Activation of Air and Concrete in Medical Isotope Production Cyclotron Facilities

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Outline

- Motivation
- Methods
- Air activation
- Concrete activation
- Conclusions

Motivation

- Neutrons produced in cyclotron target via (p,Xn)
  - can activate materials in vault
  - air activation can pose a radiological hazard to workers
  - concrete activation can become a disposal problem
- Why the renewed CNSC interest?
  - cyclotrons have become much more powerful
  - old days ~ 60 µA and 2 hour runs for F-18 FDG production
  - now could have 500 µA and 6 hour runs for Tc-99m production
  - 25 times more activation!

Methods (1/2)

Monte Carlo simulations

- MCNP5 (1% statistics)
- proton beams 18 & 24 MeV
- simplified vault geometry
- F-18 and Tc-99m neutron source spectra – assumed isotropic
- explore sensitivity of results to
  - changes in vault design
  - source energy
  - source location
  - polyethylene (with/without boron) shielding around targets

Methods (2/2)

Collaboration with UOIT co-op students

- Rob Shackelton (air activation)
- Devon Carr (concrete activation)
- Audrie Ismail (concrete activation)

Air activation

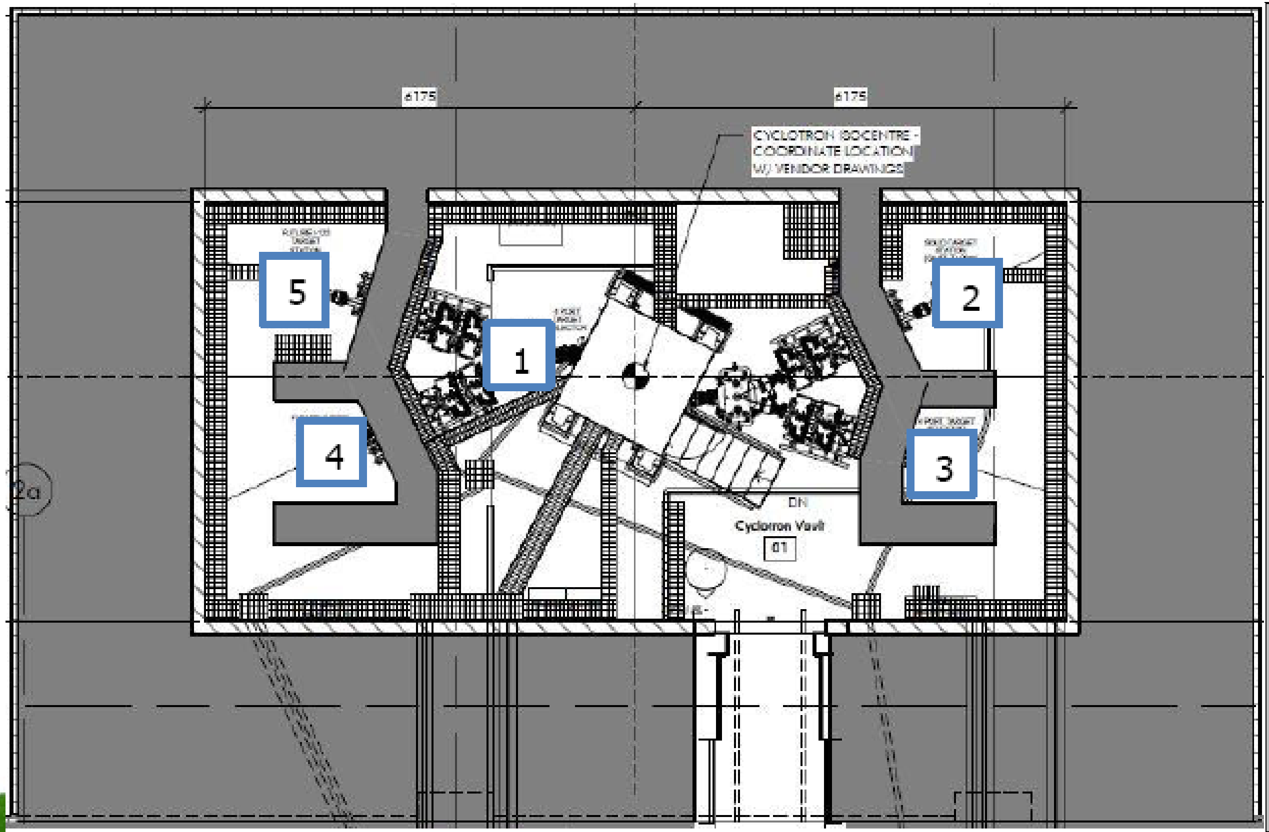
- **Nitrogen 78%of air**
  - N-14(n,p)C-14 (78% of air)
  - C-14: 5,730 year half-life, soft β – little radiological consequence
- **Oxygen 21% of air**
  - O-16(n,p)N-16 (21% of air, 10 MeV threshold)
  - N-16: 7s half life, 6 MeV γ – little radiological consequence
- **Argon ~ 1% of air**
  - Ar-40(n,γ)Ar-41 (0.93% of air)
  - Ar-41: 1.8h half life, hard β, 1.3 MeV γ

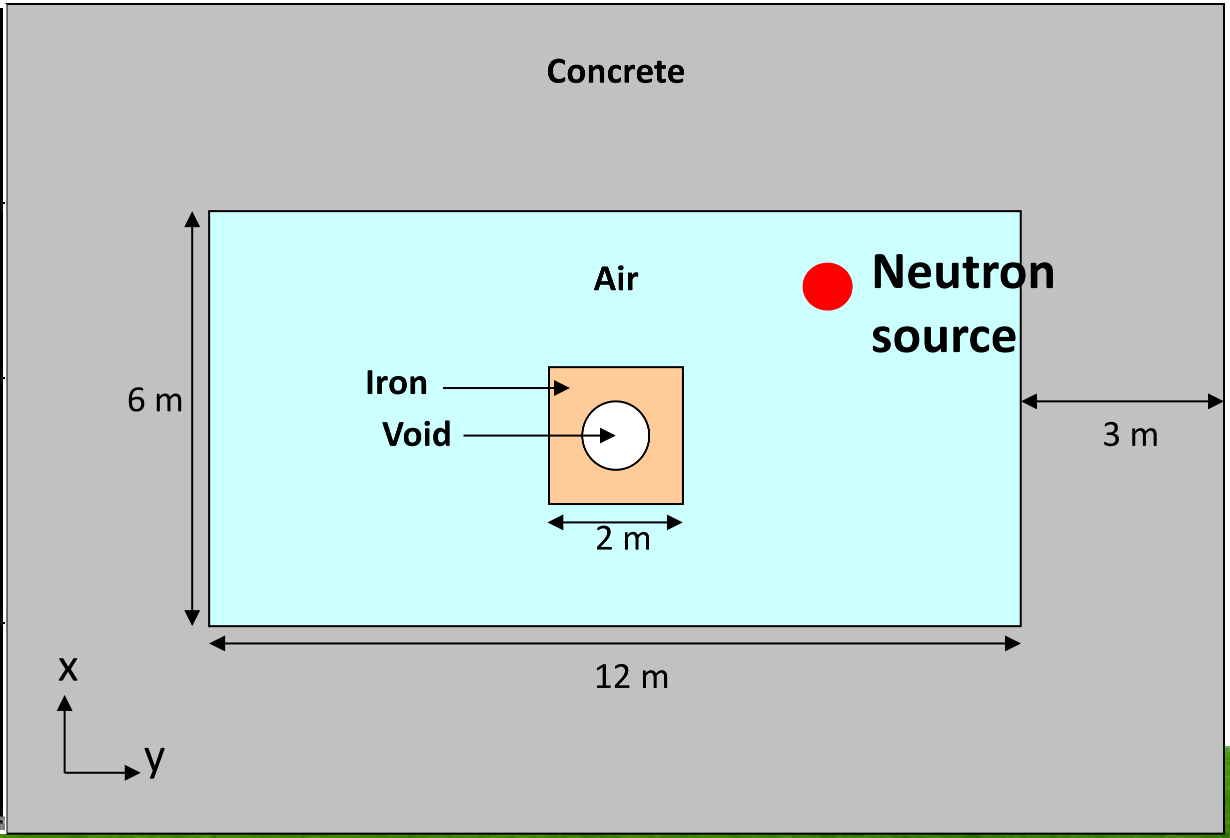
Dominant hazard is Ar-41 activity

Geometry and Materials

Go from complex to extremely simple

Reveals essential features and save computing time





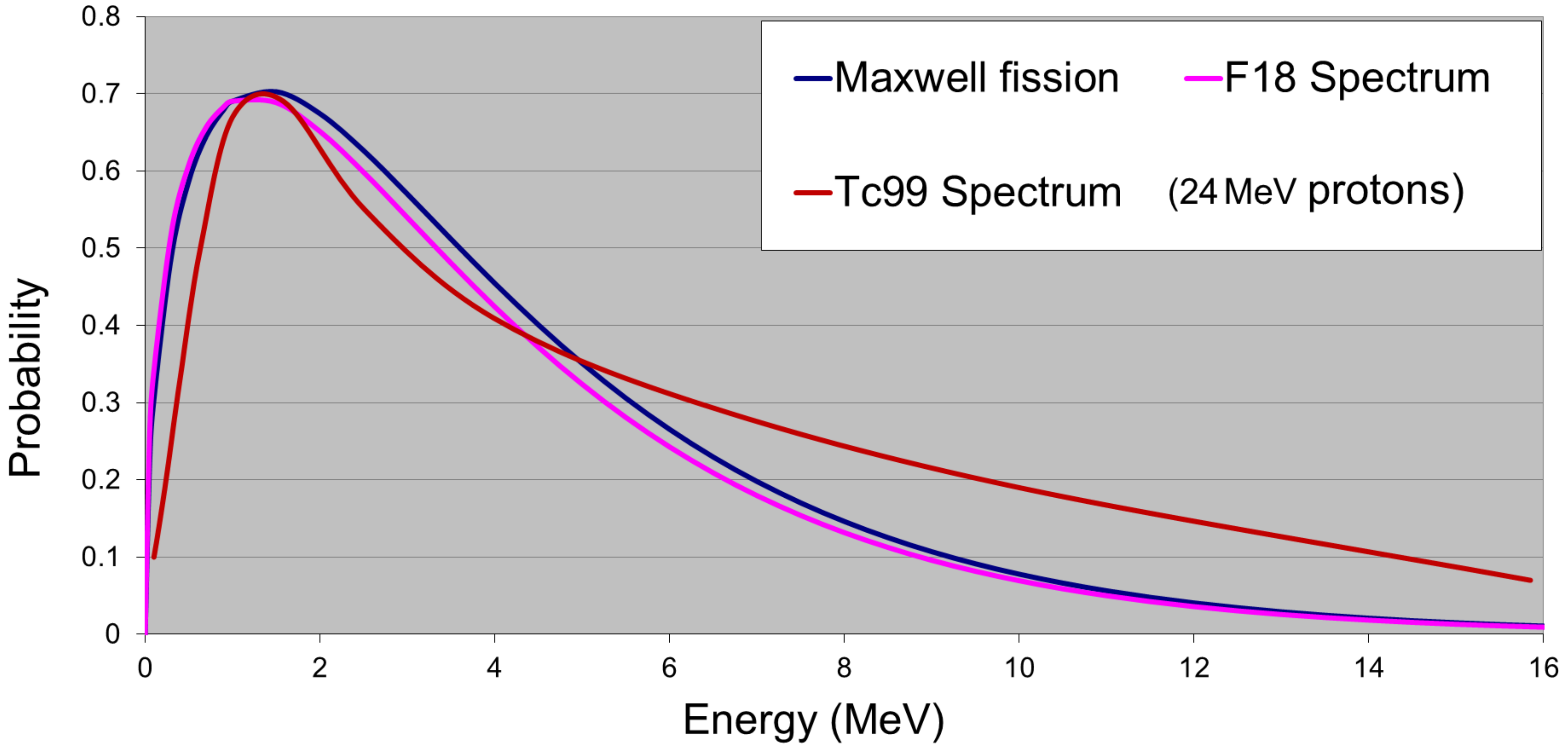
Neutron Source Spectra (1/2)

- Neutron point source emulates target during irradiation (isotropic)
- F-18 thick-target spectrum from Mendez et. al. [1]

$$N(E) = 0.27E^{0.45}e^{-\frac{E}{2.7}}$$

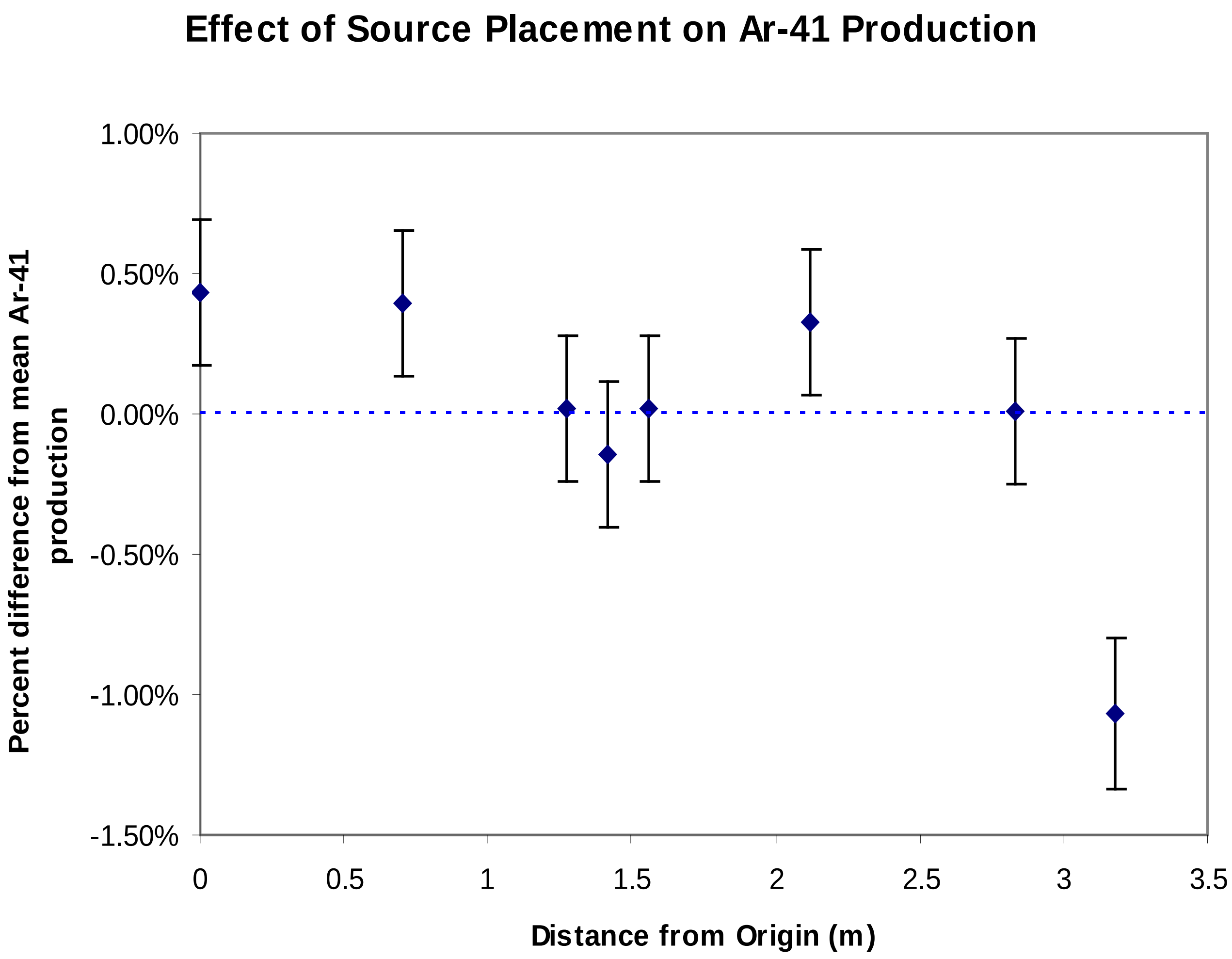
  - approximated by Maxwell fission spectrum
  - 150 logarithmically spaced energy bins
- Tc-99m spectrum from nested neutron spectrometer data [2]
  - Histogram representation
- All spectra automatically normalized by MCNP

Neutron Source Spectra (2/2)



Neutron Source Placement

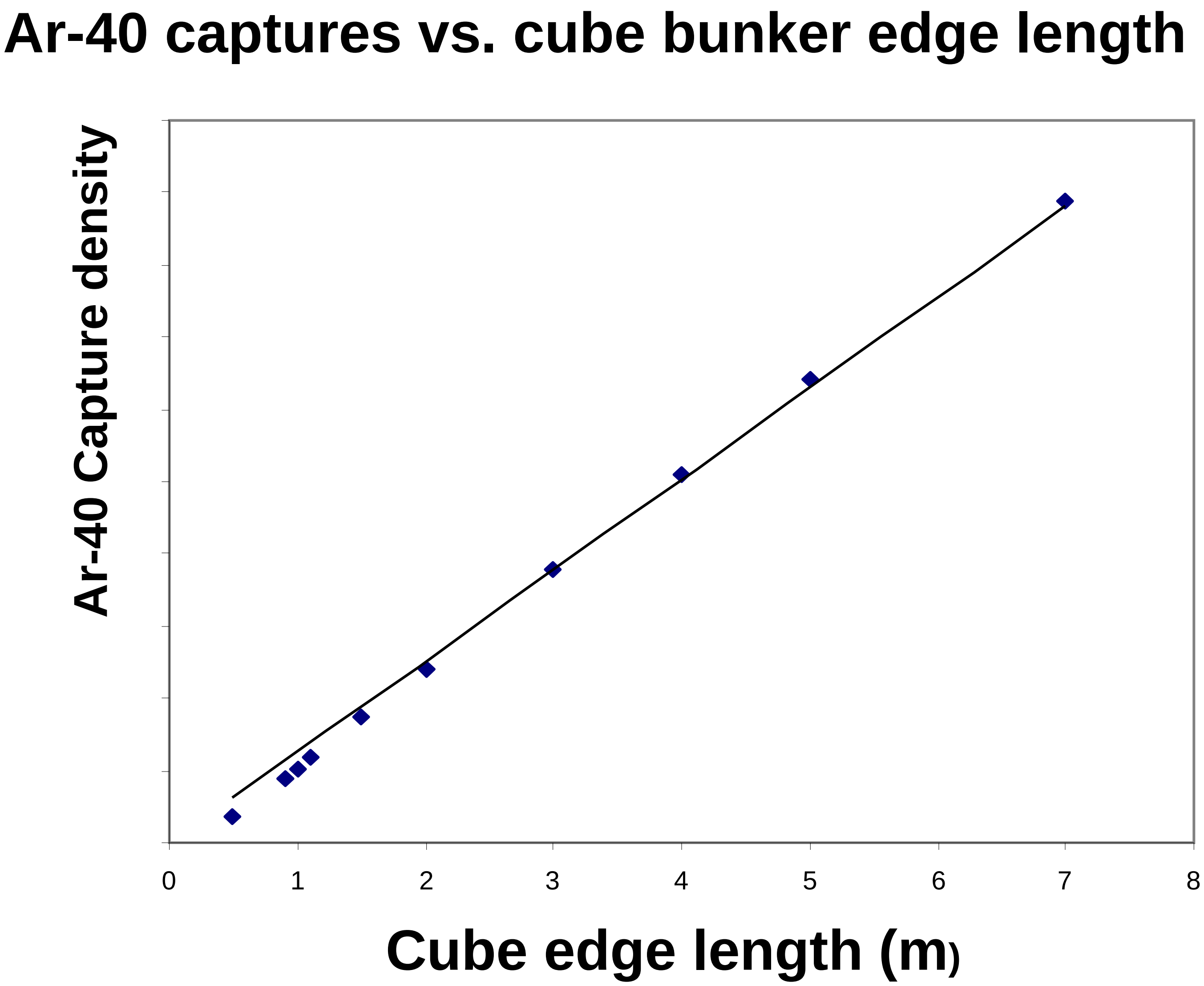
- 3m x 5m x 5m vault
- Eight source positions
- Starting at origin, moving diagonally into corner
- Considering statistical error, effect of source location is negligible



Vault Size

Linearity result of large neutron mean free path in air ~62 m

Deviation from linearity for irregular shapes and very small vaults



Cyclotron and Source Energy

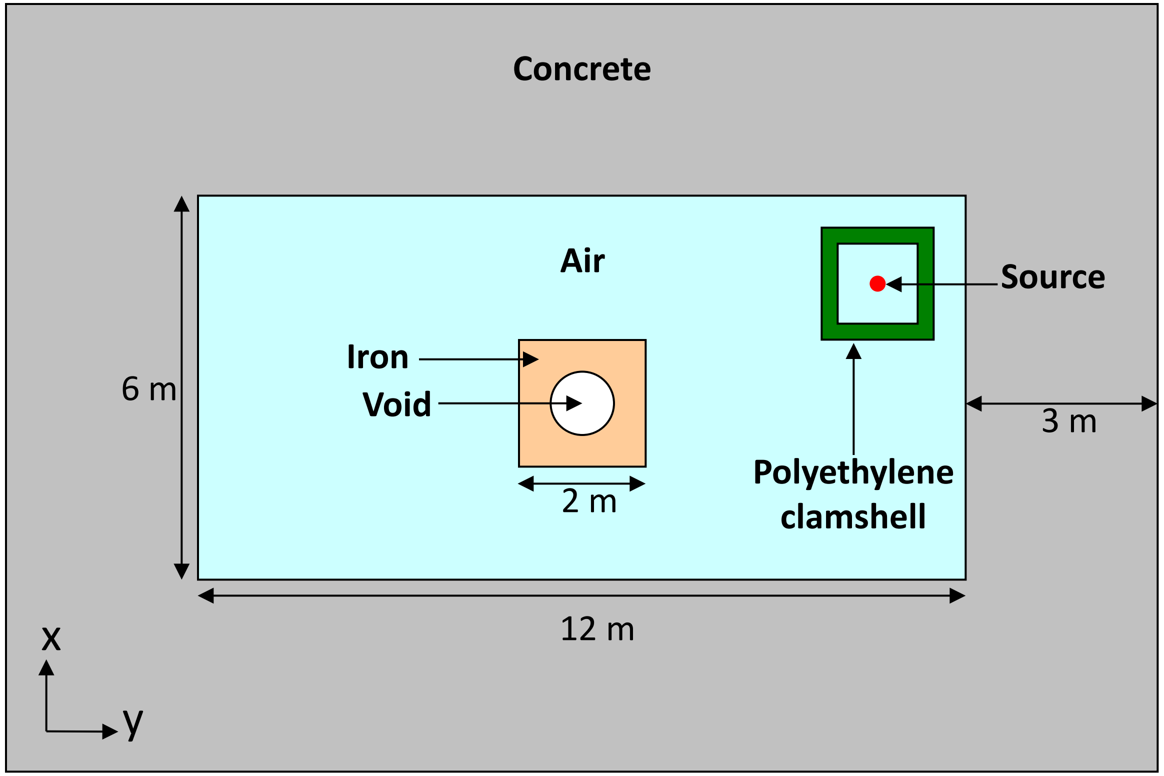
- Iron cyclotron added to centre of the room
  - Slight decrease in production
  - Cyclotron density has no effect

- Source energy tests: (3m x 6m x 12m vault with cyclotron)

Neutron energy	Ar-41 production/incident neutron/cm^3
F-18 spectrum	3.0 E-5
Tc-99 spectrum	3.6 E-5
Isotropic 1 keV	5.2 E-5
Isotropic 0.025 eV	12.0 E-5

Target Clamshell

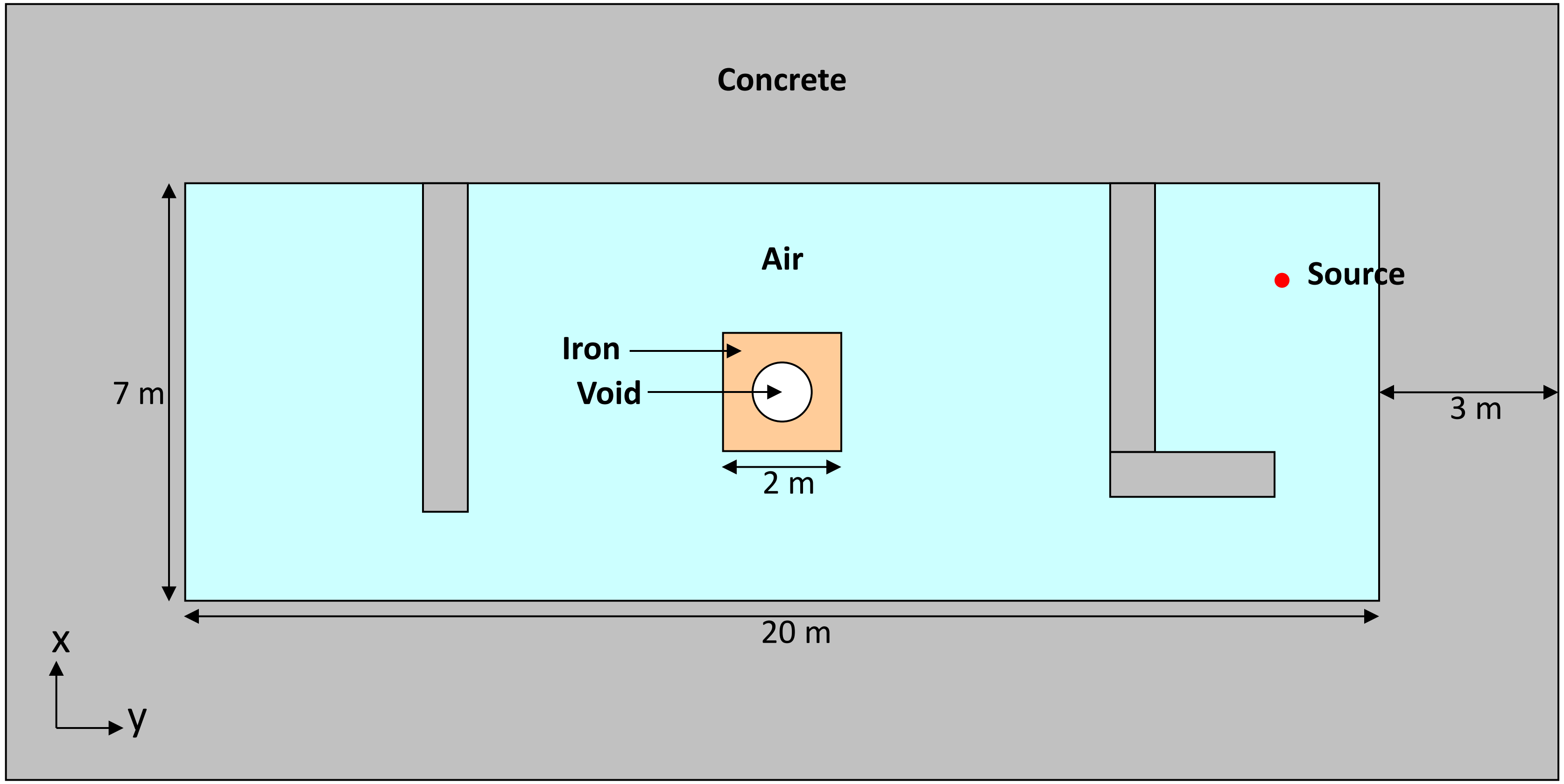
- Polyethylene target shielding
  - 5, 10, 50cm thickness
  - No boron
- Critical thickness
  - Thin shield thermalizes neutrons
  - >10cm necessary to capture
- γ shielding requirements



Partition Walls

- Wall placement below resulted in 15% decrease in Ar-41 production

- Objects in vault reduce air activation
- Justifies simplified geometry



Ar-41 Activity

- Results are in captures/neutron - How many neutrons per μA of beam?
  - IAEA TRS-468[x] gives saturation activity of F-18 at different beam energies; at saturation
- Neutron production rate = F-18 decays/s (Bq)
  - extrapolated to 24 MeV → 1.6 E10 n/s/μA
- For Mo-99 (NNS @19 MeV) → 3.2 E10 n/s/μA

- **At saturation**
- F-18 at 150 μA - (3 x 5 x 5 m vault) ~2 mCi
- Tc-99 at 750 μA – (3 x 6 x 12 m vault) ~ 12 mCi

Dosimetry – Dose rates

All dose rates in μSv/h	F-18 @ 150 μA 3m x 5m x 5m	Tc-99 @ 750 μA 3m x 6m x 12m
External gamma[3]	5.1	17.3
Inhalation[4]	0.1	0.3
Skin[5]	0.9	2.2

- Assumes saturation production
- Skin dose – upper limit – assumes no clothing

Air Activation - Summary

1. Objects in vault reduces air activation
   - Justifies simplified geometry
2. Air activation is not a problem for F-18
   - Results are for saturation of Ar-41 (half-life 1.8 h)
   - Runs are ~ 3 hours and typical F-18 runs are shorter
3. For Tc-99 may be a problem
   - Runs are ~ 6 hours and beam current ~ 3 times higher
4. Ventilation reduces problem dramatically
   - Less time to build up Ar-41 in vault and less exposure time – 1 hour air exchange time reduces dose by a factor of ~10

Concrete Activation

This produces radioactive waste, affecting decommissioning costs

1. How deep does it go?
2. What do polyethylene layers (with/without boron) do?
3. Is it on all inner vault surfaces? Or is it localized?

Literature Review (1/2)

Decommissioning a cyclotron [6]

20-year-old 17 MeV Scanditronix cyclotron (~40 µA)

- 40 tons of low-level radioactive waste including the concrete vault wall
- Activities with $\tau_{½}$ > 1 year

Isotope	Measured activity (Bq/g)	UCL (Bq/g)
Co-60	**0.068**	0.1
Cs-134	0.005	0.1
Eu-152	**0.083**	0.1
Eu-154	0.010	0.1
Mn-54 *	0.016	0.1
Total	**0.18**	**0.1**

UCL: unconditional clearance level at which material **can be thrown out as non-radioactive,**

* Made by fast neutrons via (n,p) reaction rather than (n,γ)

Literature Review (2/2)

Reactor study *[7]*

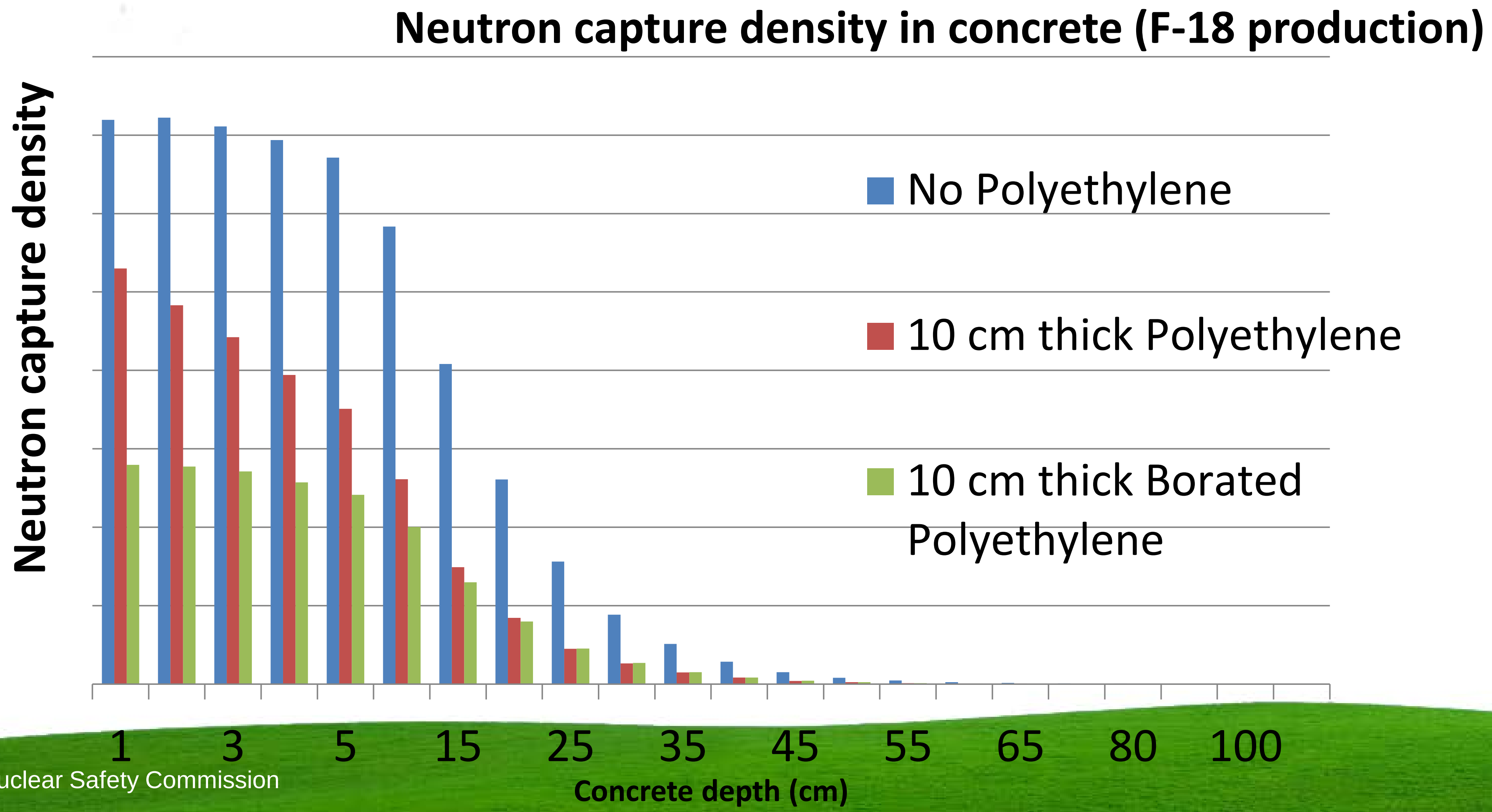
Ordinary concrete sample in TRIGA reactor in Slovenia

30 minute exposure @ neutron flux of 6.8 10^{12} n/s/cm^2

Principal activities found Eu-152 and Co-60 at 6 Bq/g

Conclusion – concrete activation could be a problem with the new cyclotrons

Absorption depth



Polyethylene around target

Poly layer thickness (cm)	Percentage of neutrons captured in poly layer	Percentage of neutrons captured in borated poly layer
5	17.1	32.0
10	52.0	65.8
15	72.5	82.9
20	82.6	91.1

- Results show how deep a sacrificial layer should be (if used)
- Regular poly is almost as good as borated poly
- Either option much cheaper than sacrificial layers in the vault
- And can be put in after vault construction

Spatial Distribution in Vault

We now know activation goes down to depth ~20cm

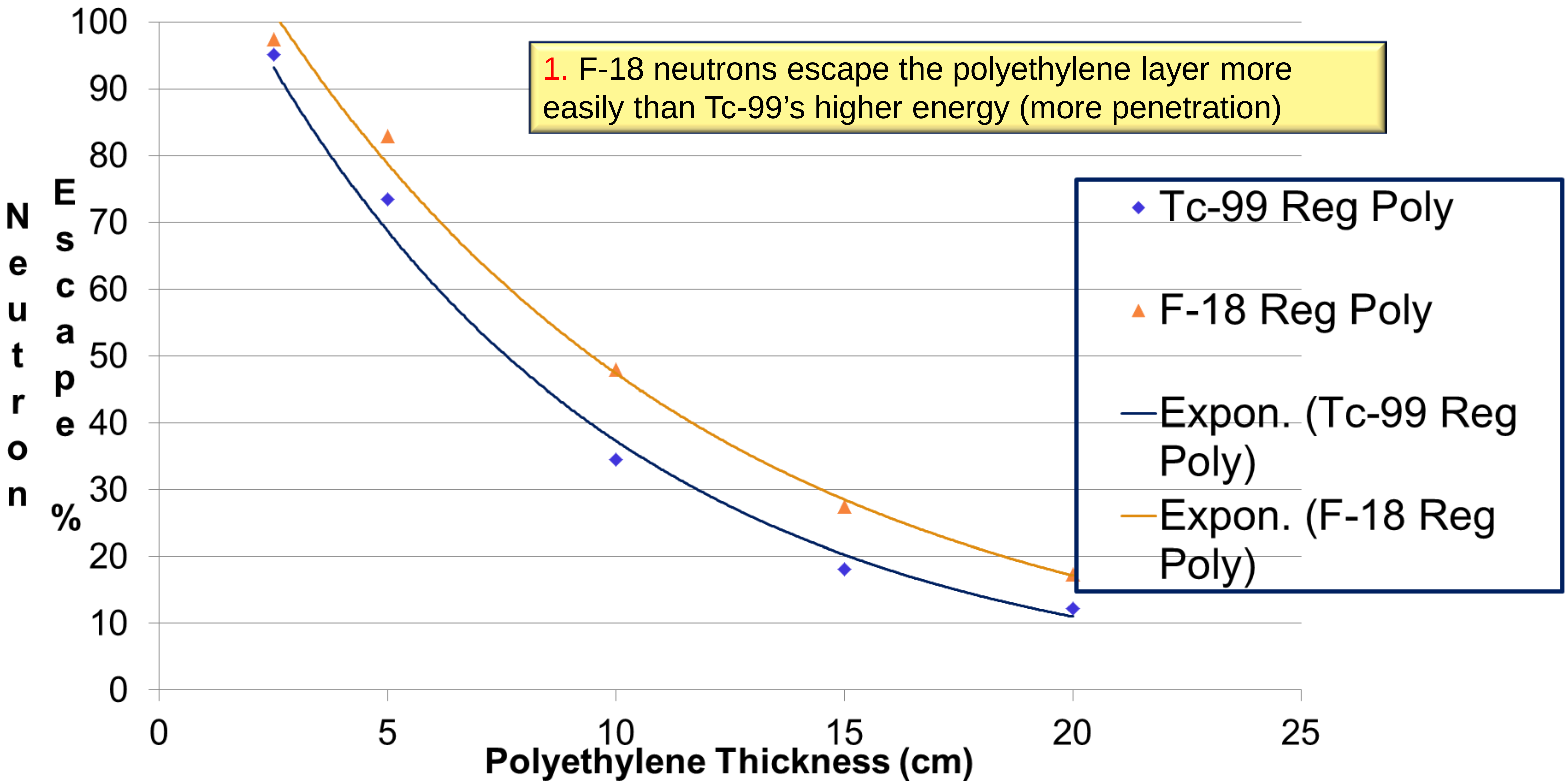
But this was averaged over whole inner vault surface

Is it on all inner vault surfaces ? Or is it localized?

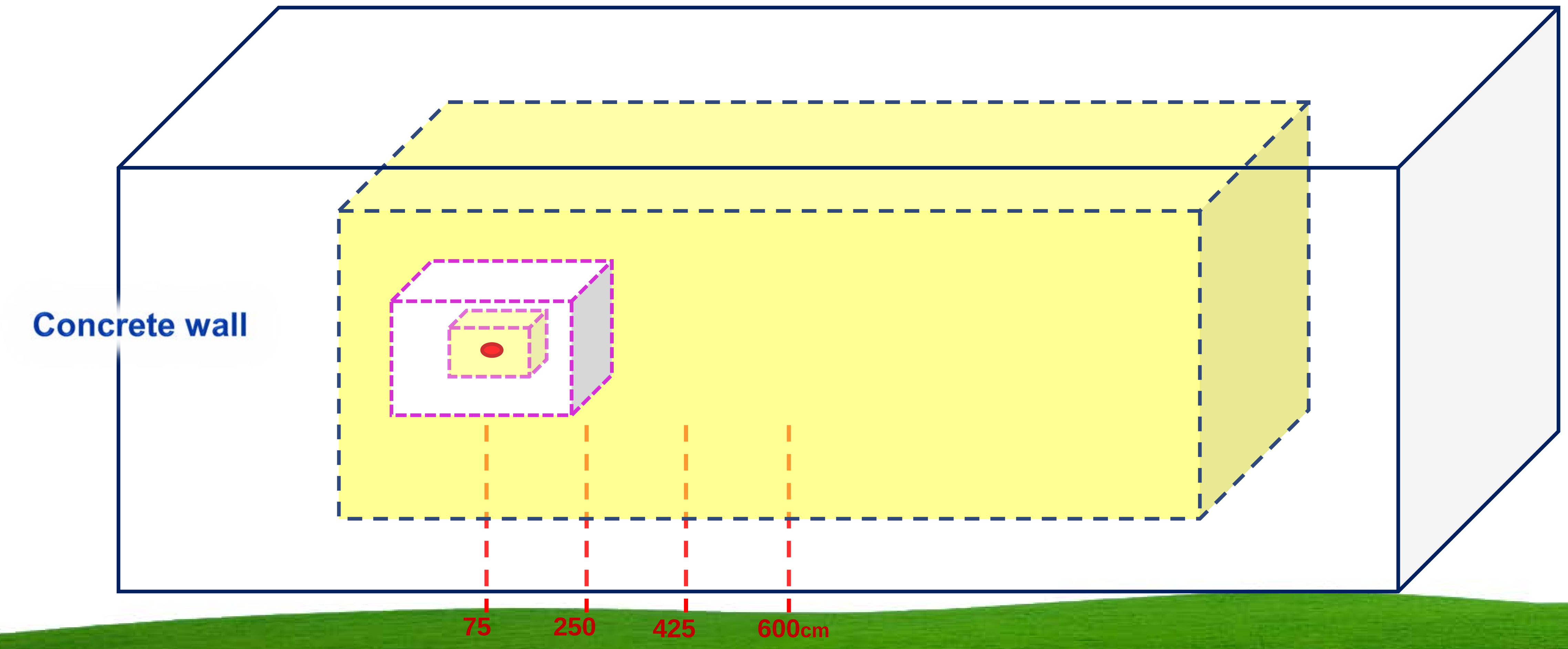
Investigated

1. Tc-99 vs F-18
2. Moving the source position inside the vault
3. Lateral distribution of neutron capture density within 1 side of the wall

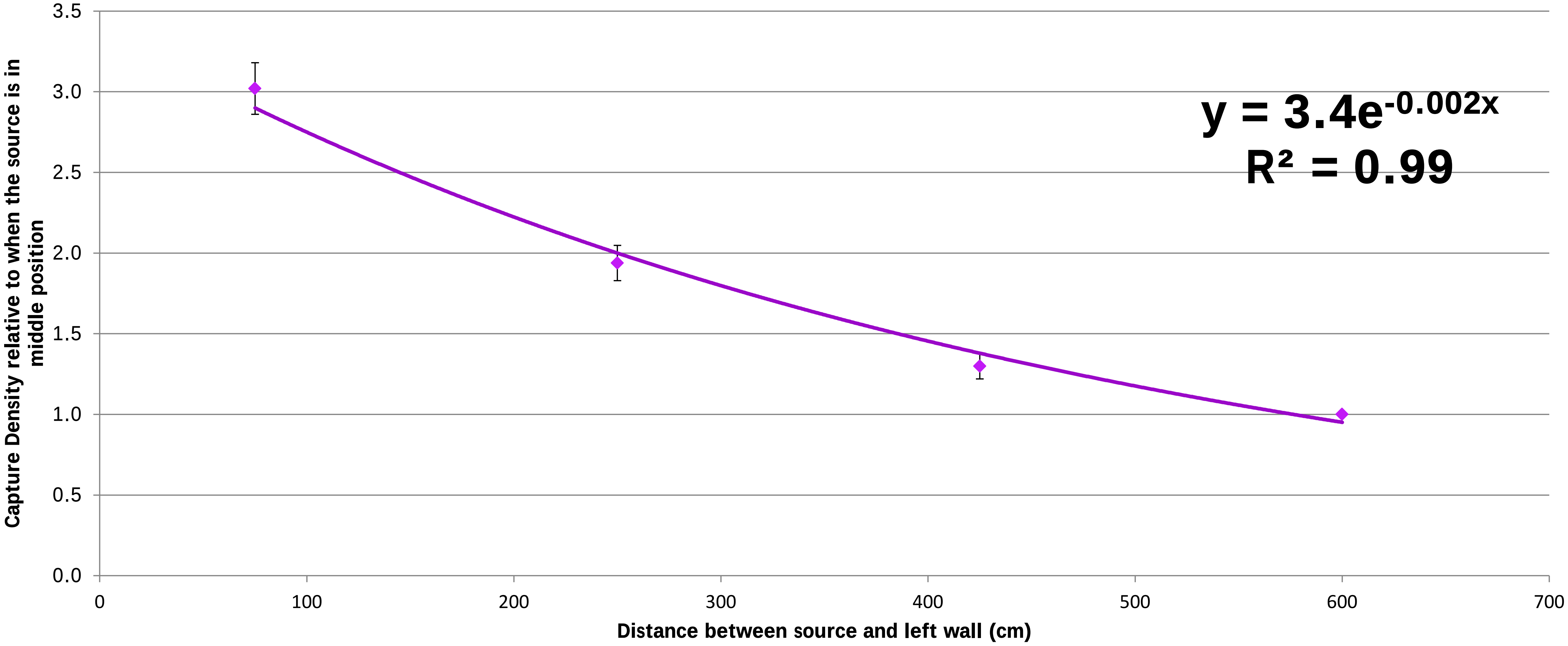
1. Neutron Escape Percentage for Regular Poly Around Target - Tc-99 versus F-18 source



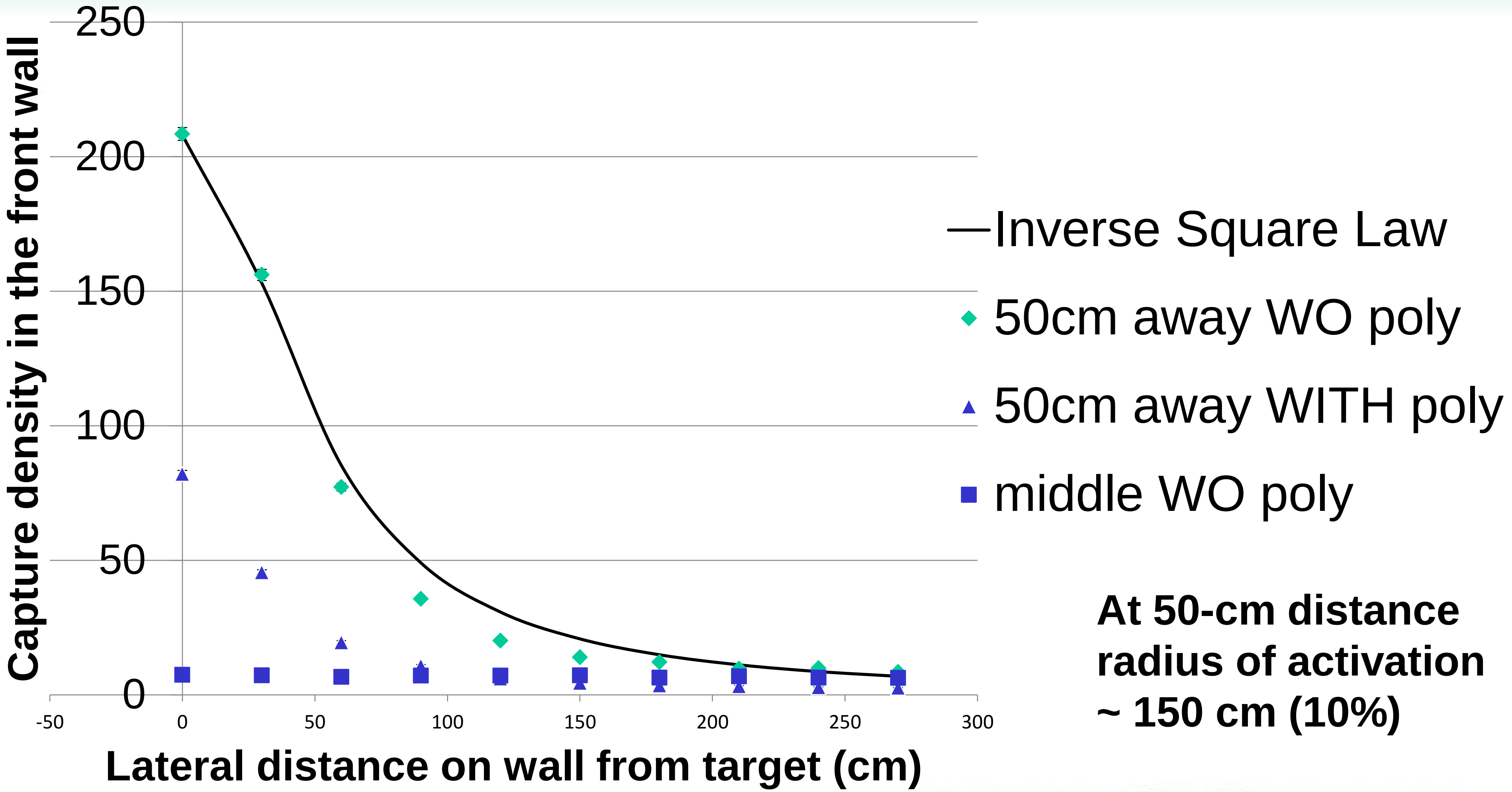
2. Moving the Source Position on Y-Axis With 20-cm Thick Polyethylene Layer



2. Relative Capture Density of the Left Wall With Respect to Tc-99 Source Position



3. Lateral Distribution of Neutron Capture Density in Near Wall



At 50-cm distance radius of activation ~ 150 cm (10%)

Regulatory Impact

1) Compare cyclotron neutron flux with TRIGA reactor flux

- After 1 year at full operation → 0.35 Bq/g of Eu-152 and 0.33 Bq/g for Co-60 ***(measurable)***
- After 25 years operation → 7 Bq/g for Eu-152 and 3 Bq/g of Co-60

2)100 x regulatory limit for disposal as non-radioactive waste

3) If no steps are taken, it will impact decommissioning cost and possibly financial guarantee

Conclusions

- **Air activation** – Not a big deal and easily controlled through
  - ventilation
  - restricted access for a few hours (normal to allow cyclotron to cool off)
  - detection of Ar-41 by area monitor in vault
- **Concrete activation**
  - could be a challenge for decommissioning
  - localized to concrete near target – including floor

For both problems, suggest borated poly around target

- **Experiment – activate sample of vault concrete & analyze by γ spectroscopy**
- Reactor neutron spectrum not quite the same as cyclotron neutron spectrum
- Your concrete may have different impurities

Questions?

References

[1] Mendez R. et. al. “Study of the neutron field around a PET cyclotron”. IRPA11 conference presentation, May 2004.

[2] Debeau J. et. al. “The Measurement of Neutron Energy Spectra in the High Neutron Flux Environments of Medical Accelerators Using Nested Neutron Spectrometer” (2013 CRPA conference poster).

[3] *Ar-41 source modeled as uniform equivalent sphere. Specific Gamma constant* from Delacroix D. et al., Rad. Prot. Dosimetry v. 98, no. 1 pp. 9-18 (2002). [4] Thériault B. “Inhalation dose coefficient for Ar-41”, private com. (2012).

[5] *β skin dose coefficient linearly extrapolated from data of* - Fell TP, Rad. Prot. Dosimetry V. 36 No. 1, pp. 31-35 (1991).

[6] Sunderland J. et al. “Considerations, measurements and logistics associated with low energy cyclotron decommissioning (2011)”, AIP Conf. Proc. 1509, 16 (2012): http://dx.doi.org/10.1063/1.4773931.

[7] Zagar T., Ravnik M., “Measurement of Neutron Activation In Concrete Samples”, Proc. Int. Conf. “Nuclear Energy In Central Europe 2000”.

Initial Results

- 3m x 5m x 5m empty vault with point source