PROPOSAL

Measurement of the $^{35}Cl(n,\gamma)$ cross section at n_TOF EAR1

I. Porras[1], T. Wright[2], S. Bennett[2], F. Ogállar[1,3], J. Praena[1], M. Sabaté-Gilarte[3], P. Torres-Sánchez[1], F. Arias de Saavedra[1], C. Lederer-Woods[4], S.J. Lonsdale[4], R. Garg[4], M. Dietz[4] and the n_TOF Collaboration.

[1]Universidad de Granada, Spain
[2]University of Manchester, UK
[3]European Organization for Nuclear Research, Switzerland
[4]University of Edinburgh, UK

Plan of the talk

- Motivations
- Status of data
- Setup proposed and counting rate estimations
- Request of protons

Motivations

Dual interest: Health and Environment

- **1. Boron Neutron Capture Therapy**
  Cl present in brain and in skin at 0.3%.
  Important for dose calculations in healthy tissue

- **2. Nuclear power plants**
  Cl present in the materials of the reactor
  Important for criticality calculations and waste repositories

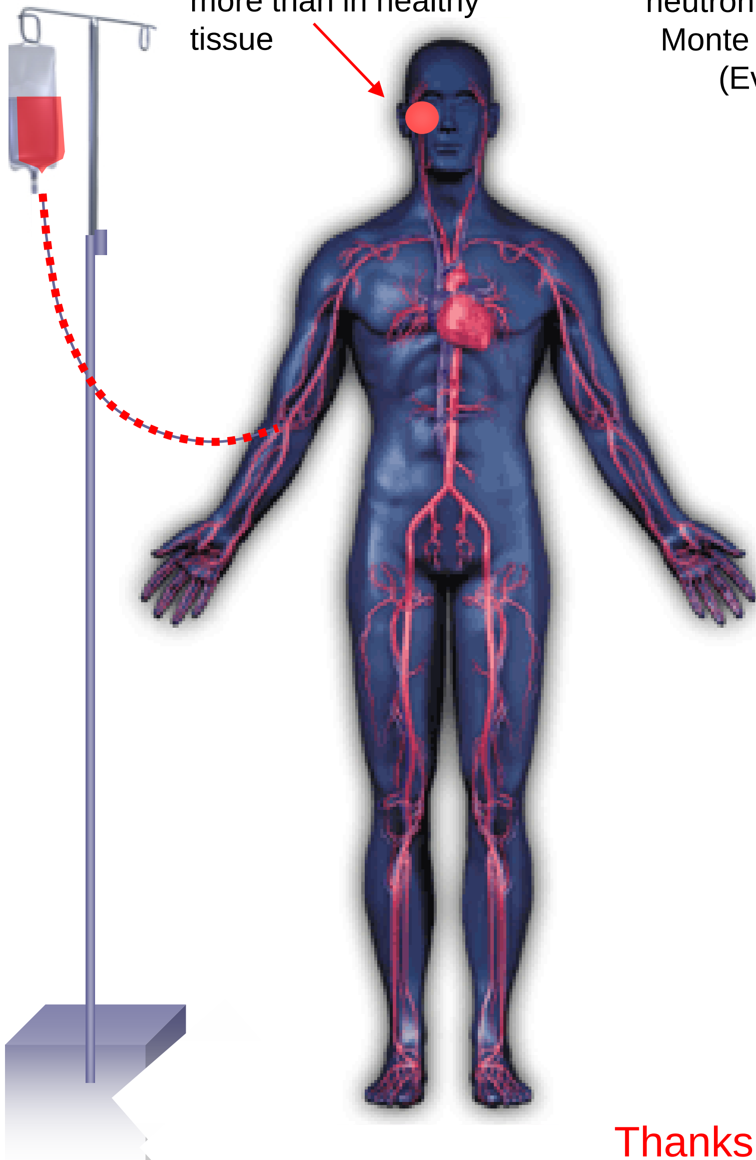
75% of Cl is ^{35}Cl, its capture cross section is so large (44 b. at thermal) that even at low concentrations its activation is important.

1. Boron Neutron Capture Therapy (BNCT)

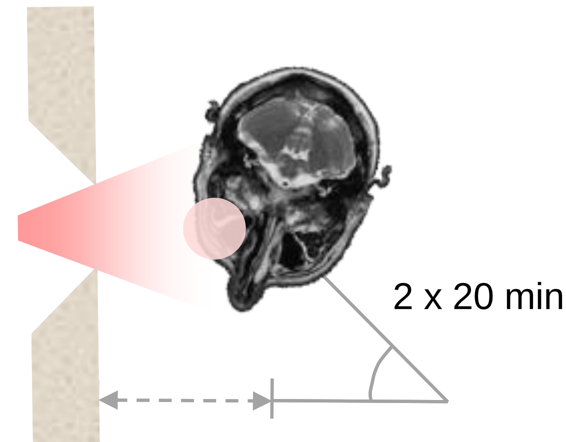
BNCT: an effective one-day treatment

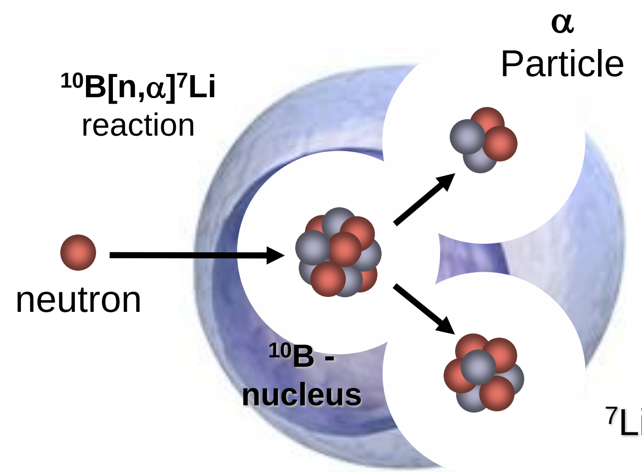
I.v. infusion of boron compound 4 hours before irradiation (BPA: 400 mg/kg)

Boron accumulates in malignant cells 4 times more than in healthy tissue



Irradiation with epithermal neutron beam according to Monte Carlo calculations (Evaluated data)

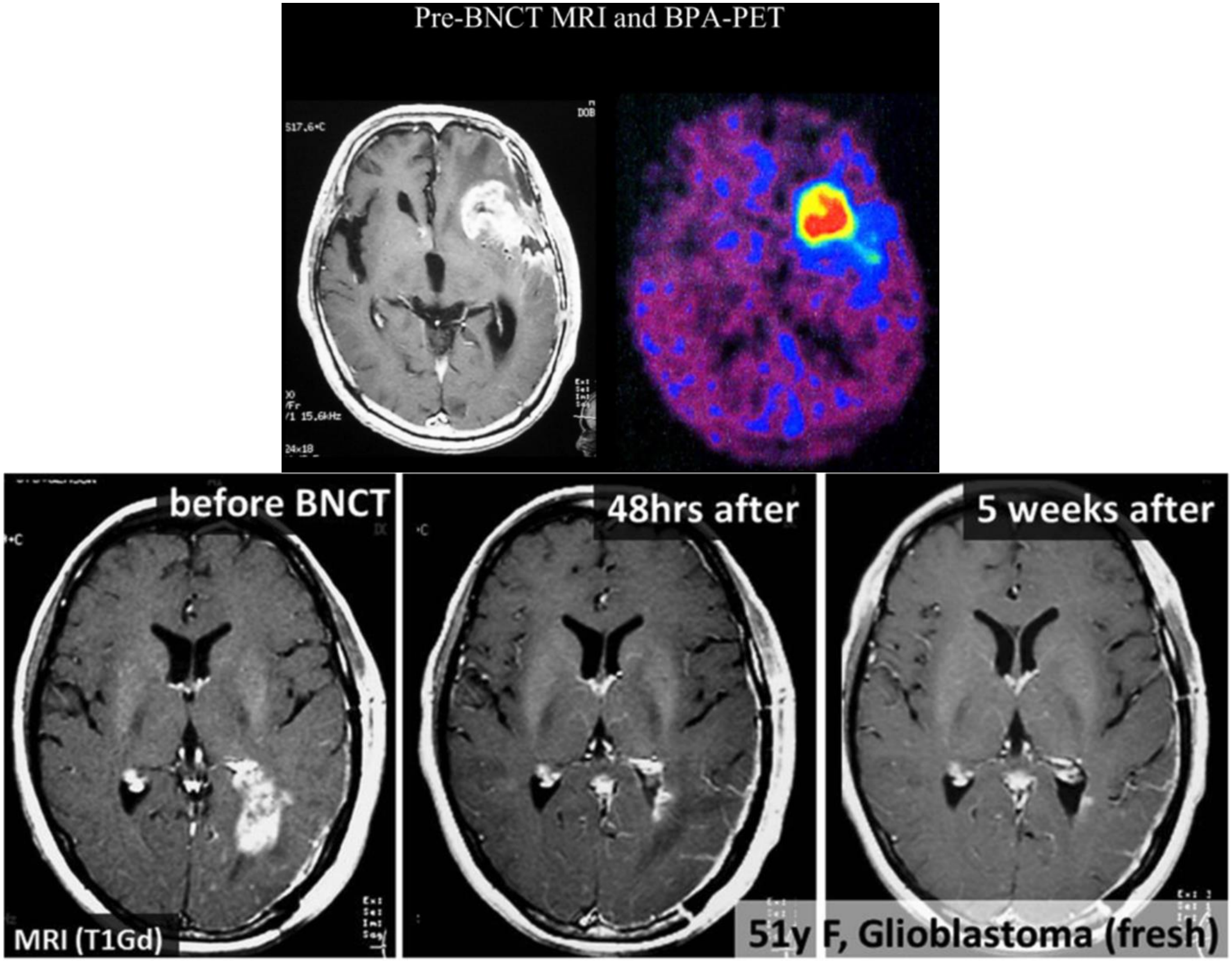




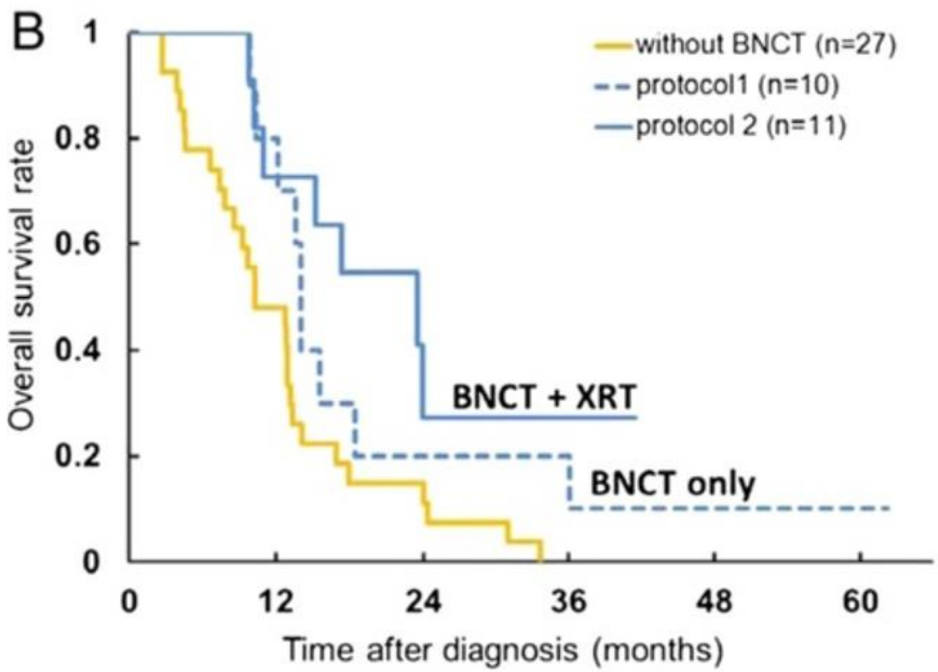
^{10}B capture neutrons and produces heavy ions which has a range (5-9 um) of the order of the cell size. The biological effectiveness is much greater than conventional RT.

Thanks to L. Kankaanranta & M. Kortesniemi

BNCT: unique RT option for infiltrative tumors



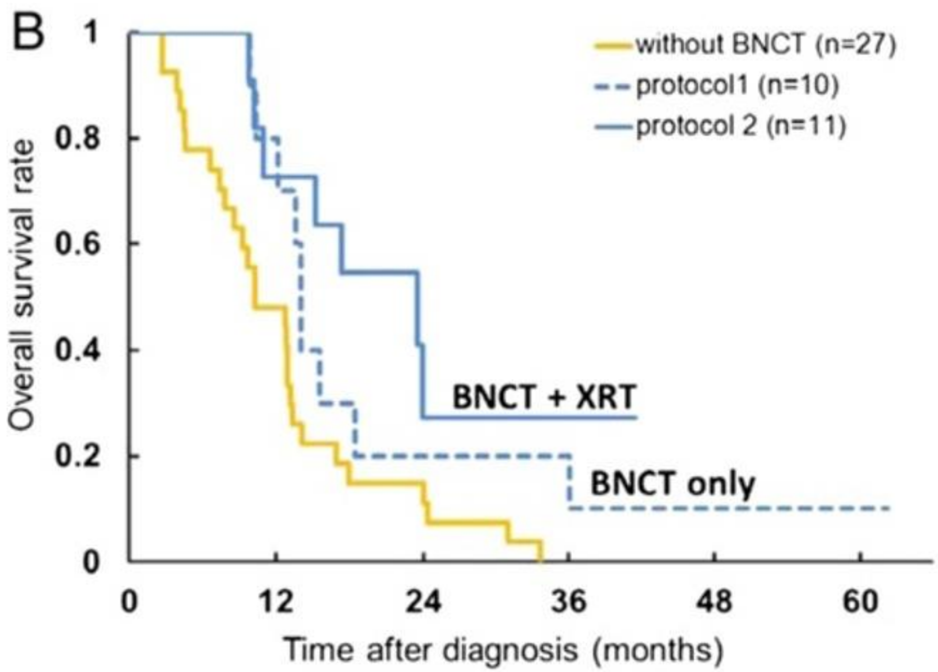
BNCT: promising therapy for brain tumors



Outcome of BNCT Osaka clinical trial for 21 brain tumor patients, combined with X-Ray Conventional Radiotherapy, and compared to standard treatments

S. Kawabata, S-I. Miyatake, T. Kuroiwa et al.,
J. Radiat. Res. 50, 51-60 (2009).

BNCT: promising therapy for brain tumors



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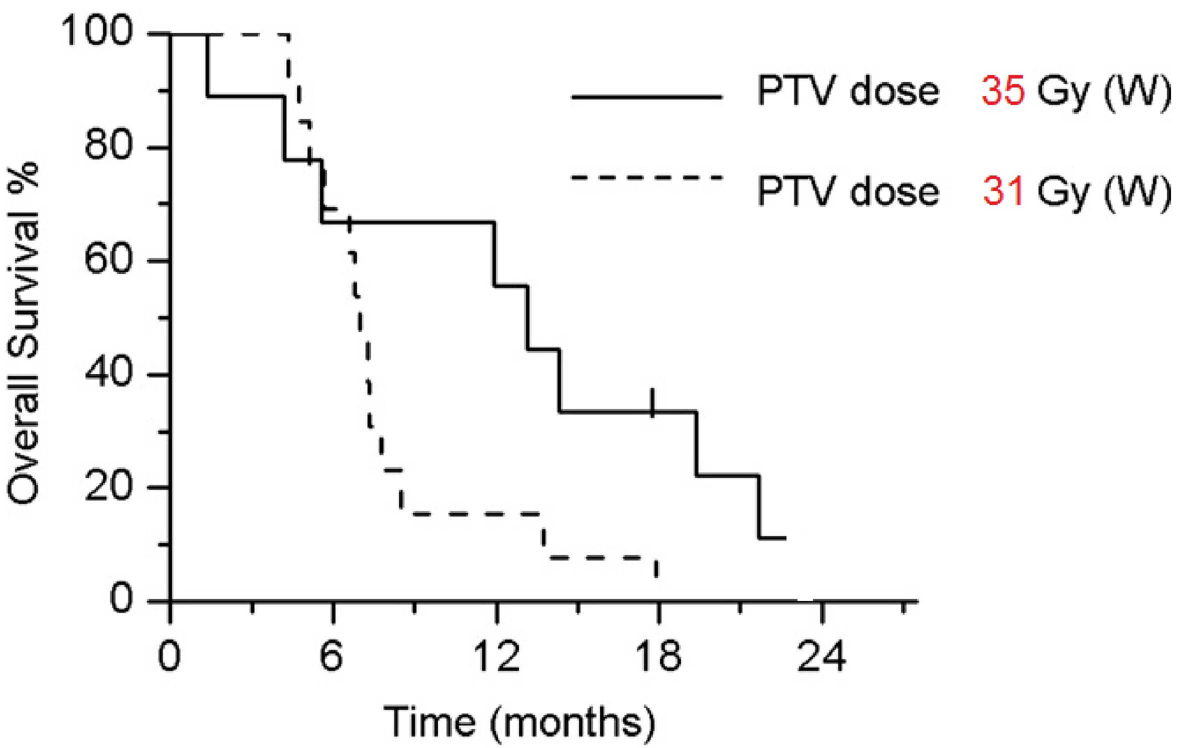
Can we optimize the treatment further?

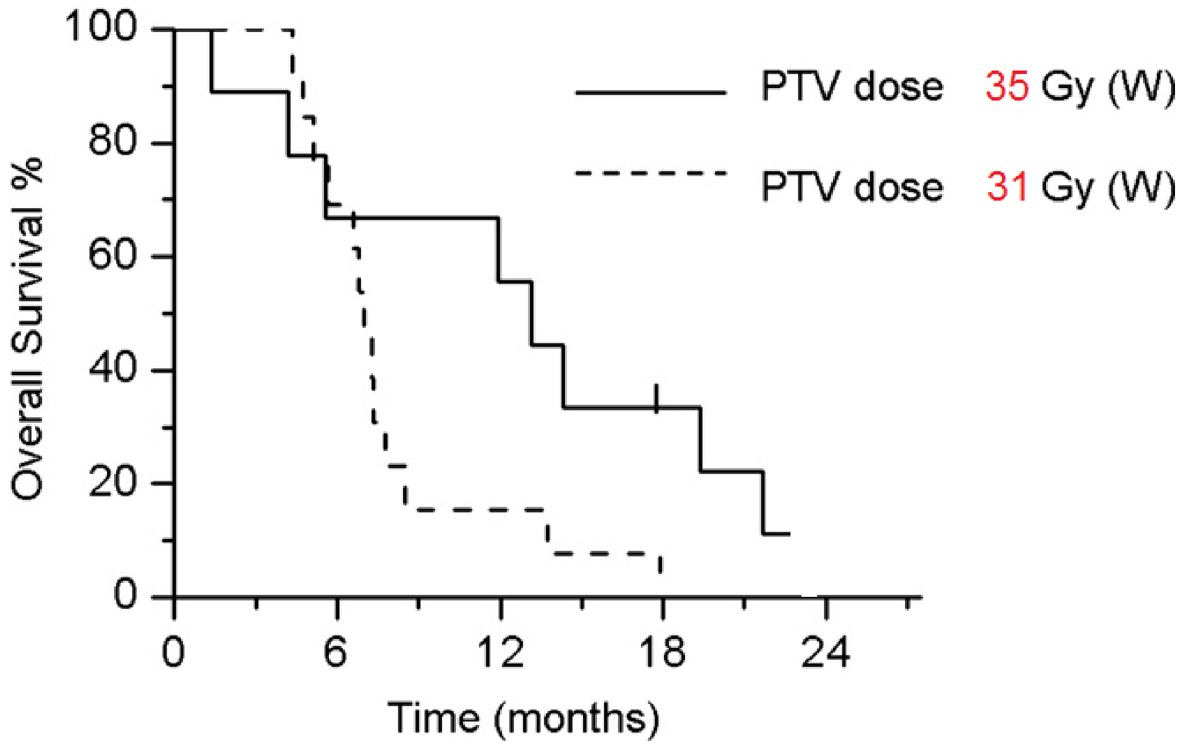
Adjusting dose may improve outcome



Outcome of BNCT Finnish trial for 22 brain tumor patients (Glioblastoma) depending on dose delivered on Planning Tumor Volume

L. Kankaanranta, T. Seppala, H. Koivunoro et al., **Int. J. Radiat. Oncol. Biol. Phys. 80, 369-376 (2011).**

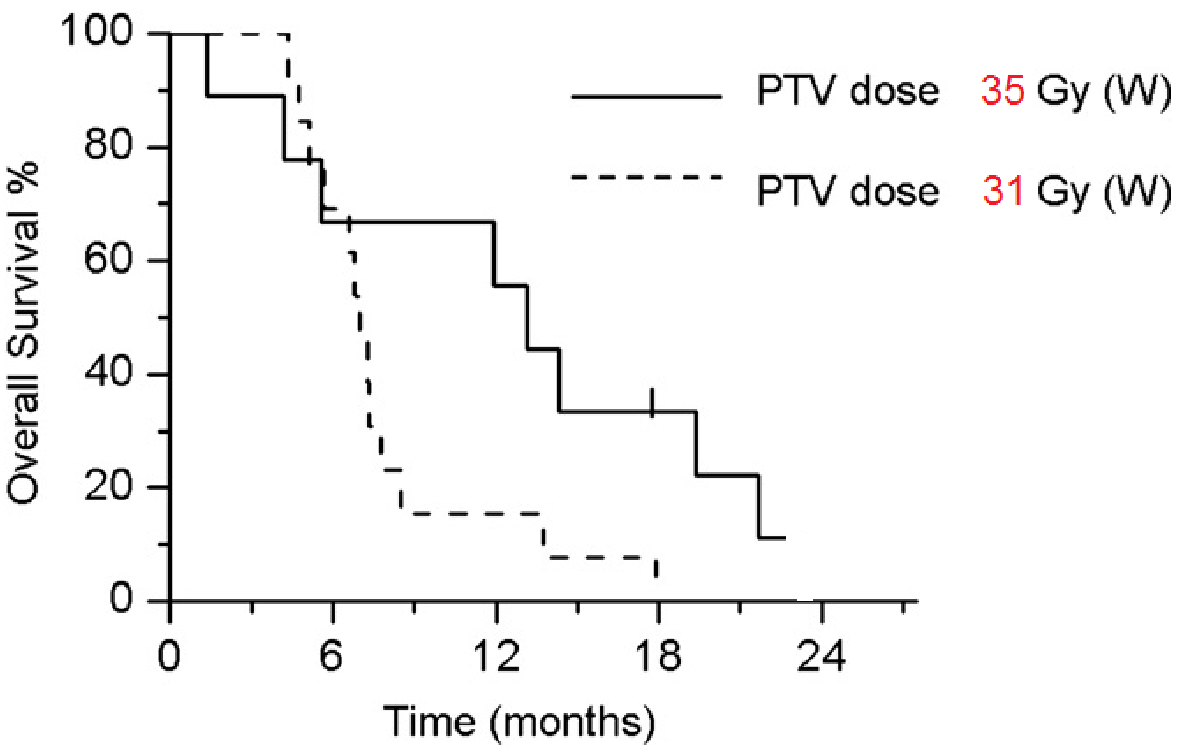
Adjusting dose may improve outcome



But …

Dose PTV (Gy)	Dose healthy tissue (Gy)
35 →	12.1

Adjusting dose may improve outcome

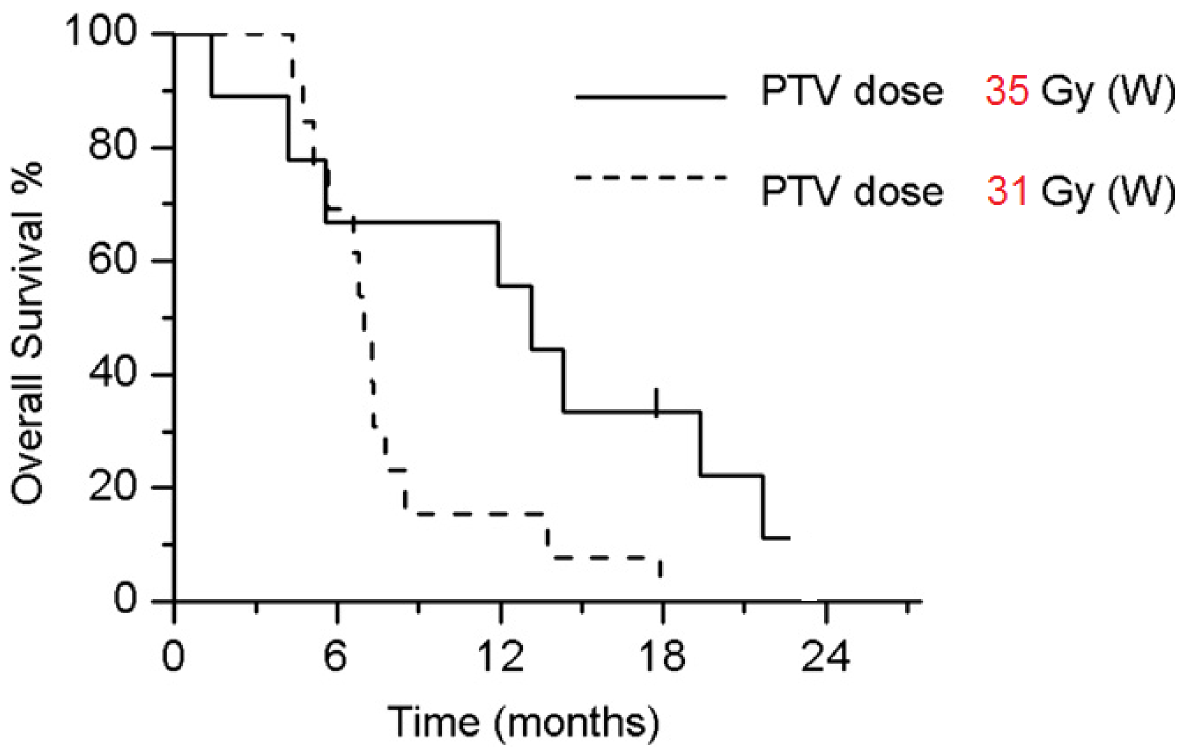


But …

Dose PTV (Gy)	Dose healthy tissue (Gy)
35 →	12.1
	16 (max tolerable)

large safe margin, because of uncertainties

Adjusting dose may improve outcome



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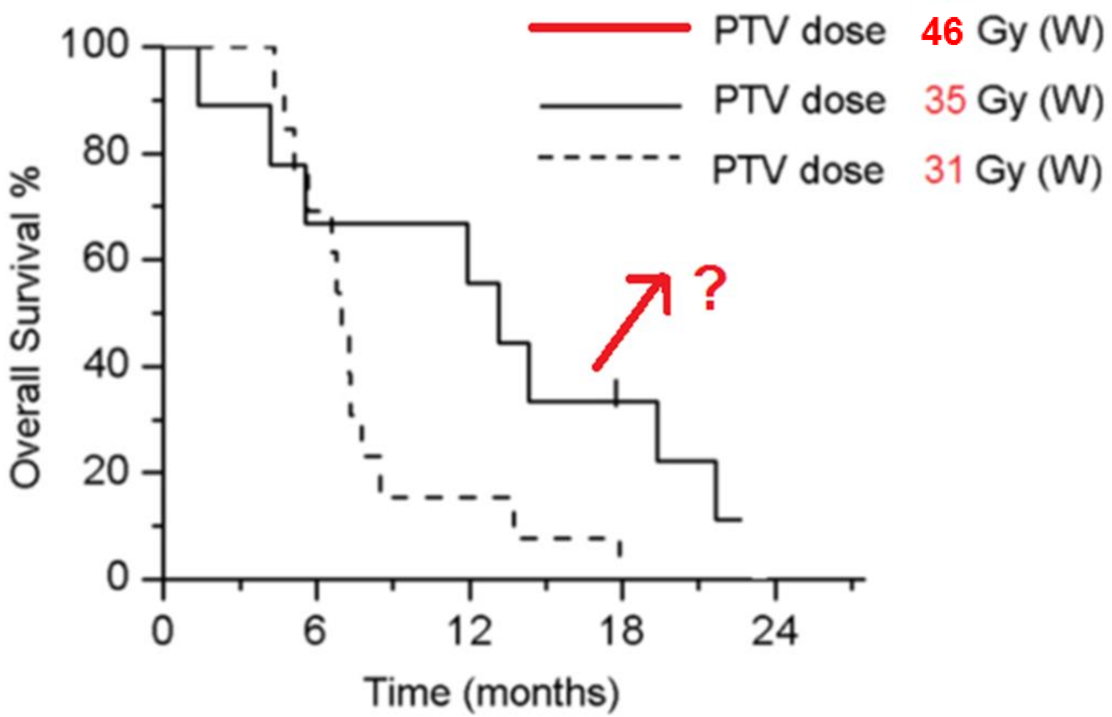

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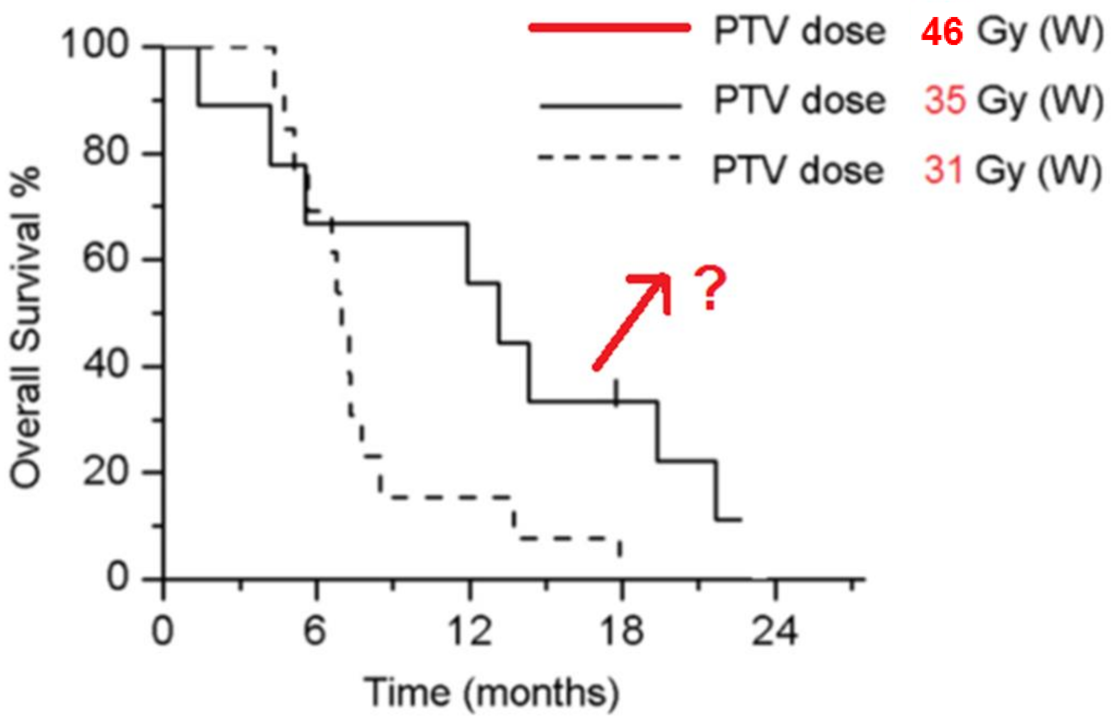


A more precise dose calculation (includes improving all cross sections) could lead to optimize treatment time

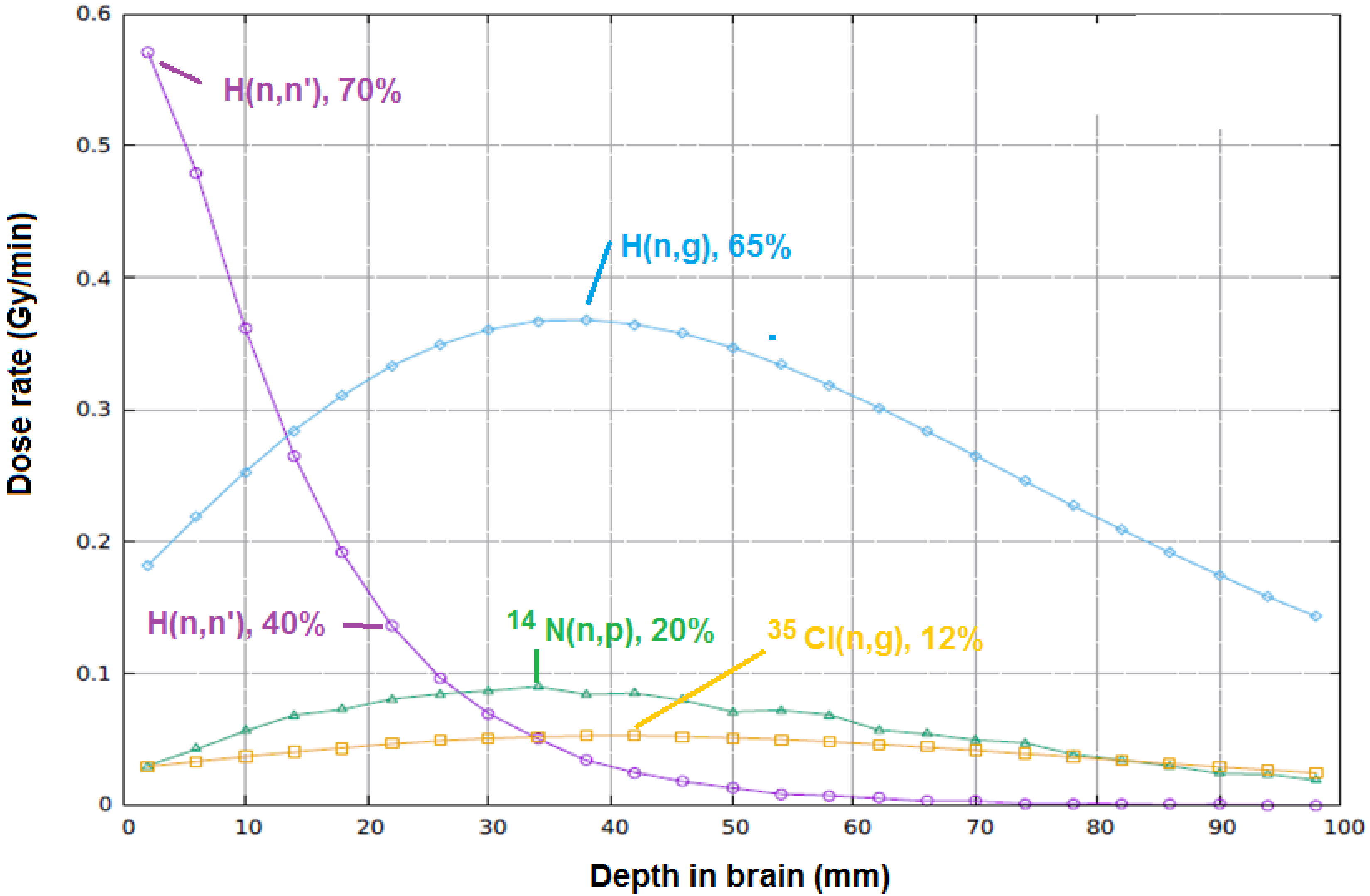
⇒ **Better therapeutic outcome and possibly survival**

But …

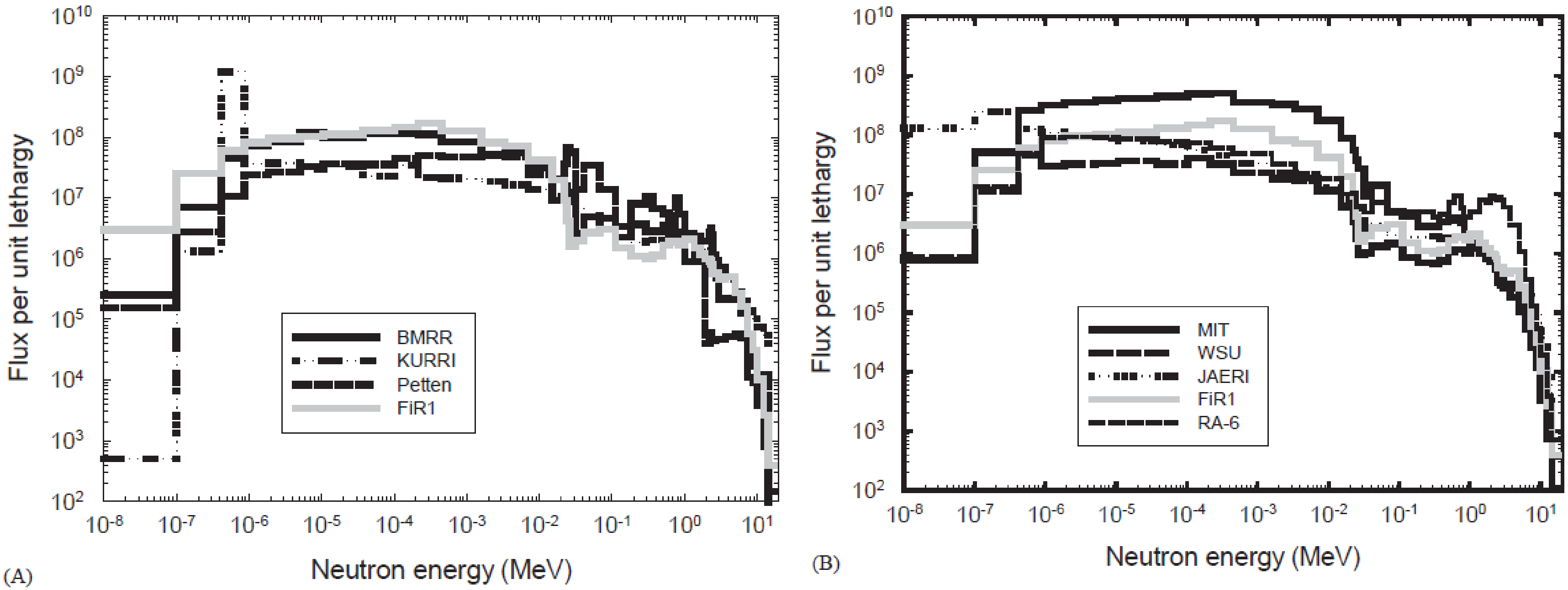
Dose PTV (Gy)	Dose healthy tissue (Gy)
35 →	12.1
46 ←	16 (max tolerable)

large safe margin, because of uncertainties

Most important processes contributing to dose in brain (healthy tissue)



Energy range of interest: thermal to 100 keV



(A) (B)

Spectrum of the main neutron beams for BNCT

I. Auterinen, T. Serén, K. Anttila et al.,
Applied Radiation and Isotopes 61 (2004) 1021–1026

2. Nuclear fission reactors

Chlorine in fission reactors

- ^{35}**Cl is present in:**
  - Polyvinyl chloride pipes (is 57% Cl by weight)

  R. O. Sayer, K. H. Guber, L. C. Leal, Phys. Rev. C73, 044603 (2006)
  - Chlorides are impurities in fuel cladding.

  IAEA-TECDOC-927: Influence of water chemistry on fuel cladding behaviour
  - Cl in nuclear grade graphites (purification processes).

  LLWR Environmental Safety Case: Cl-36 Mobility in Reactor Circuits and its Potential Significance for the ESC 2011 Inventory, LLWRP 1106: SERCO/E003756/008

- ^{36}**Cl is produced in fission reactors by** ^{35}**Cl(*n,γ*) capture**

Motivations

RESEARCH PROFILES

Detection of ^{36}Cl in nuclear reactor waste

When most people think of radioactive waste, elements such as uranium, plutonium, or even americium come to mind. Very few would immediately name chlorine as a problematic element for radioactive waste disposal. But one of chlorine's radioactive isotopes, ^{36}Cl, has a particularly long half-life (hundreds of thousands of years) and is extremely water-soluble, making it a potential long-term threat to the environment if it is not disposed of properly. For ^{36}Cl content to be determined accurately, it must first be separated from its matrix and from other radionuclides, which can be a difficult task if the ^{36}Cl is embedded in the components of a nuclear reactor. In the April 15 issue of *Analytical Chemistry* (pp 3126–3134), Xiaolin Hou and colleagues at the Technical University of Denmark present a new set of meth-

normal waste, it will be quite cheap," says Hou.

Of the materials tested in this paper, only graphite has a previously reported method for extraction of ^{36}Cl. In that case, the chloride was leached from the alkali fusion at high temperatures with NaOH and Na_2CO_3 to a fused cake, which is then leached with water to extract the chloride.

For all the materials, the solutions containing the chloride are precipitated as AgCl. At this point, the scientists faced another challenge. Hou explains, "In most papers, they do the measurement of the ^{36}Cl by just dissolving the silver chloride in a very high concentration of ammonium solution and then doing the measurements, but . . . if the chloride content is high, you need quite a lot of ammonium to dissolve it." In addition, the dissolved silver is not very stable over the time period necessary for the liquid scintillation counting process and tends to precipitate. "If there are particles formed in the solution, it will change the counting efficiency so you cannot get a very good result," says Hou.

JUPITERIMAGES

When this nuclear reactor is decommissioned, Hou and colleagues' methods could help authorities safely dispose of the construction materials.

Cl is a major concern in radioactive waste due to:

- Half-life of 301000 y.
- High mobility (extremely water soluble)

Motivations

Nuclear Science

ISBN 978-92-64-99072-2

Mobile Fission and Activation Products in Nuclear Waste Disposal

Workshop Proceedings
La Baule, France
16-19 January 2007

OECD 2009
NEA No. 6310

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

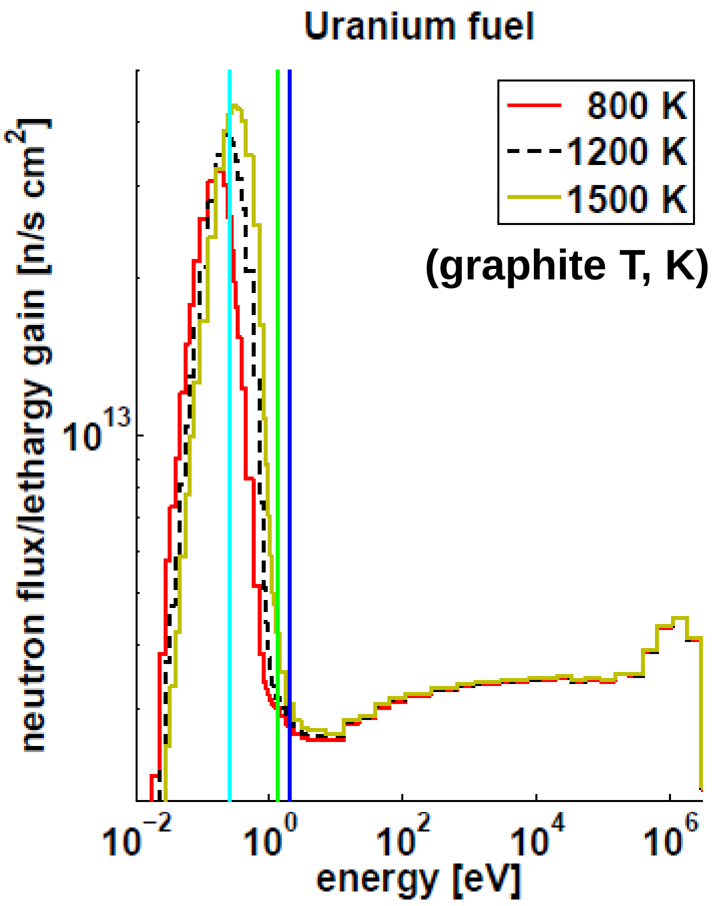
- Cl-36 inventories in low and intermediate level waste stores are of interest to ANDRA because Cl-36 contributes 77% (5.7E-3 mSv/y) of the peak dose at Centre de l'Aube.

- The JGC Corporation in Japan are concerned with Cl-36 issues in Low and Intermediate Level Waste because Cl-36 is assessed to be the major contributor to dose following disposal of this waste, assumed to arise due to use of river water for irrigation.

In order to accurately predict the amount of ^{36}Cl in irradiated fuel, the ^{35}Cl(n,γ) cross section must be known to a high level of accuracy

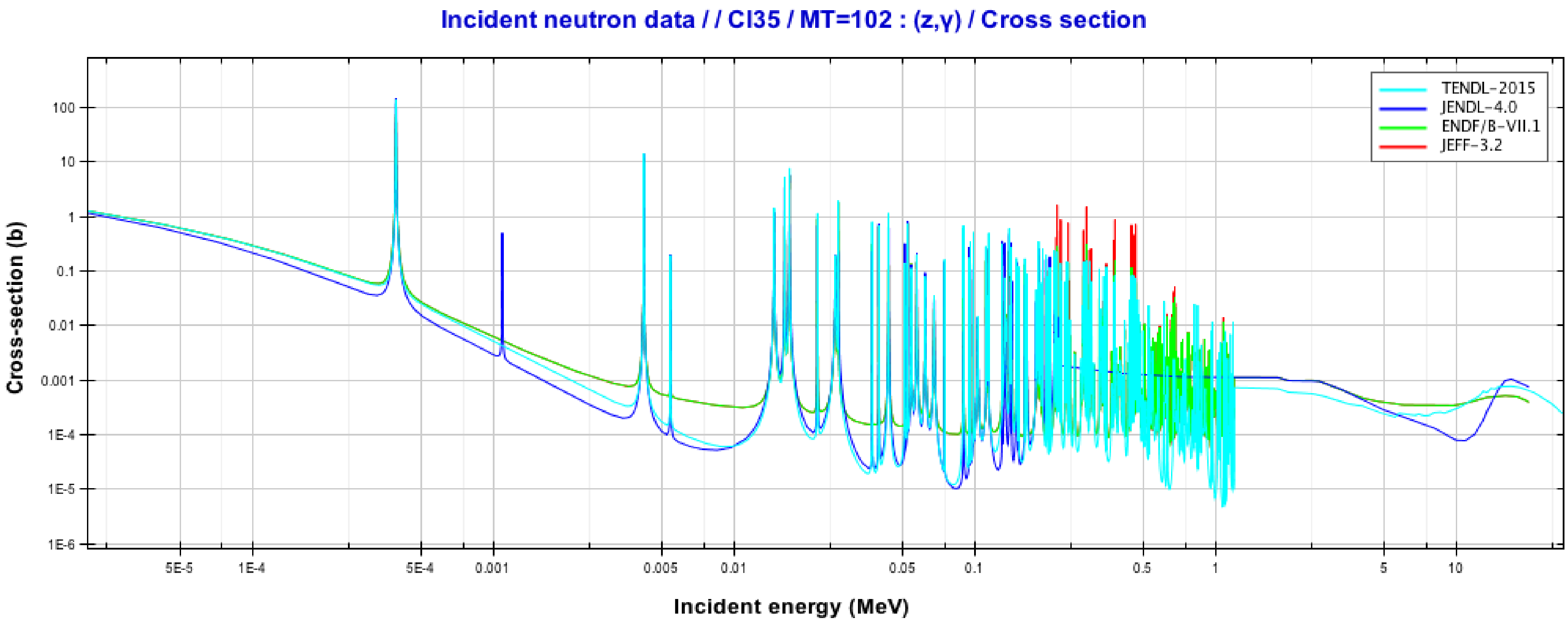
Energy range of interest

thermal to
~ 1 MeV



Status of data

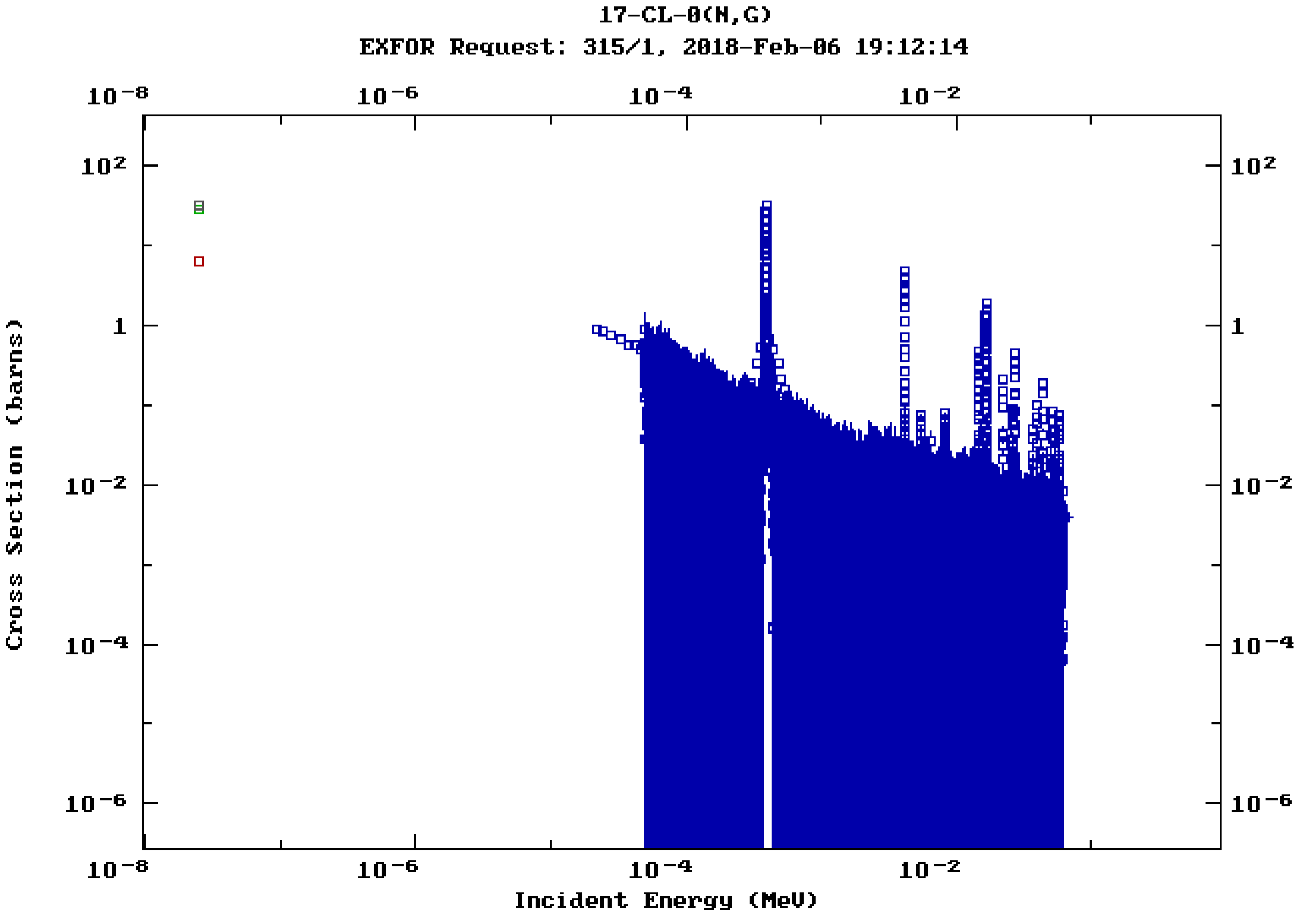
Evaluations



Uncertainties of 10% between evaluations:

too much for precise estimation of ^{36}Cl production inside the reactor core.

EXFOR

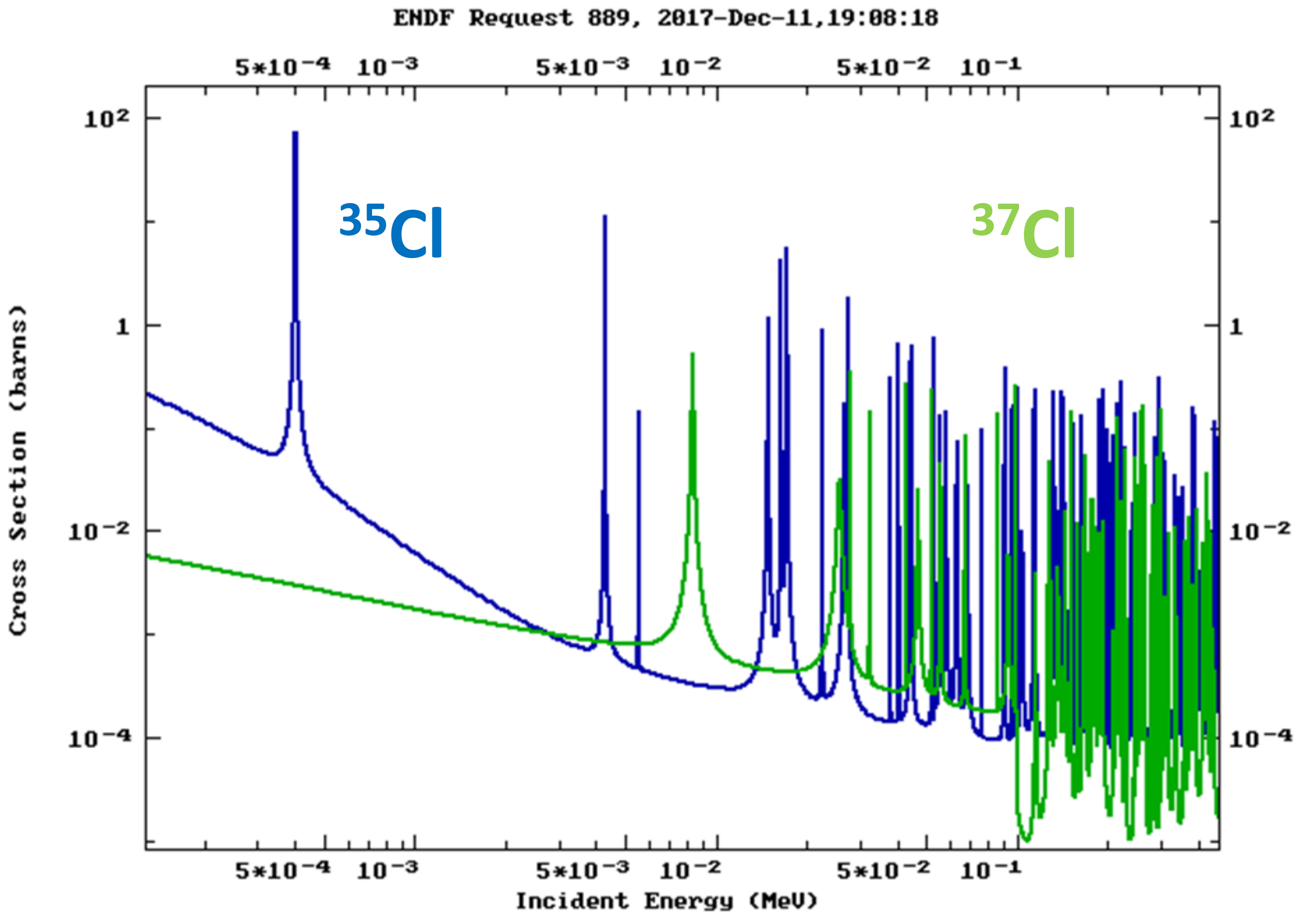


Only one capture measurement in resonances:

K. H. Guber, R. O. Sayer, T. E. Valentine, et al, *Phys. Rev. C* 65, 058801 (2002)

Discrepances in the measurement at thermal point

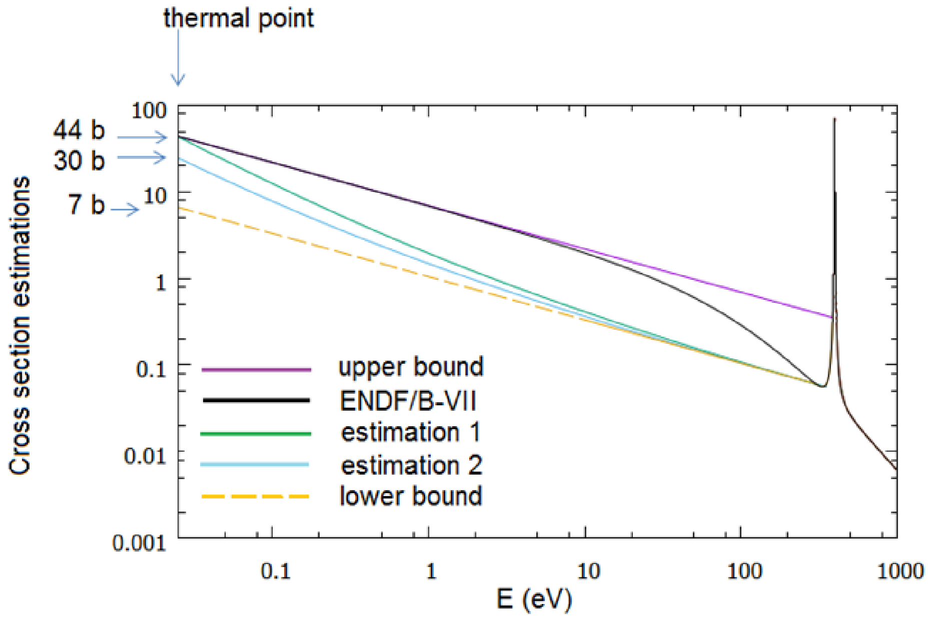
All measurements with nat Cl (75% ^{35}Cl, 25% ^{37}Cl)



We propose the first measurement with pure 35Cl

Low E behaviour: bound state

ENDF: Use bound state to fit the thermal value of Mughabghab.
-75 eV to -180 eV reported from different authors, different possibilities for low energy behaviour



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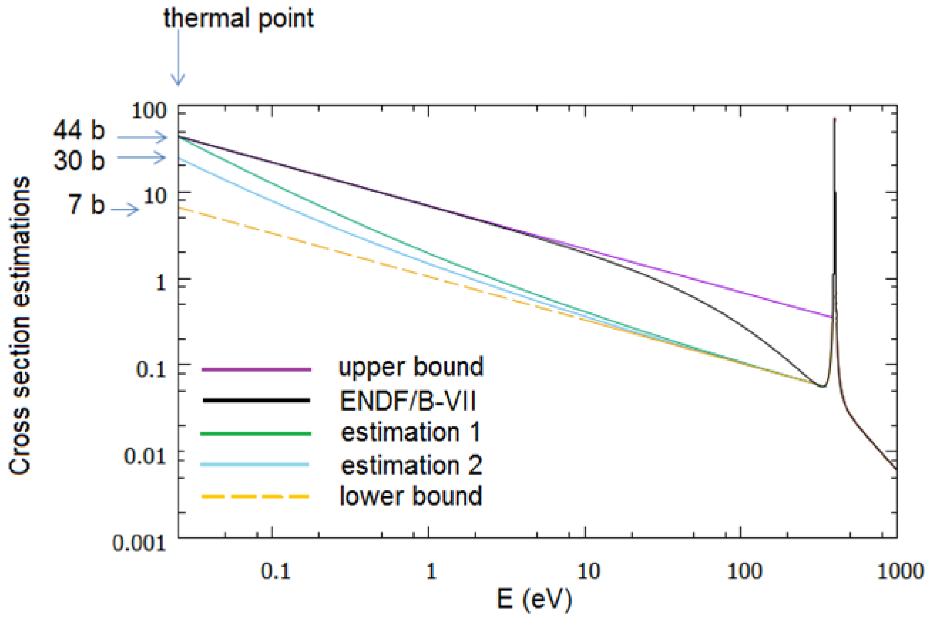

Has this any impact on BNCT dose delivered to brain?

MC Simulations for Snyder head model with different cross section

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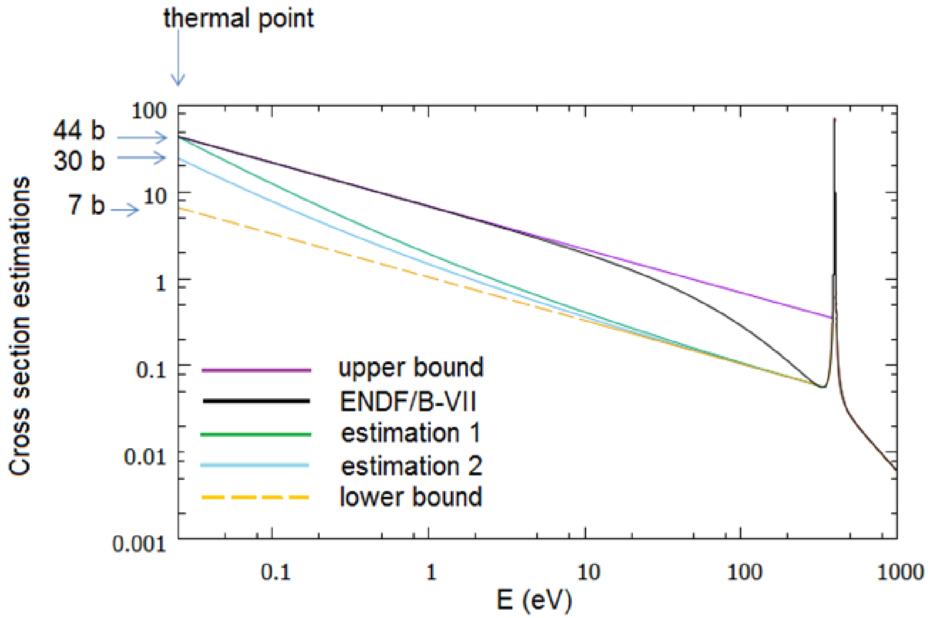

	Dose 35Cl (n,γ)	%Dose 35Cl(n,γ)	Total Dose
Upper bound	0.516	**12.446**	4.1830
ENDF/B-VII	0.514	**12.410**	4.1439
Estimation 1	0.551	**13.196**	4.1421
Estimation 2	0.325	**8.167**	3.9803
Lower bound	0.081	**2.162**	3.7660

Low E behaviour: bound state

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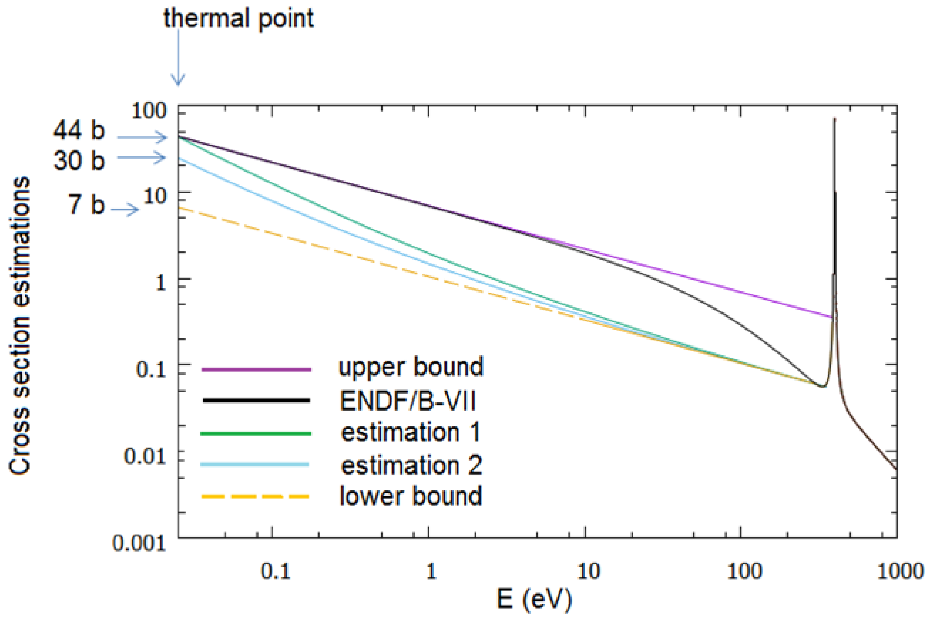

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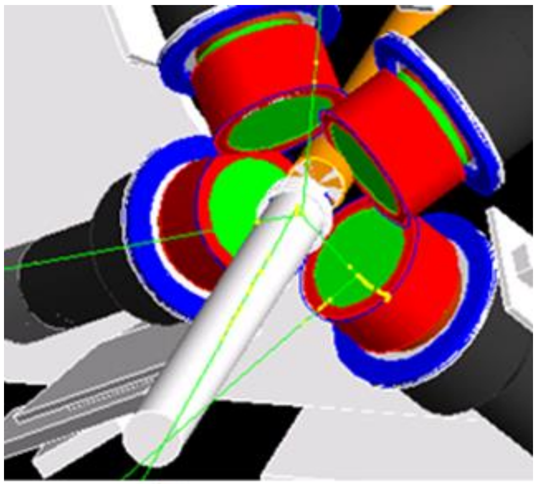
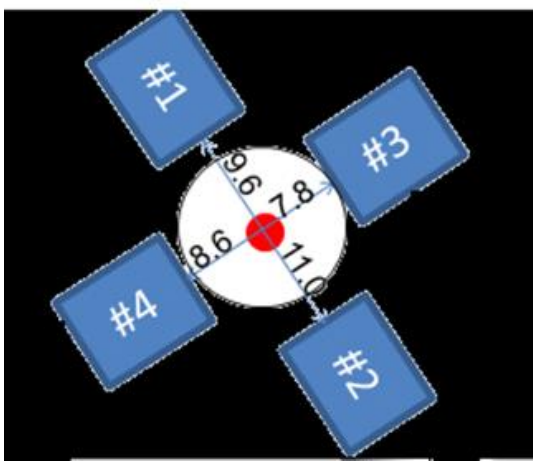
MC Simulations for Snyder head model with different cross section

ICRU: Doses must be known with less than 5% uncertainty

Set up and counting rate

Setup





Capture Setup in EAR-1

- 4 x C6D6 detectors, (neutron insensitive)

Commonly used in capture measurements at n_TOF

Samples of $Na^{35}Cl$ or $K^{35}Cl$

Nominal mass	Accurate mass	% Natural abundance	Chemical form	Enrichment available %
^{35}Cl	34.968852721 (69)	75.78 (4)	KCl, NaCl, $BaCl_2$, AgCl	83 - 99+
^{37}Cl	36.96590262 (11)	24.22 (4)	KCl, AgCl, $BaCl_2$, $PbCl_2$	42 - 86+

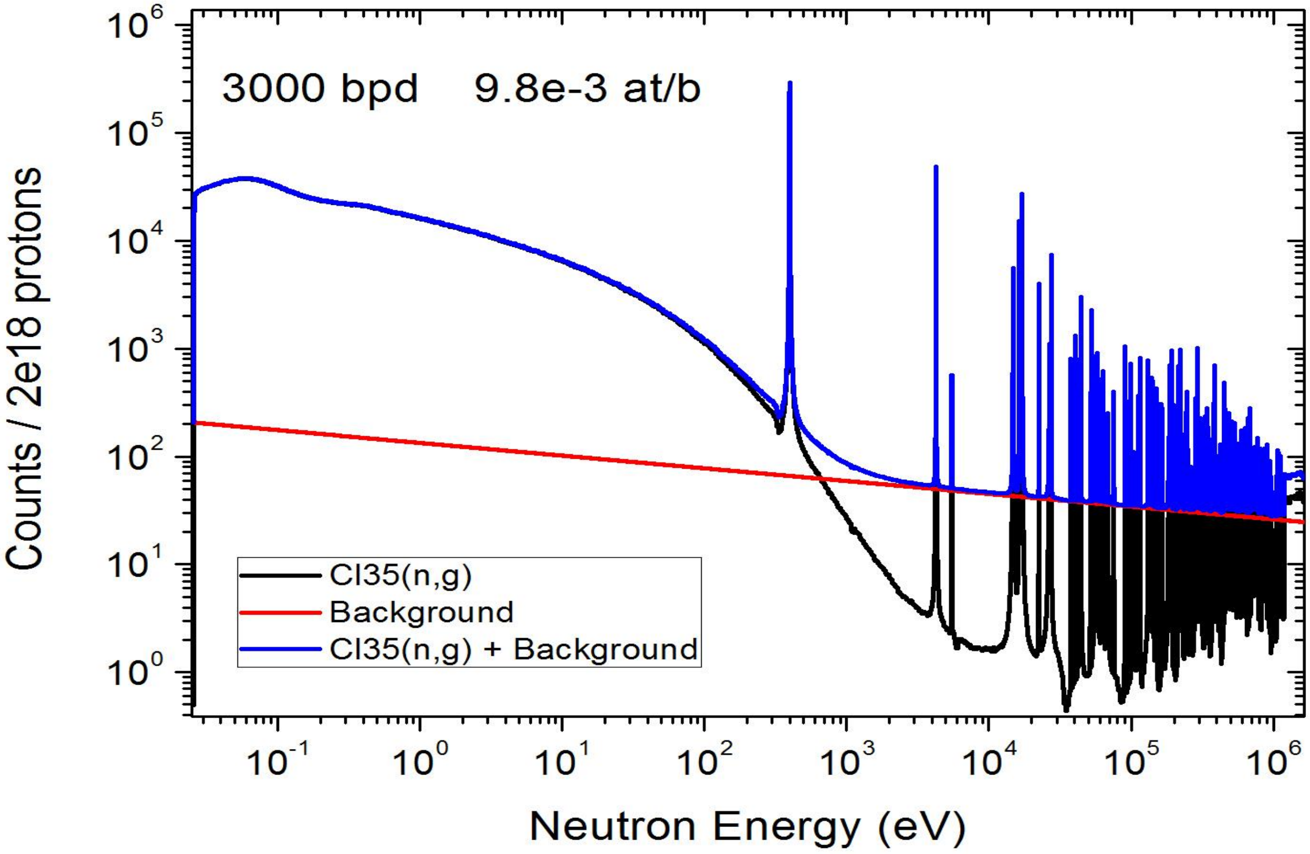
Chlorine Isotopes Cl-35 and Cl-37 are used to study the toxicity of environmental pollutant and are usually supplied in the form of NaCl.

Already produced:

500 mg
3.2e-3 at/b

250 mg
1.6e-3 at/b

Counting rate estimation



Protons requested

1.5×10^{18} protons for 35Cl measurement
0.5×10^{18} protons for normalization and background

Total 2×10^{18} protons

2 PhD students involved

- F. Ogállar (University of Granada, Spain)
  Low energy values and application to dose calculations (together with 35Cl(n,p) recently performed) for BNCT of brain tumors

- S. Bennett (University of Manchester, UK)
  Resonance analysis and application to calculations of 36Cl activity

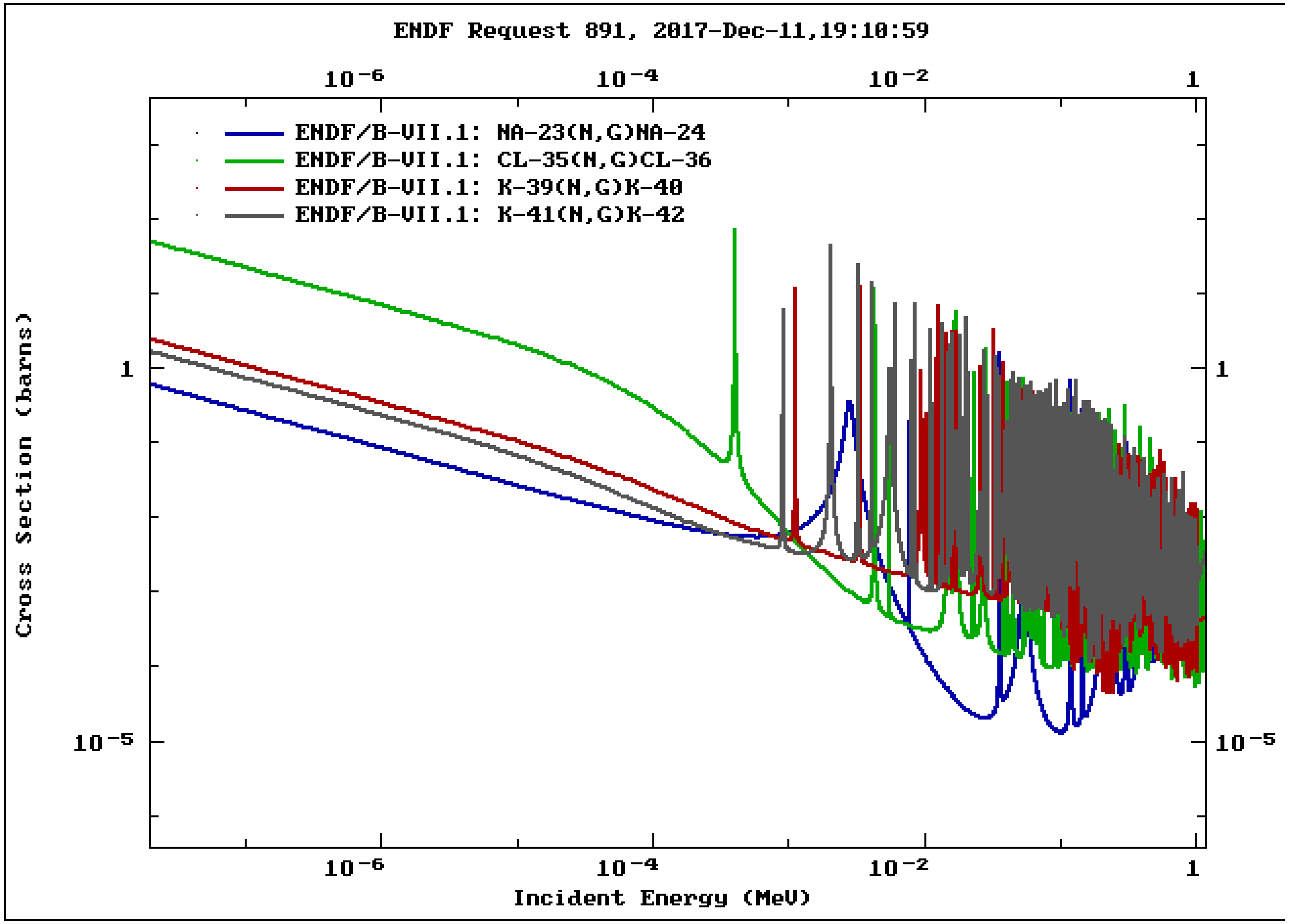
Close coordination between groups for set-up and analysis

Conclusions

- **Low E data (missing data) are important for BNCT**
- **Data in the resonance region (uncertain, only one measurement wilth nat Cl) important for radioactive waste calculation**
- **Feasible experiment: samples and detector available and count rate above bacground guaranteed**
- **Two PhD students from different universities benefiting from the analysis and applications**

THANK YOU FOR YOUR ATTENTION

Sample of $Na^{35}Cl$ or $K^{35}Cl$

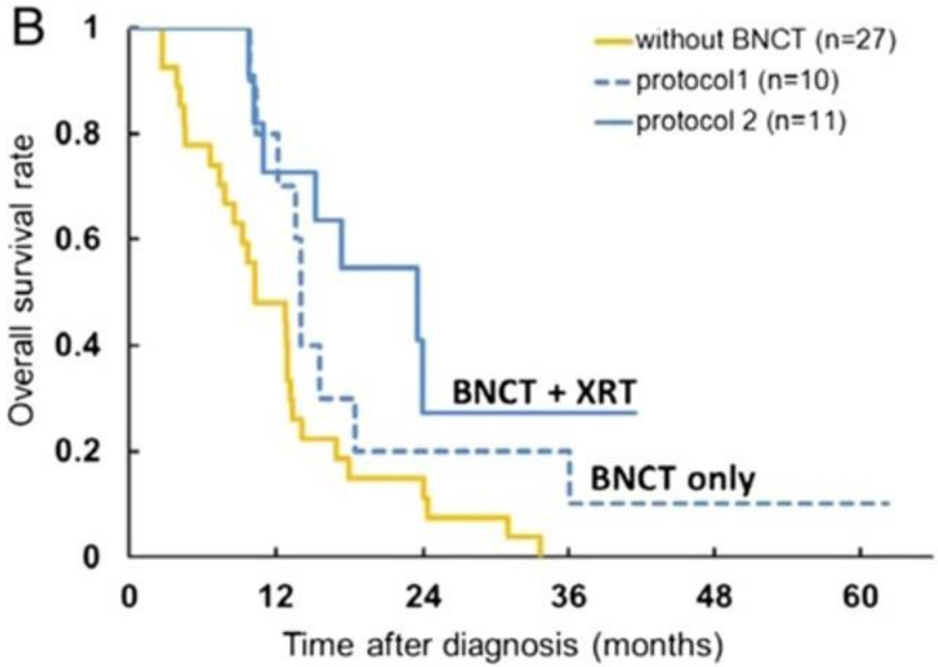


Already produced:

500 mg 2 mm

250 mg 1 mm

BNCT: promising therapy for brain tumors



Outcome of BNCT Osaka clinical trial for 21 brain tumor patients, combined with X-Ray Conventional Radiotherapy, and compared to standard treatments

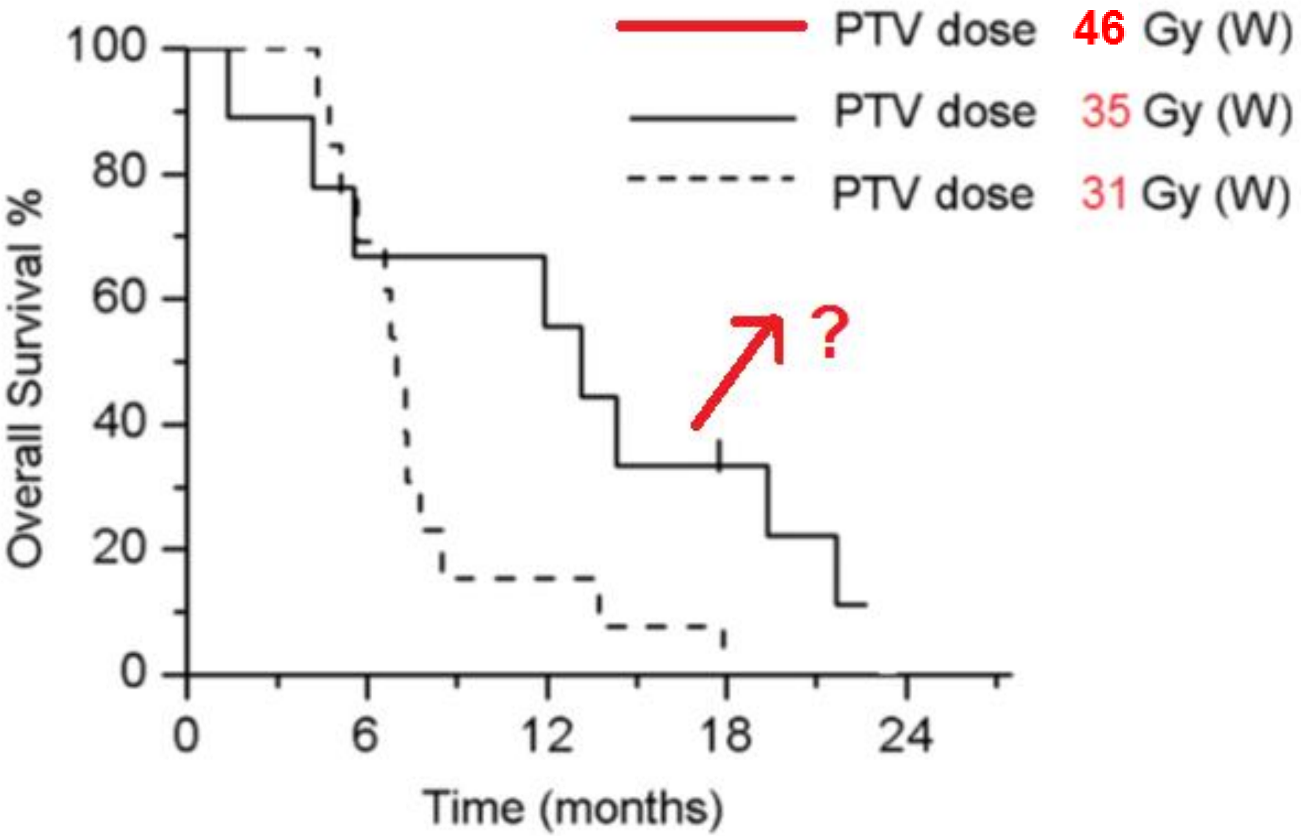
S. Kawabata, S-I. Miyatake, T. Kuroiwa et al.,
J. Radiat. Res. (Tokyo) 50, 51-60 (2009).

BNCT Treatment Planning – Example of protocol

- Finnish clinical trial – mean values
- Dose not accurately known - BNCT is planned with a safe margin

	Brain tumor (PTV)	Healthy tissue (limiting factor)
Fractionated Dose (Gy)	60 (30 sessions of 2 Gy)	60 (30 sessions of 2 Gy)
Current dose of the BNCT treatment (Gy-Eq) – single session	35	12.1
Current photon equivalent dose of the BNCT treatment (Gy-Eq) – fractionated	50.5 (25 sessions of 2 Gy)	41.5 (21 sessions of 2 Gy)
Target photon equivalent Dose BNCT treatment (Gy-Eq)	71.0 (35 sessions of 2 Gy)	60 (30 sessions of 2 Gy)
Target BNCT dose (Gy-Eq)	46.0	16.0

Adjusting dose may improve outcome



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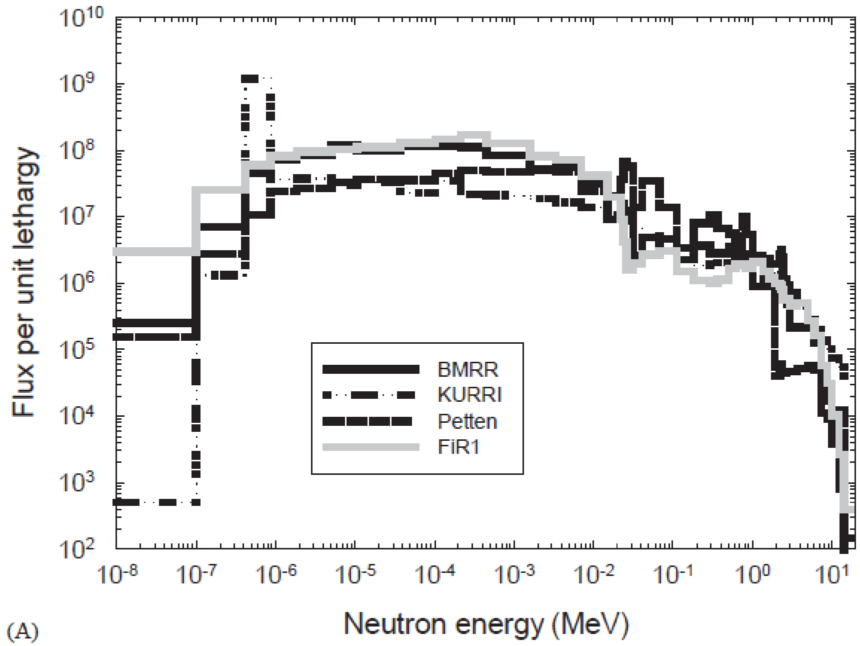
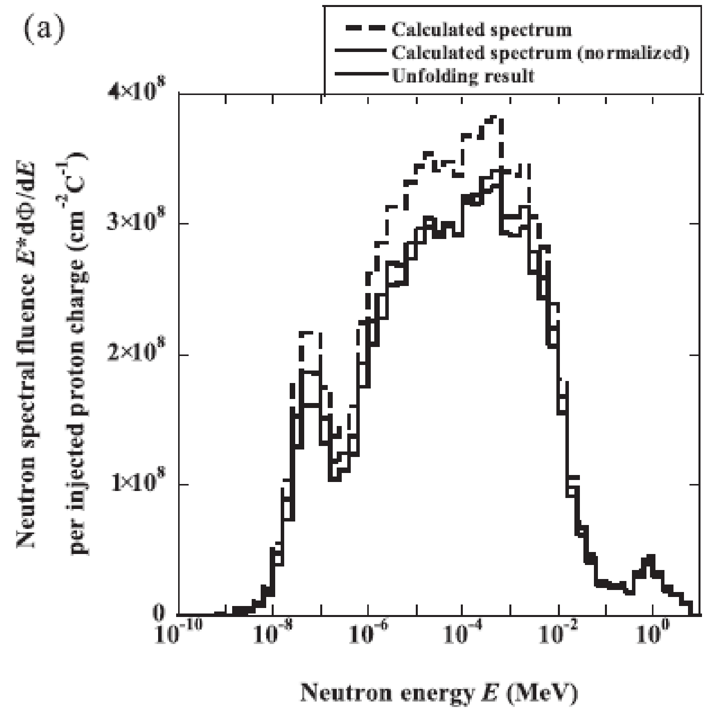




Spectrum of the main neutron beams for BNCT compared to the accelerator one of Ibaraki based on ^{9}Be(p,n) reaction

I. Auterinen, T. Serén, K. Anttila et al.,

Int. J. Applied Radiation and Isotopes 61 (2004) 1021–1026

ENDF/B-VII.1

ENDF/B-VII.1:

PHYSICAL REVIEW C 73, 044603 (2006)
R-matrix analysis of Cl neutron cross sections up to 1.2 MeV
R. O. Sayer, K. H. Guber, L. C. Leal, et al.

> For resonances above 500 keV, the capture widths were set to the average capture width of the resonances observed in the capture measurements. Average ^{35}Cl capture widths were 606 MeV for *s* waves and 860 MeV for *p* waves

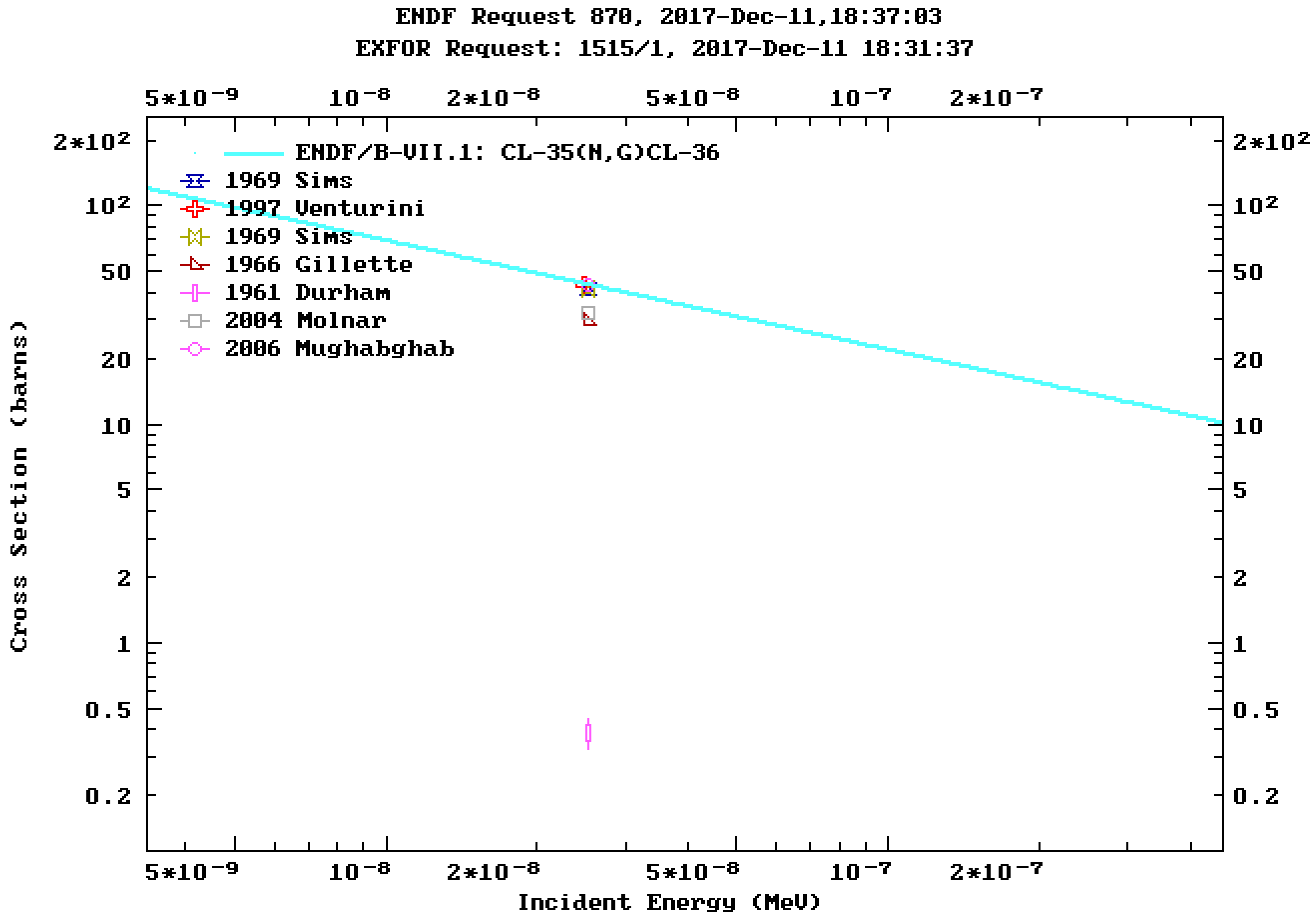
Only one capture measurement in resonances:

PHYSICAL REVIEW C 65, 058801 (2002)
New Maxwellian averaged neutron capture cross sections for ^{35,37}Cl
K. H. Guber, R. O. Sayer, T. E. Valentine, et al

Capture measurements performed only for ^{nat}Cl

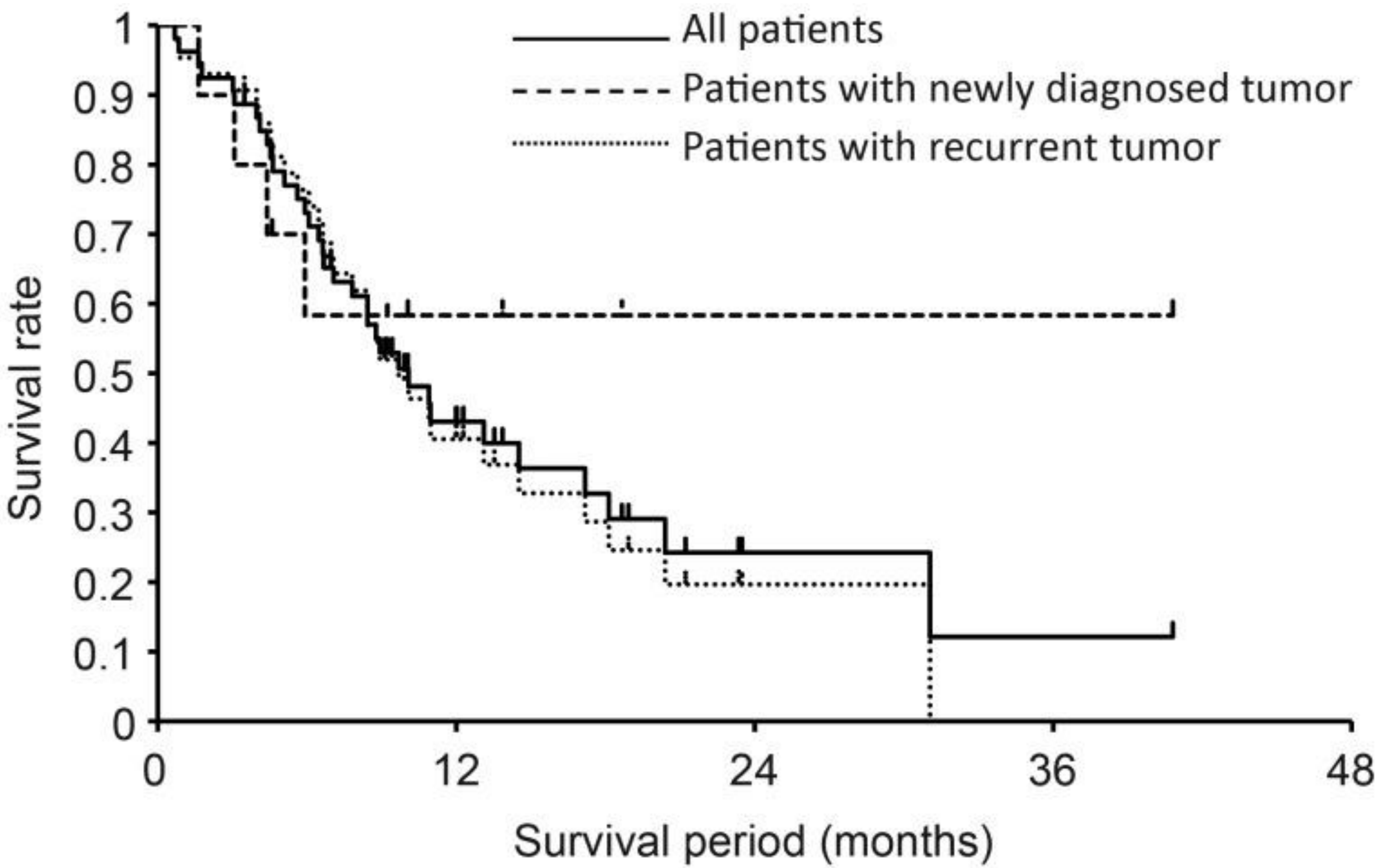
(75% ^{35}Cl, 25% ^{35}Cl)

Thermal value



BNCT for head and neck tumors

Suzuki M, Kato I, Aihara T, et al. Head & Neck tumors:
J Radiat Res. 2014 Jan; 55(1): 146–153.
doi: 10.1093/jrr/rrt098

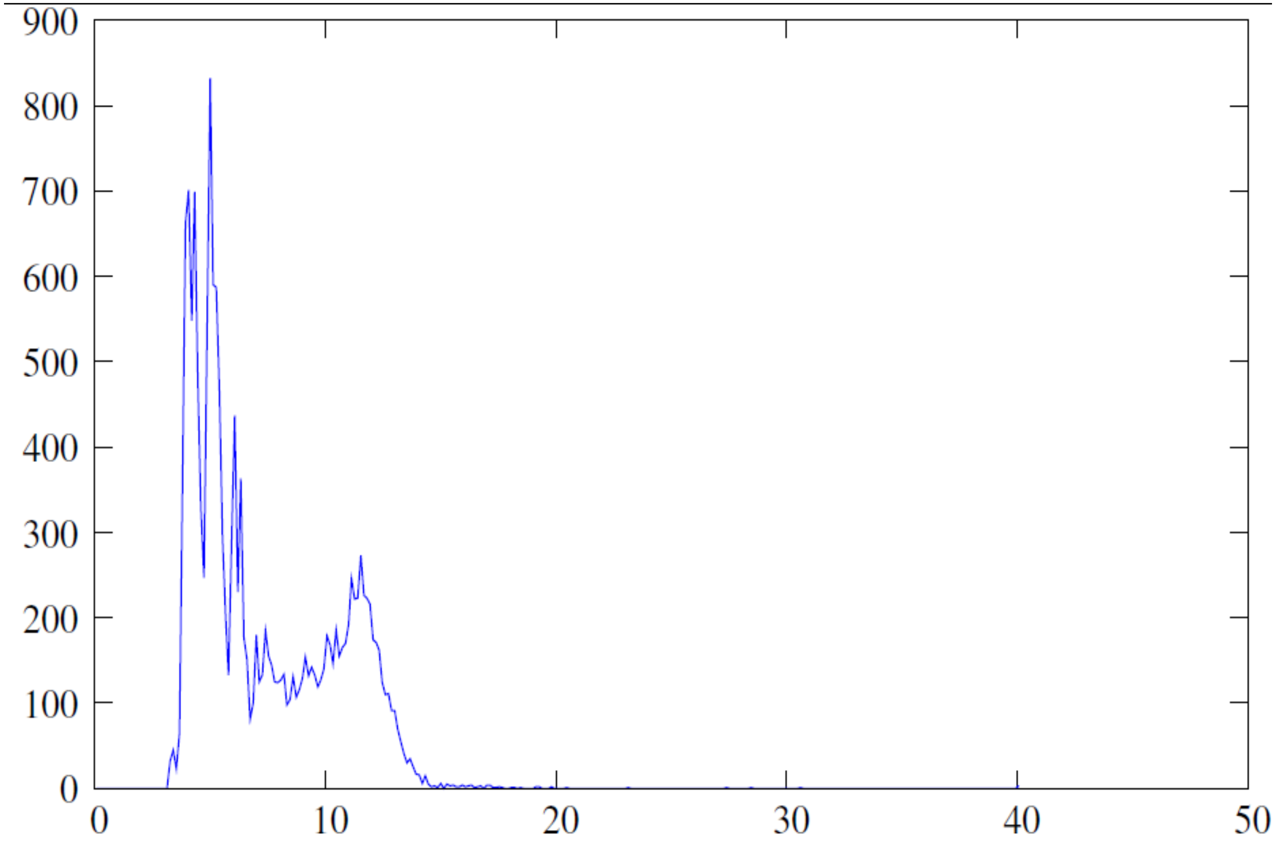


Detector response:

88Y source: γ’s of 898.0 (93.7%) and 1836 keV (99.2%)

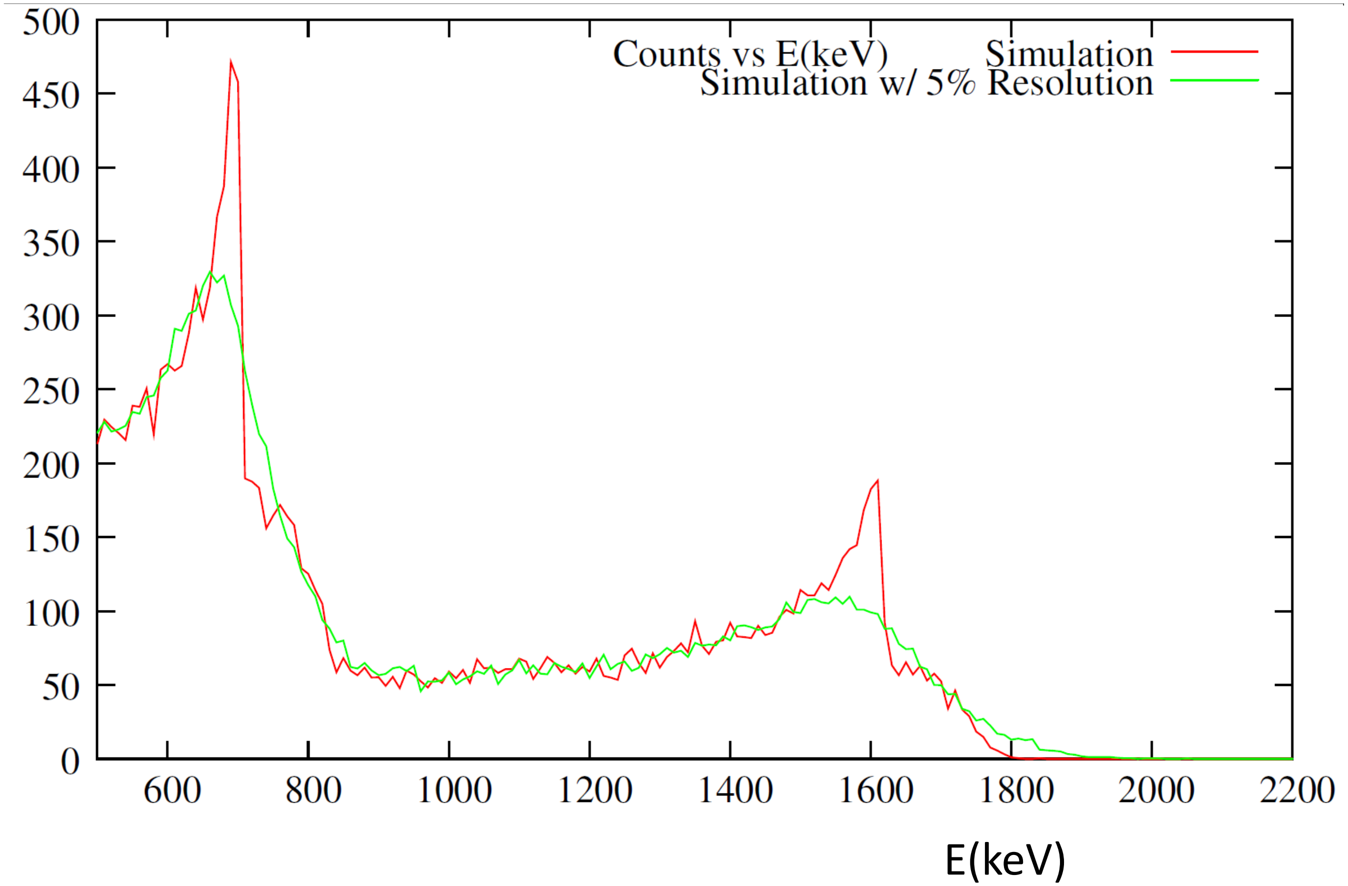
Plot of the calibraton run, counts vs channels:

Run#200437, B6D62@900V FS 0.5V, Offset 200mV, Threshold -228



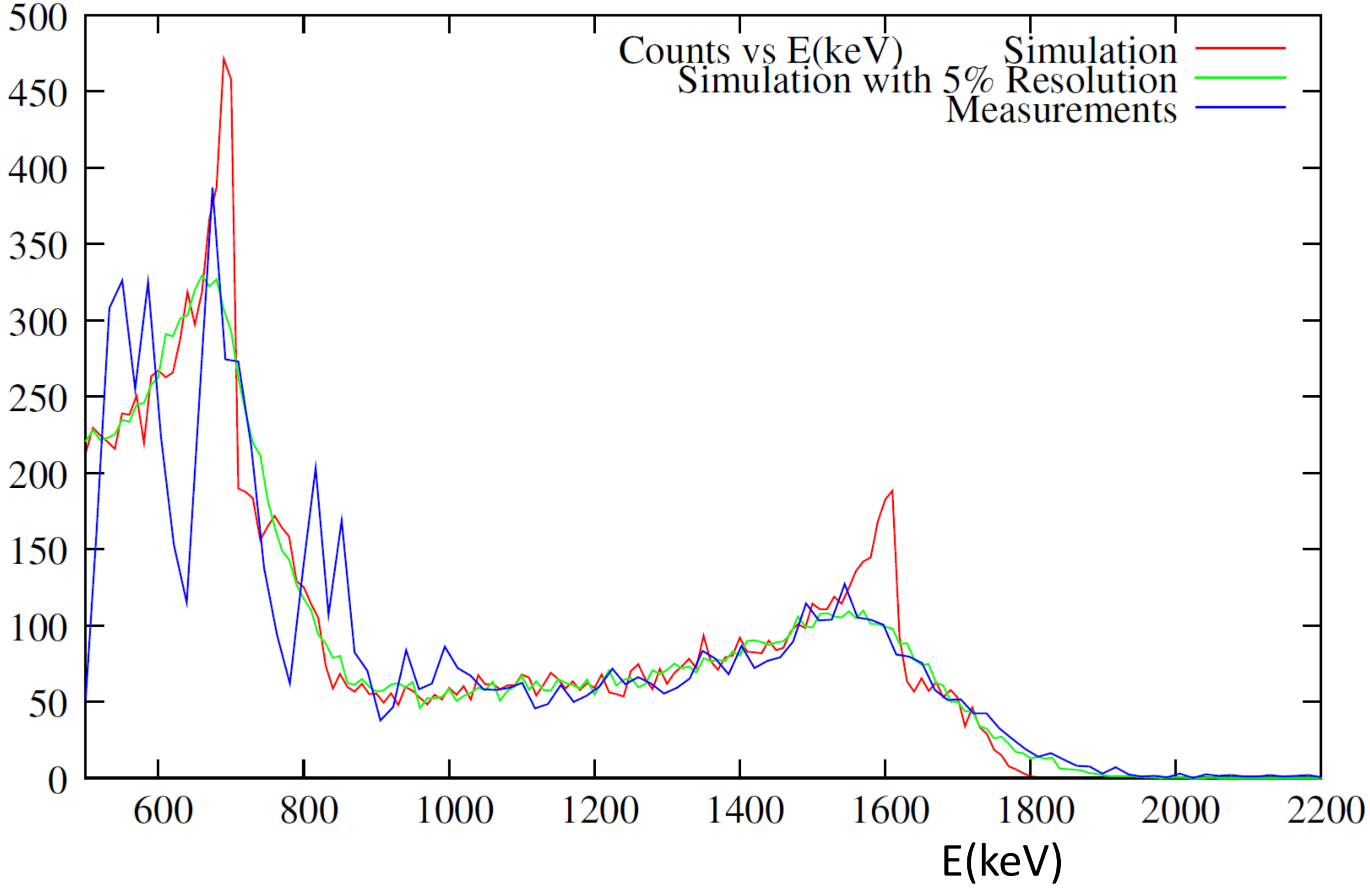
Detector response:

Convolution with a gaussian resolution



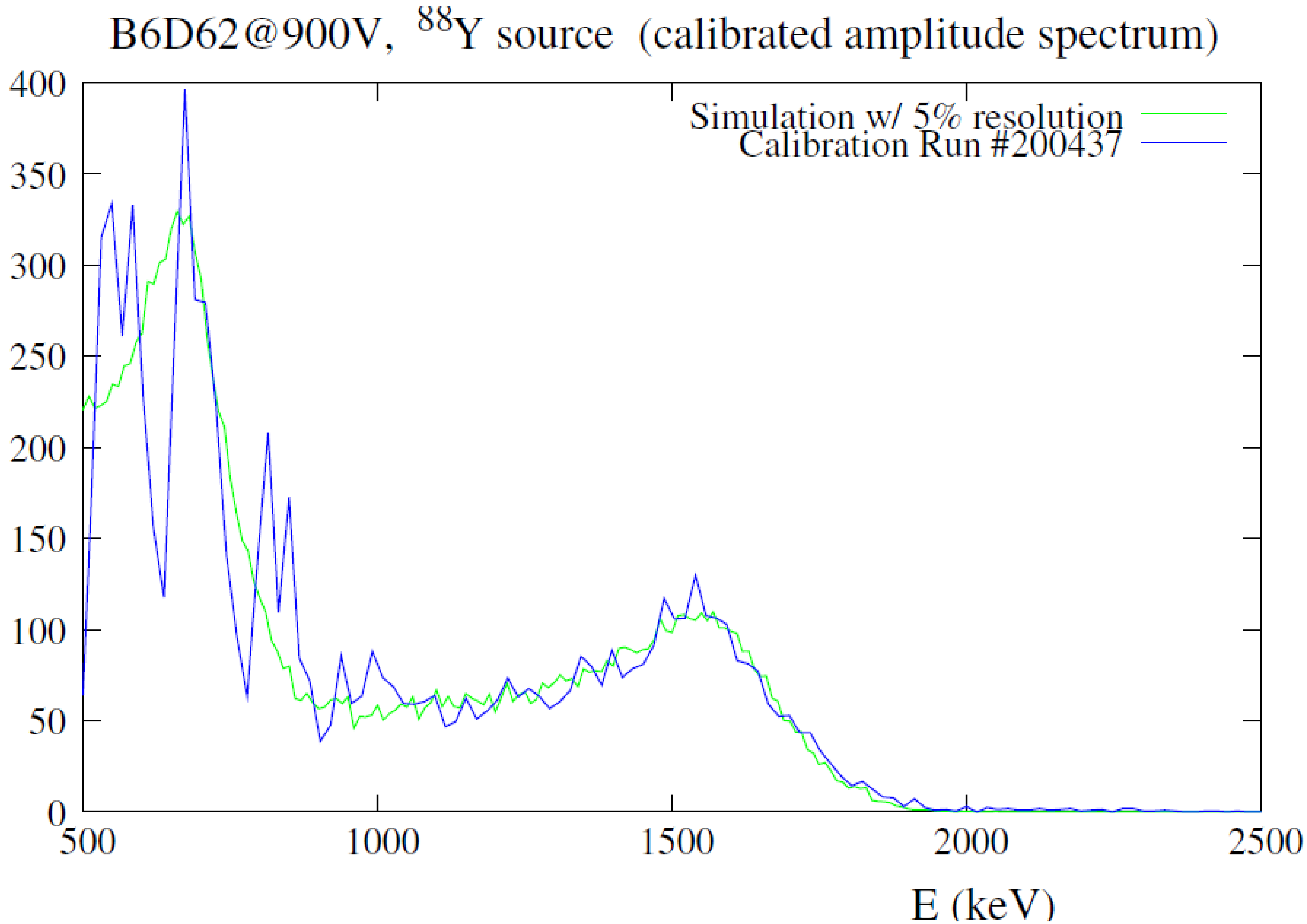
Detector response:

Linear transformation channel-energy of the experimental spectrum



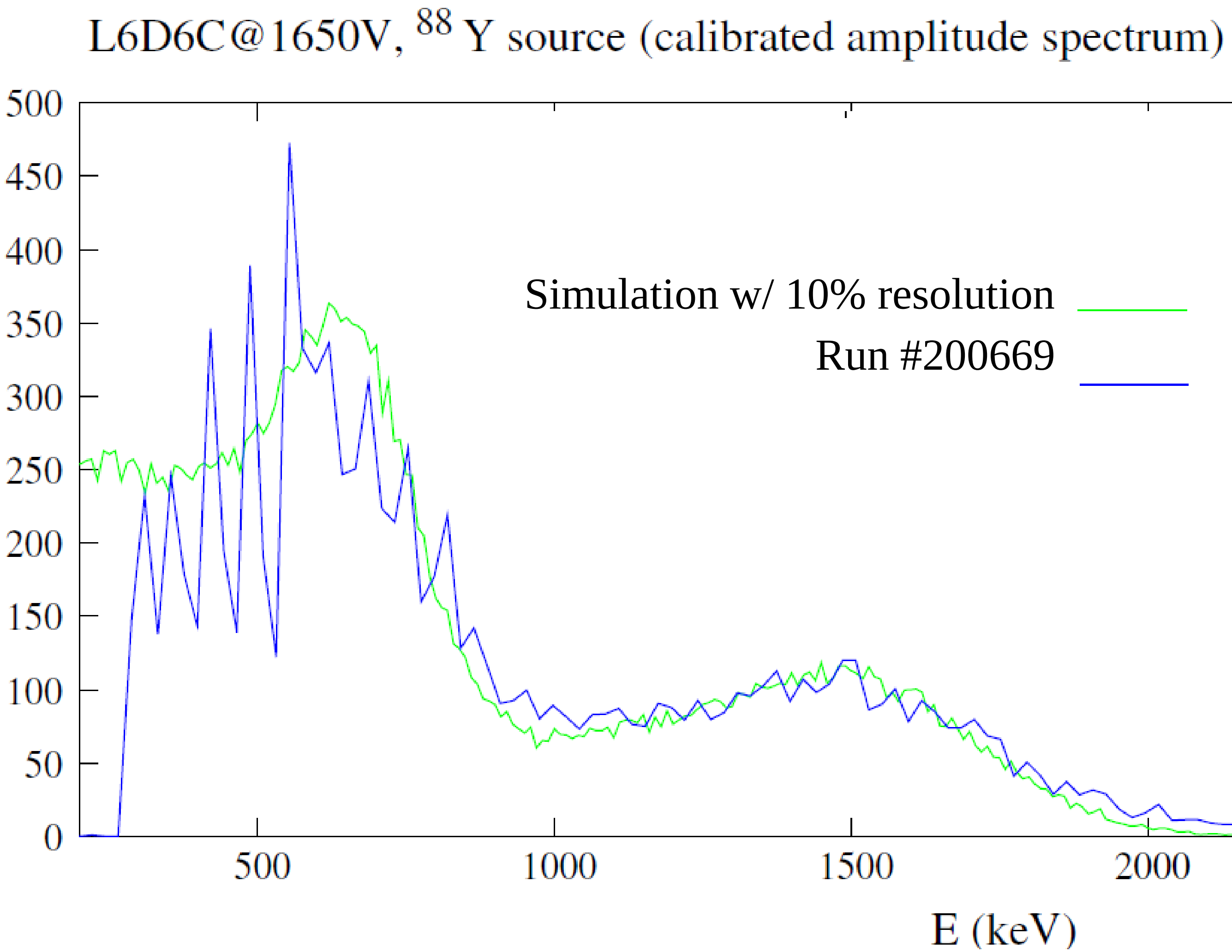
Detector response: calibration B6D6

E (keV) = *a* *n* (ch) + *b* *a* = 133.25 keV/ch *b* = 10 keV

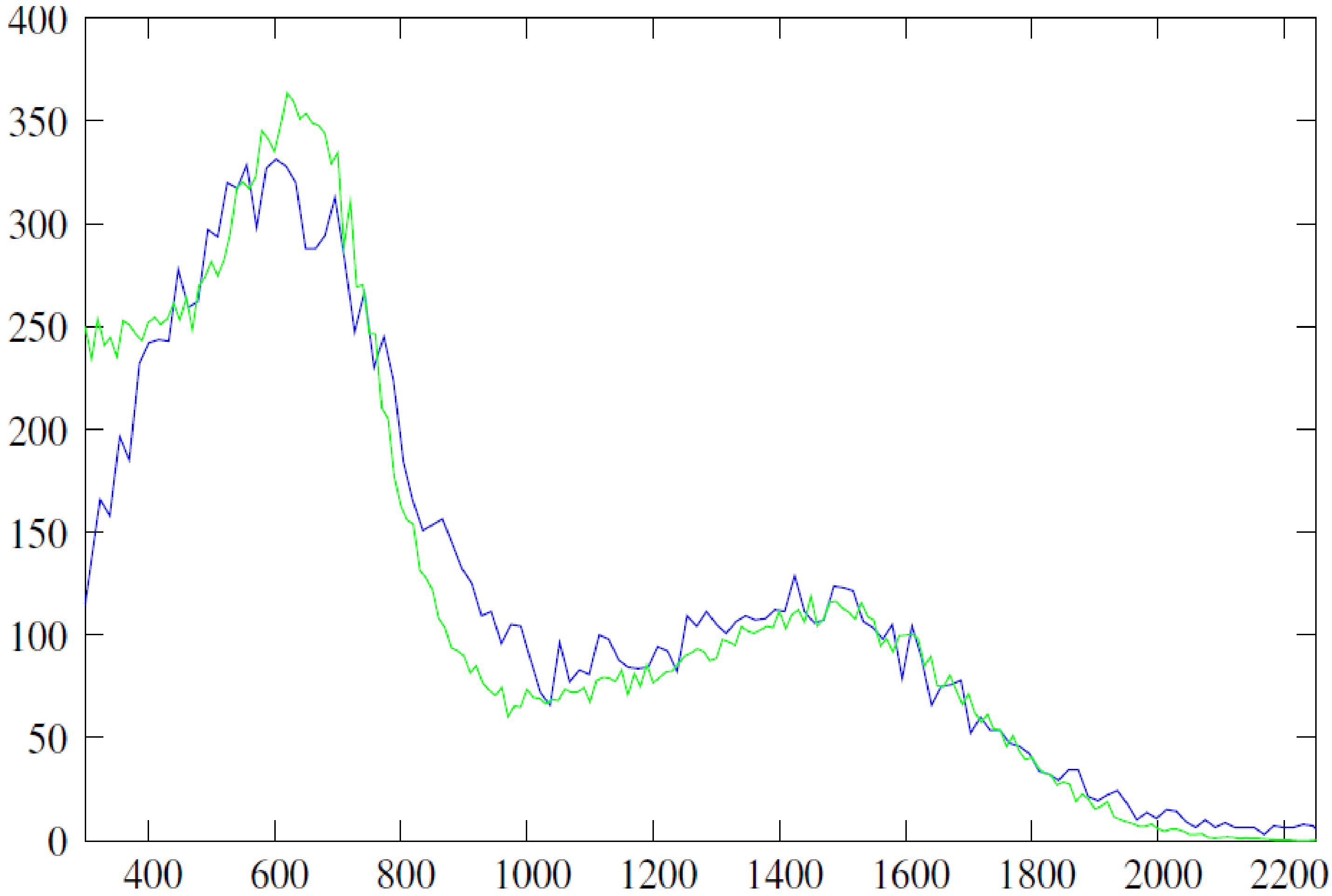


Detector response: calibration L6D6

E (keV) = a n (ch) + b a = 68.21 keV/ch b = 142 keV



Using area instead of amplitude



Calibration results:

Detector	a (keV/ch)	b (keV)	Run #
B6D61@800V	49.	82.	200440
B6D63@800V	71.	59.	100745
B6D63@900V	24.	62.	100744
L6D6B@1500V	120.	0.	100741
L6D6B@1600V	65.	0.	100742
L6D6C@1600V	85.	70.	200440

Background measurements EAR-2

All measurements: Full beam in vacuum

Configuration September 2014:

Detectors: B6D62@900V, L6D6C@1600V

Positions of detectors:

Top	#200465, 467-471	FS: 0.5V, Offset 200mV, Thr -228
Middle	#200446-454	FS: 0.5V, Offset 200mV, Thr -228
Low	#200442-444	FS: 0.5V, Offset 200mV, Thr -228

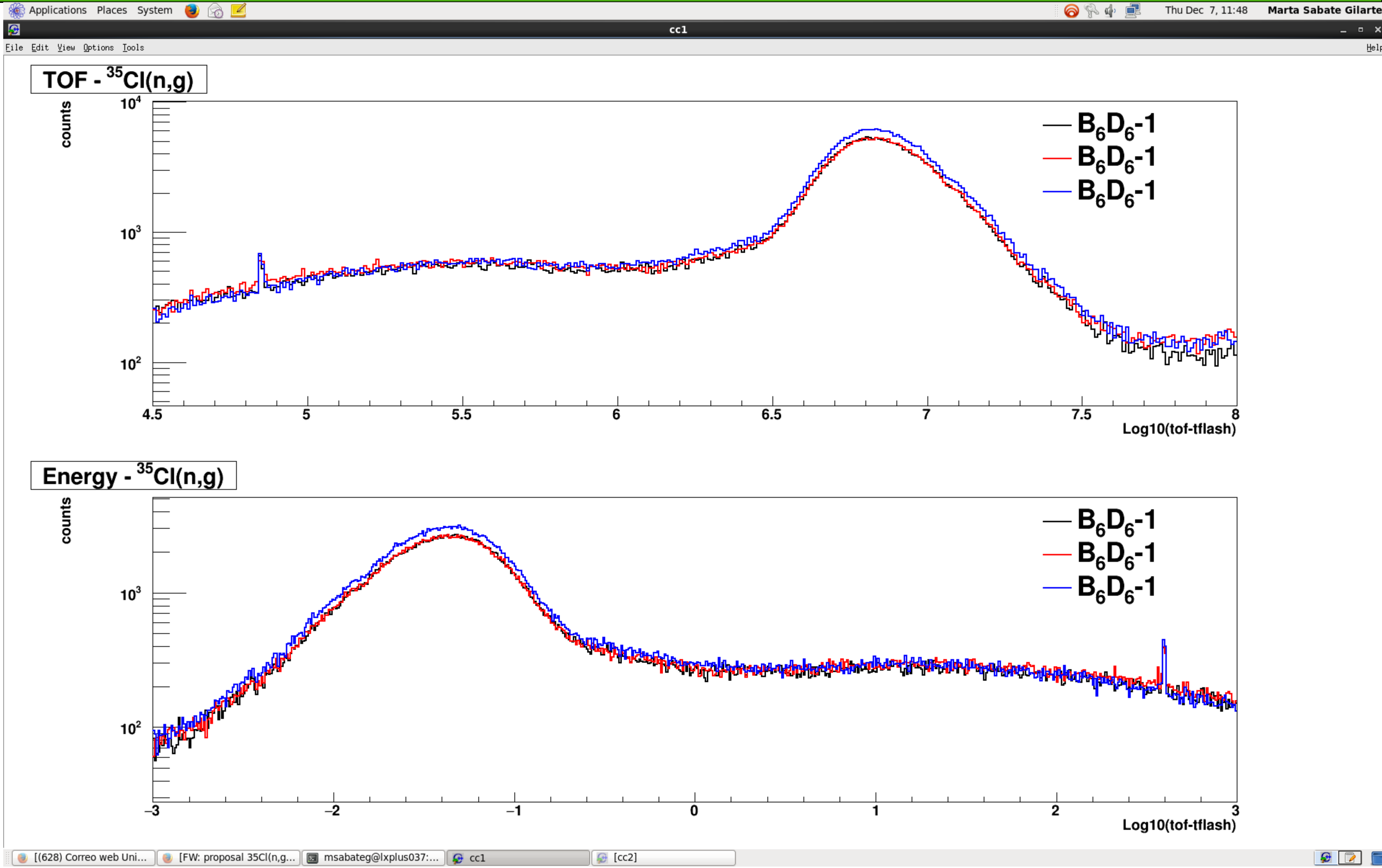
Configuration October 2014 (after shielding):

Detectors: B6D61@800V, B6D62@970V, L6D6C@1650V

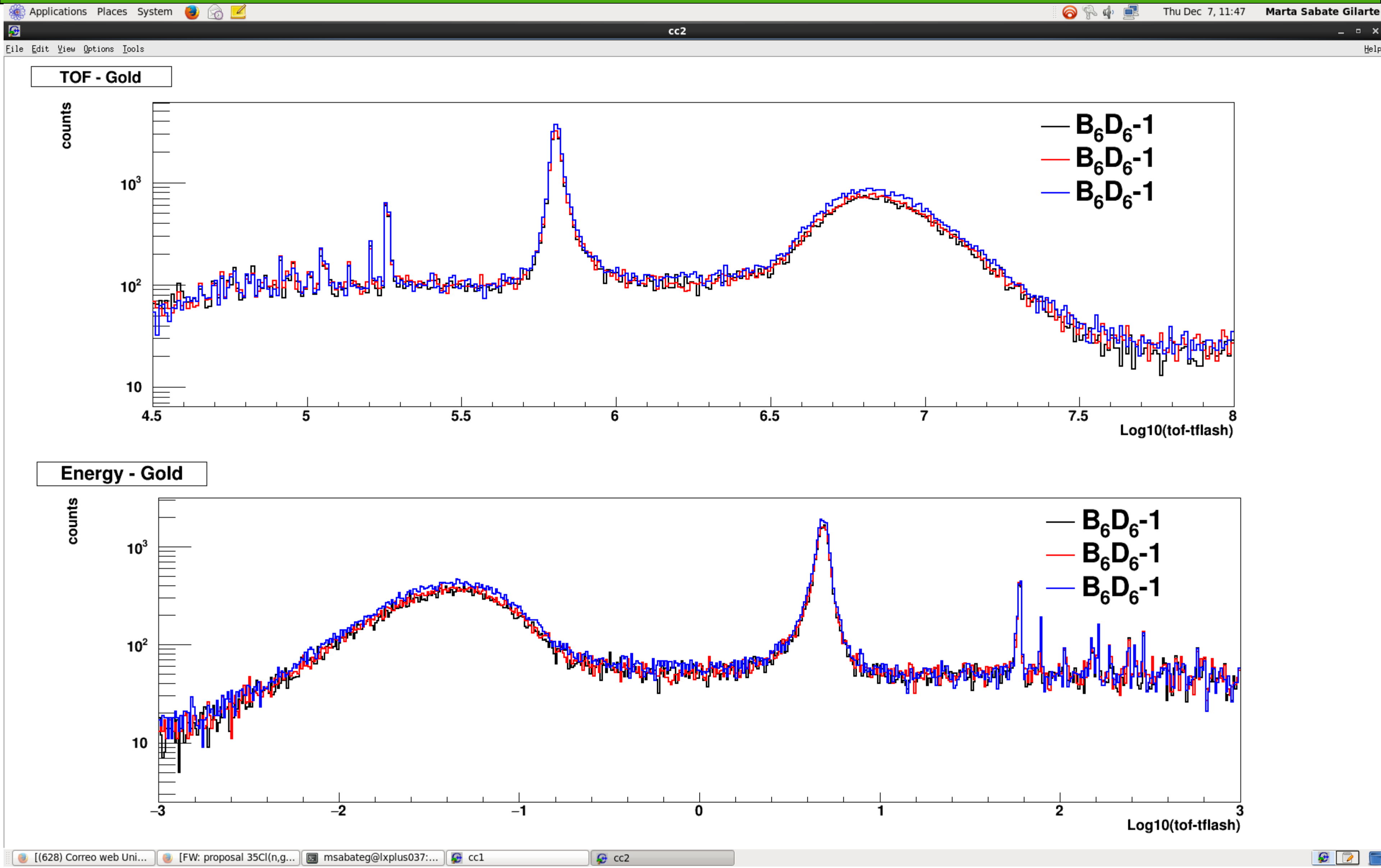
Positions of detectors:

Top	#200630-631	FS: 0.5V, Offset 200mV, Thr -240
Middle	#200696-708	FS: 1.0V, Offset -50mV, Thr -240
Low	#200626-627	FS: 0.5V, Offset 200mV, Thr -240

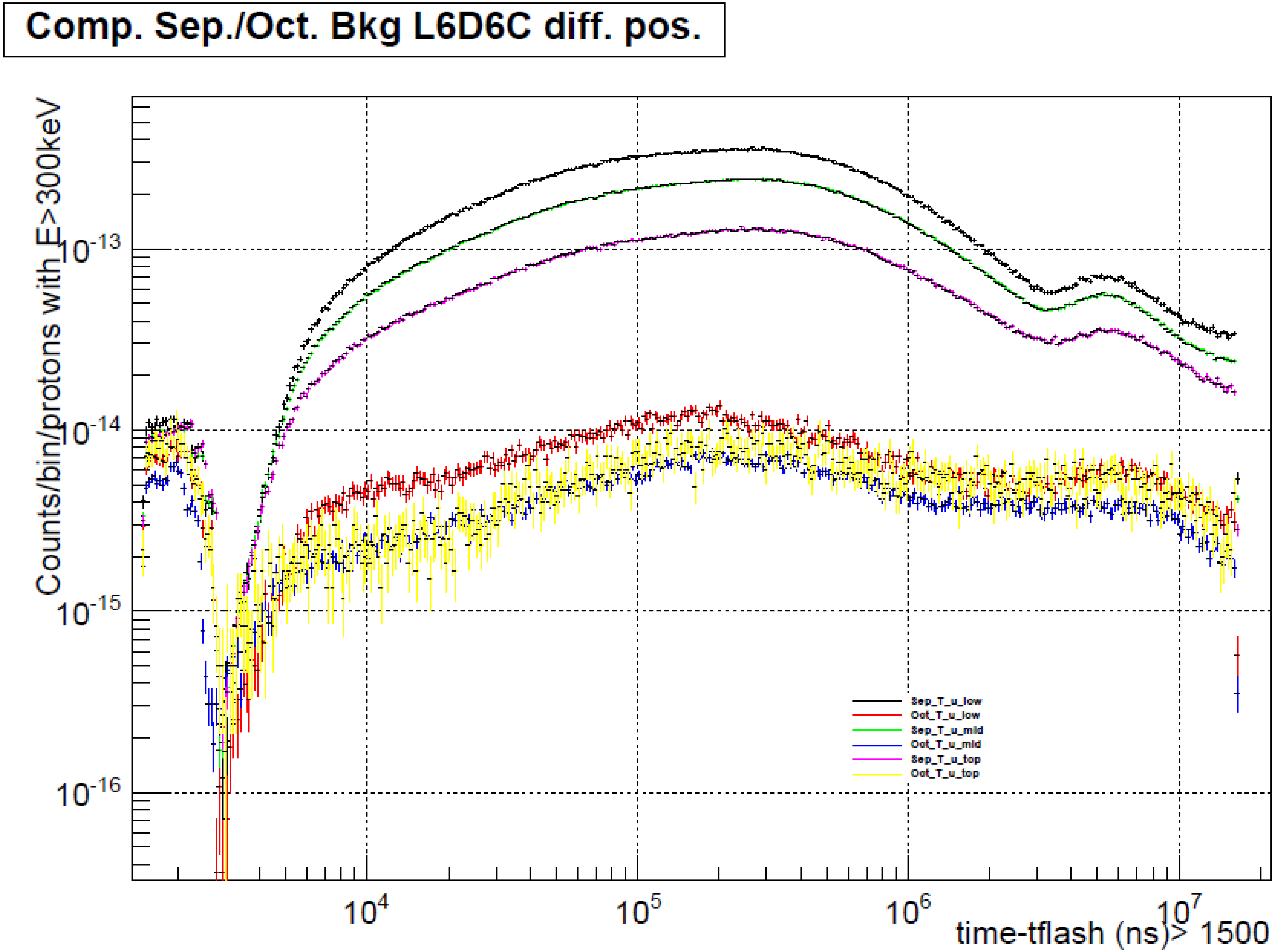
Tests EAR-2 (thans to Frank Gunsing)



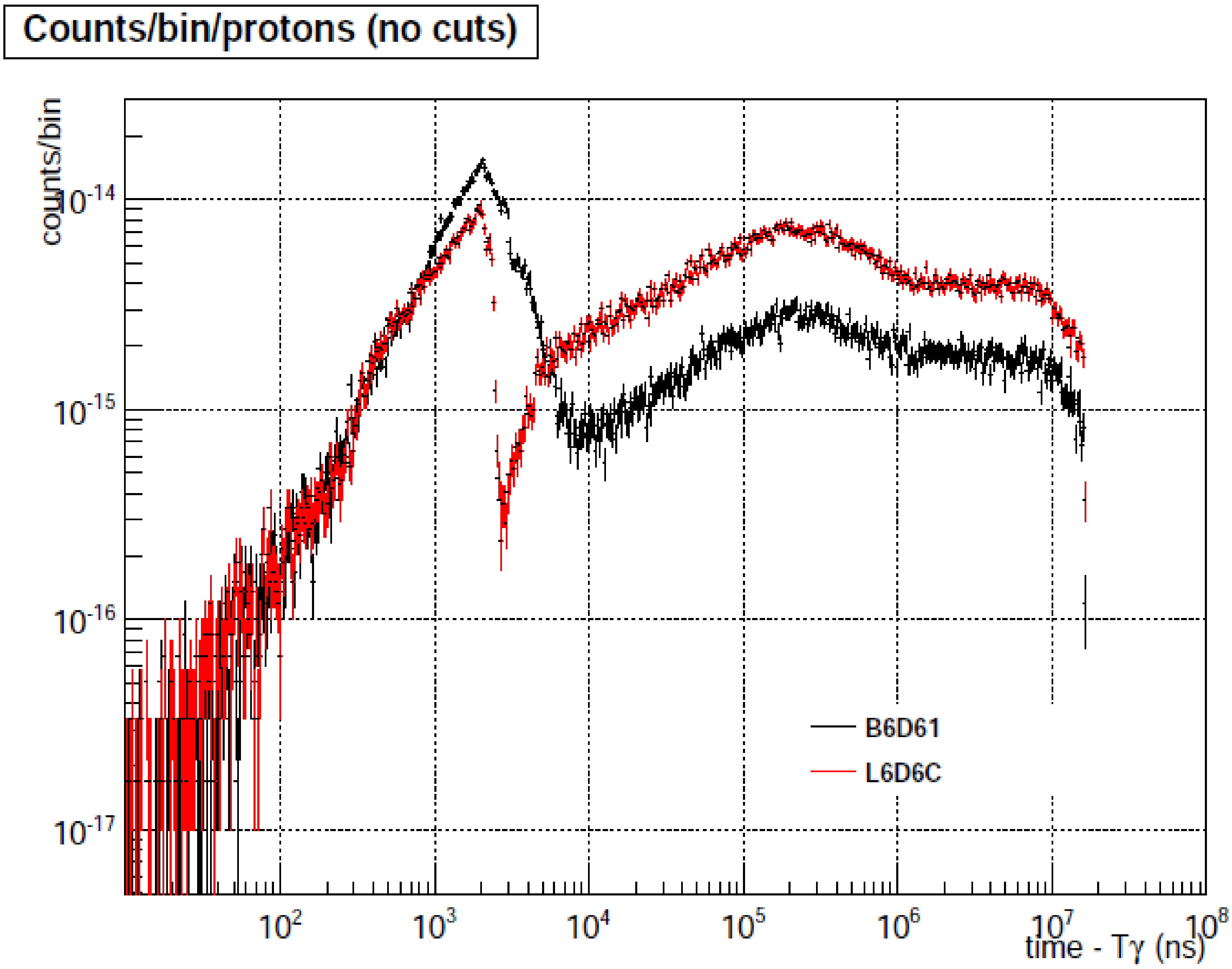
Gold for normalization



Background measurements EAR-2



Gamma-flash recovery



Detectors

- The response detector fits well the simulations assuming an energy resolution ΔE = const E
- B6D6: ΔE/E = 5%, L6D6: ΔE/E = 10%
- Efficiency and recovery from γ-flash of L6D6 are superior

BNCT Treatment Planning

Tissue		Dose prescribed with photons (Gy)	Photon Isoeffective Dose of the BNCT treatment (Gy-Eq)	Target photon Isoeffective Dose BNCT treatment (Gy-Eq)
Healthy tissue	Fractionated:	60 (30 sessions of 2 Gy)	41.5 (21 sessions of 2 Gy)	60 (30 sessions of 2 Gy)
	Single-fraction:	16	12.1	16.0
Brain tumor	Fractionated:	60 (30 sessions of 2 Gy)	50.5 (25 sessions of 2 Gy)	71.0 (35 sessions of 2 Gy)
	Single-fraction:	41.5	46.7	61,6 (PTVdose of 50 Gy)

- BNCT delivered with safe margin (12.1 Gy to healthy tissue, could receive up to 16 Gy)
- A more precise dose calculation (improving all cross sections) could lead to optimize treatment time ⇒ **Better therapeutic outcome and possibly survival**