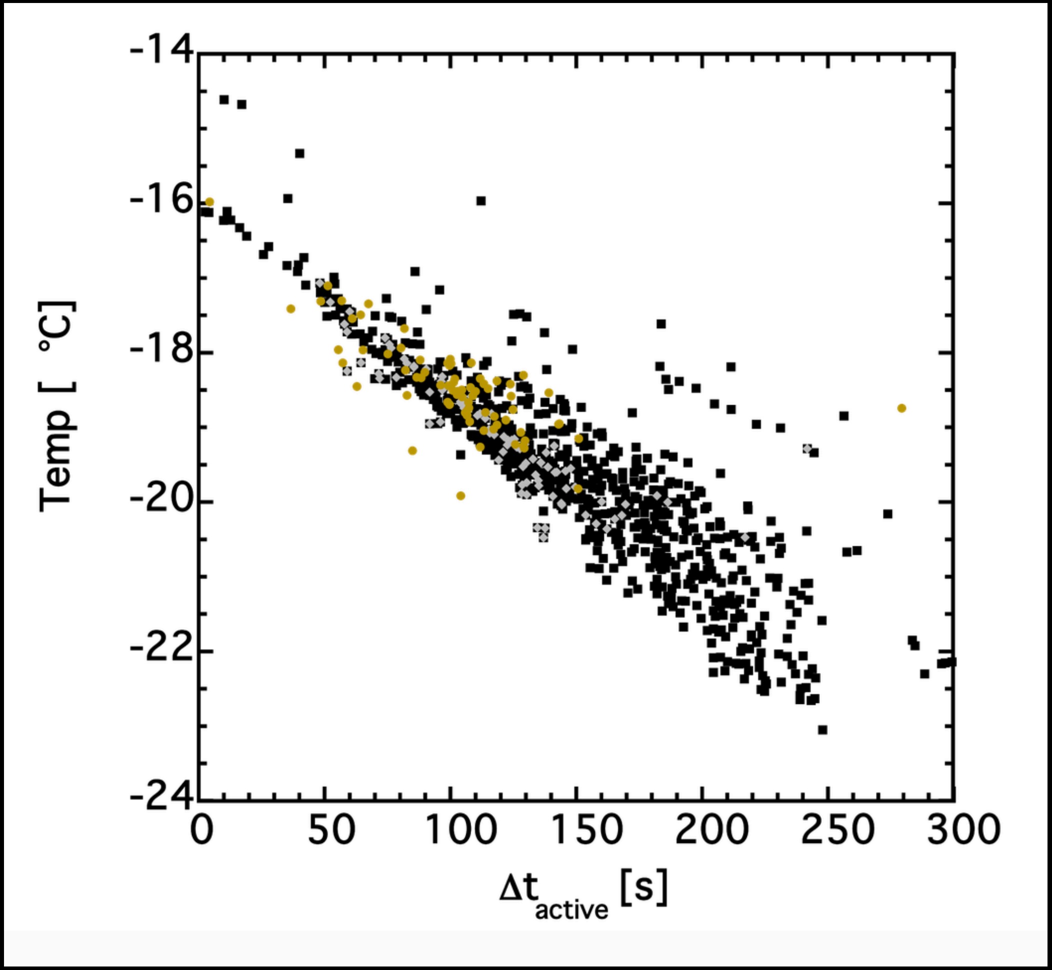


time water spent “active” (< -16 °C)

Calibration Type	$< \Delta t_{act} >$ [s]	σ	$< T_{min} >$ [°C]	σ
Ctrl (no source) ’17	190.6 $^{\pm 2.8}_{\pm 4.2}$	-	-21.46 $^{\pm 0.07}_{\pm 0.09}$	-
AmBe Top no Pb	183.1 $^{\pm 3.5}_{\pm 3.2}$	-1.1	-21.85 $^{\pm 0.10}_{\pm 0.39}$	-0.9
AmBe Top yes Pb	150.5 $^{\pm 4.1}_{\pm 17}$	-2.2	-20.33 $^{\pm 0.13}_{\pm 0.17}$	4.7
^{137}Cs γ-ray Top	201.8 $^{\pm 3.6}_{\pm 11}$	0.9	-21.21 $^{\pm 0.12}_{\pm 0.23}$	0.9
Control 2018	149.5 $^{\pm 1.9}_{\pm 7.6}$	-	-19.63 $^{\pm 0.04}_{\pm 0.19}$	-
FWBe side (thin)	137.7 $^{\pm 16}_{\pm 0.0}$	-0.7	-19.71 $^{\pm 0.29}_{\pm 0}$	-0.2
FWBe side (thick)	124.6 $^{\pm 2.9}_{\pm 9.9}$	-1.9	-19.33 $^{\pm 0.07}_{\pm 0.27}$	0.9
AmBe side no Pb	158.9 $^{\pm 4.9}_{\pm 23}$	0.4	-19.96 $^{\pm 0.09}_{\pm 0.44}$	-0.7
AmBe Top no Pb	154.9 $^{\pm 3.8}_{\pm 12}$	0.4	-19.73 $^{\pm 0.09}_{\pm 0.31}$	-0.3
AmBe Top yes Pb	113.8 $^{\pm 4.4}_{\pm 2.1}$	-3.9	-18.88 $^{\pm 0.08}_{\pm 0.01}$	3.5
^{252}Cf Top yes Pb	102.5 $^{\pm 4.0}_{\pm 3.8}$	-4.9	-18.46 $^{\pm 0.07}_{\pm 0.22}$	3.8

Graphical form of course: numbers don't agree with last slide, because this is with fits




-14
-16
-18
-20
-22
-24
0
50
100
150
200
250
300
Temp [°C]
Δt_{active} [s]


VISSANI