“Nuclear data for applications: Evaluation methodology and examples”

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OUTLINE

- ☐ Introduction
- ☐ What is Nuclear Data Evaluation?
- ☐ ND evaluation methods
- ☐ Selection of experimental data and EXFOR
- ☐ Experimental uncertainties and correlations
- ☐ Modelling uncertainties and model defects
- ☐ Bayesian approaches: GLSQ
- ☐ Selected ND evaluation challenges

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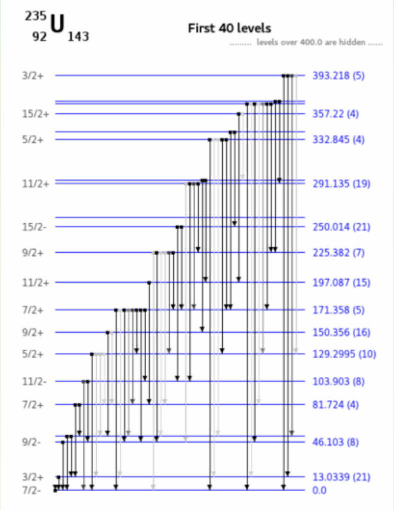
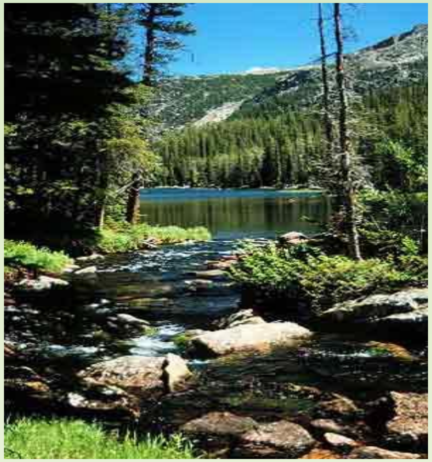
What is Nuclear Data?

The numbers underlying nuclear applications



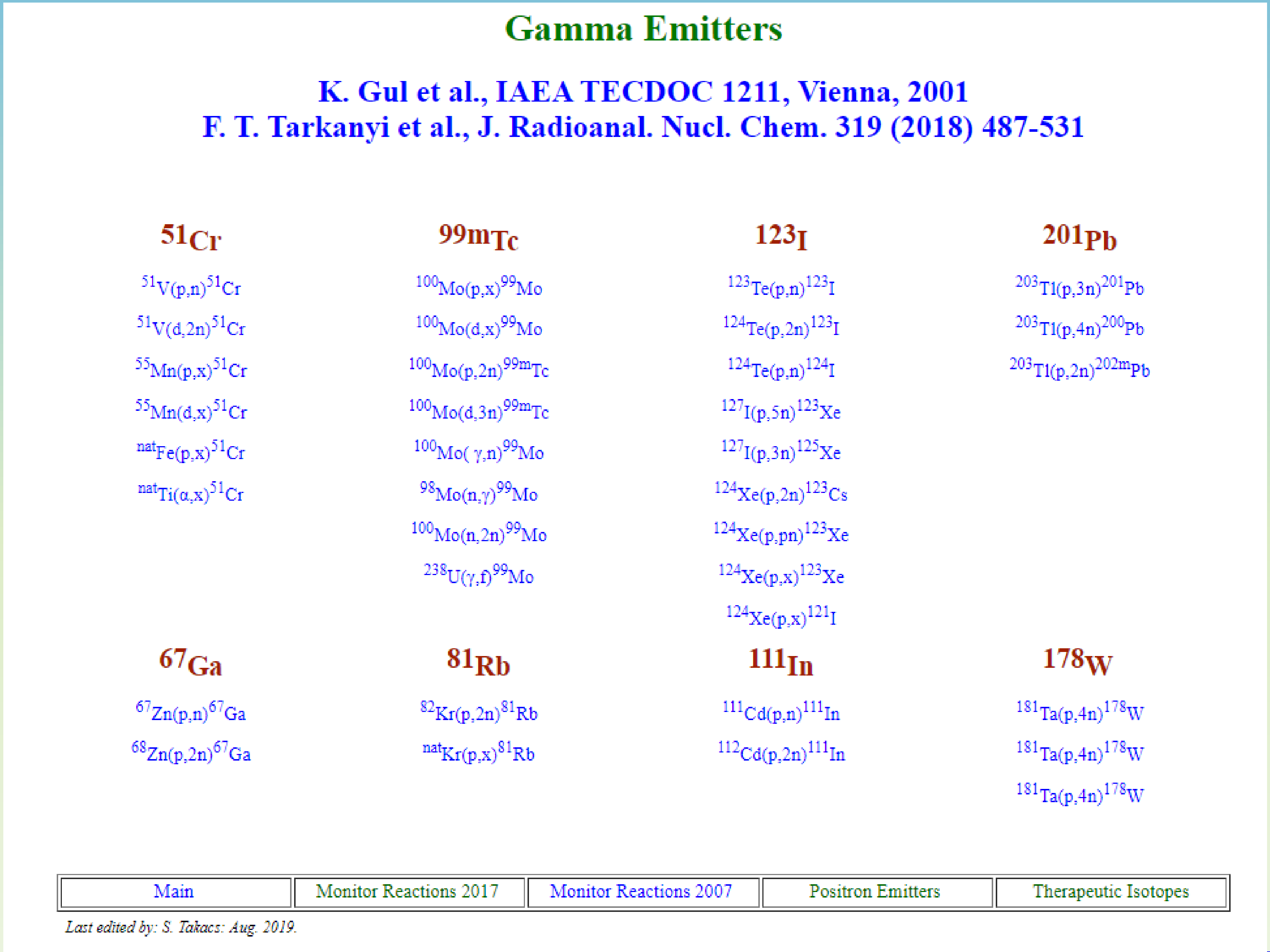


Nucl. Sciences & Applications: Serving Basic Human Needs

THREE DISTINCT MEDICAL FIELDS

- Diagnostic radiology
  - 100% diagnostic
- Radiotherapy
  - 100% treatment
- Nuclear medicine
  - 80% diagnostic
  - 10% treatment

Multidisciplinary team: physicians, physicists, radiographers,..



Gamma Emitters

K. Gul et al., IAEA TECDOC 1211, Vienna, 2001
F. T. Tarkanyi et al., J. Radioanal. Nucl. Chem. 319 (2018) 487-531

^{51}Cr

^{51}V(p,n)^{51}Cr
^{51}V(d,2n)^{51}Cr
^{55}Mn(p,x)^{51}Cr
^{55}Mn(d,x)^{51}Cr
^{nat}Fe(p,x)^{51}Cr
^{nat}Ti(α,x)^{51}Cr

^{99m}Tc

^{100}Mo(p,x)^{99}Mo
^{100}Mo(d,x)^{99}Mo
^{100}Mo(p,2n)^{99m}Tc
^{100}Mo(d,3n)^{99m}Tc
^{100}Mo(γ,n)^{99}Mo
^{98}Mo(n,γ)^{99}Mo
^{100}Mo(n,2n)^{99}Mo
^{238}U(γ,f)^{99}Mo

^{123}I

^{123}Te(p,n)^{123}I
^{124}Te(p,2n)^{123}I
^{124}Te(p,n)^{124}I
^{127}I(p,5n)^{123}Xe
^{127}I(p,3n)^{125}Xe
^{124}Xe(p,2n)^{123}Cs
^{124}Xe(p,pn)^{123}Xe
^{124}Xe(p,x)^{123}Xe
^{124}Xe(p,x)^{121}I

^{201}Pb

^{203}Tl(p,3n)^{201}Pb
^{203}Tl(p,4n)^{200}Pb
^{203}Tl(p,2n)^{202m}Pb

^{67}Ga

^{67}Zn(p,n)^{67}Ga
^{68}Zn(p,2n)^{67}Ga

^{81}Rb

^{82}Kr(p,2n)^{81}Rb
^{nat}Kr(p,x)^{81}Rb

^{111}In

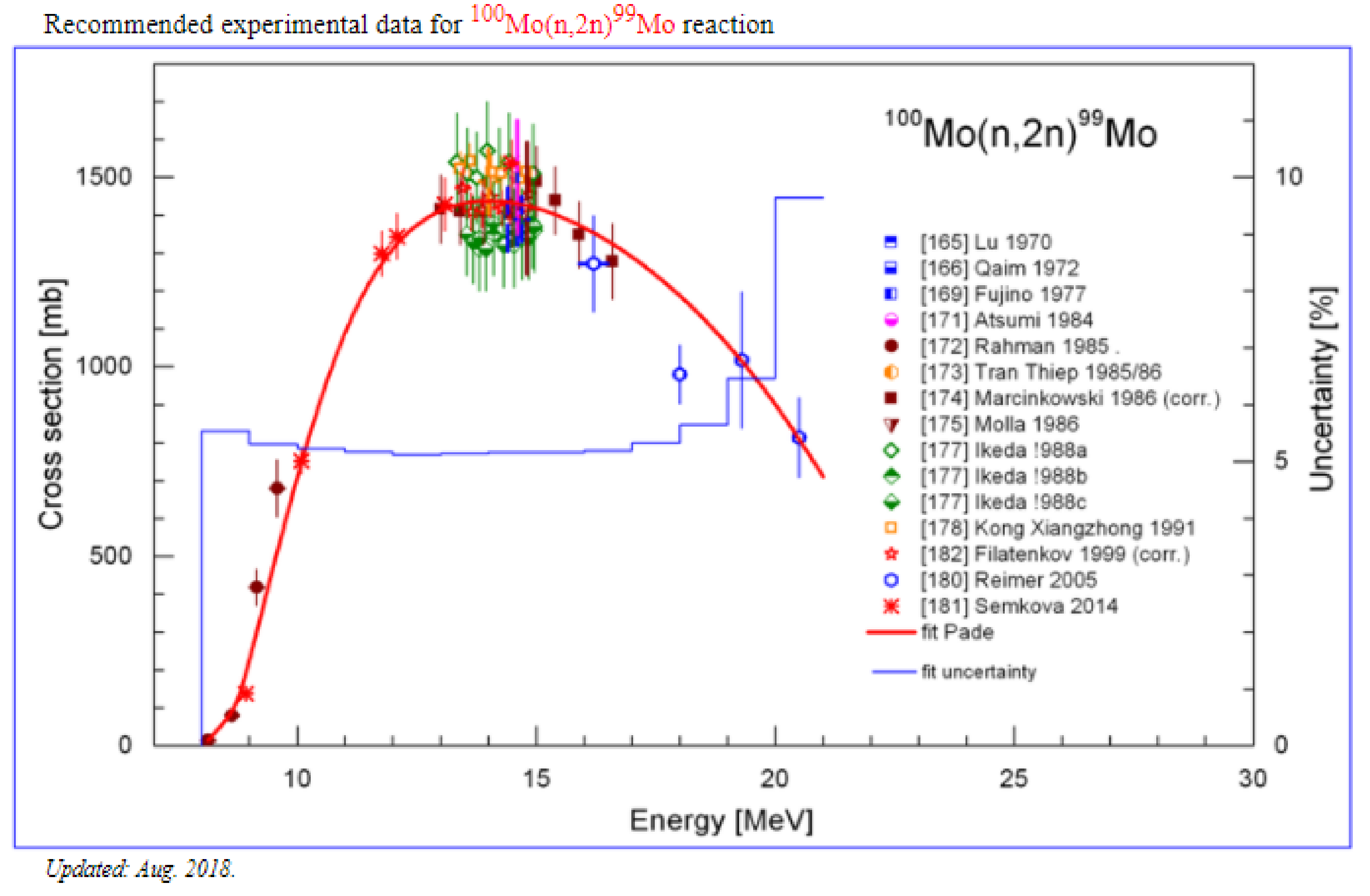
^{111}Cd(p,n)^{111}In
^{112}Cd(p,2n)^{111}In

^{178}W

^{181}Ta(p,4n)^{178}W
^{181}Ta(p,4n)^{178}W
^{181}Ta(p,4n)^{178}W

Last edited by: S. Takacs: Aug. 2019.

Mo-99 production via (n,2n) reaction (deployed in Japan)



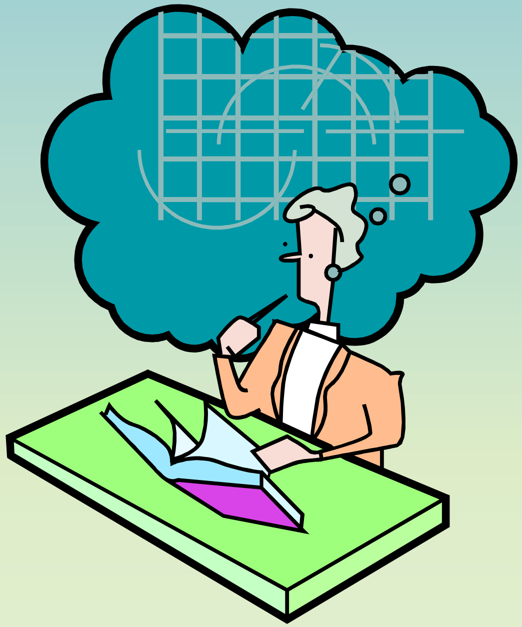
Recommended experimental data for $^{100}Mo(n,2n)^{99}Mo$ reaction

Updated: Aug. 2018.

What is Nuclear Data Evaluation?

What do engineers need?

I need cross sections …

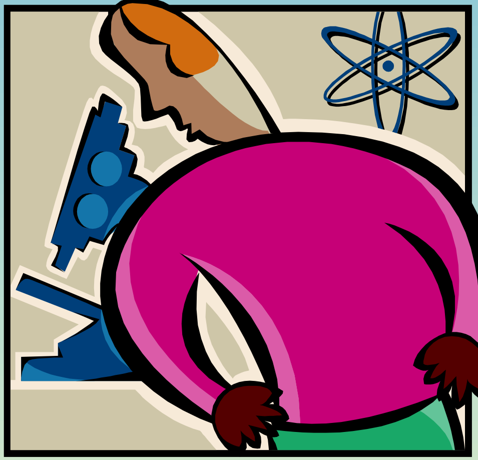


$$-\nabla D_{(1)}\nabla\phi_{(1)}+\Sigma_{a(1)}\phi_{(1)}=\frac{1}{k}\left[\sum_{g}\nu_{(g)}\Sigma_{f(g)}\phi_{(g)}\right]+\Sigma_{(2\to1)}\phi_{(2)}$$

$$-\nabla D_{(2)}\nabla\phi_{(2)}+\Sigma_{a(2)}\phi_{(2)}=\Sigma_{(1\to2)}\phi_{(1)}$$

What theoreticians provide?

Here you are …
It is all described in
my article in the journal !



$$\sigma_a\left(U,J,\pi\right)=\frac{\pi}{k^2}\frac{\left(2J+1\right)}{\left(2I+1\right)\left(2i+1\right)}\sum_{S=\left|I-i\right|}^{I+i}\;\sum_{l=\left|J-S\right|}^{J+S} f\left(l,\pi\right)T_l^{a}\left(\varepsilon\right)$$

Or even worse …
Please use my calculations …

1.00000E-03	2.43758E+01	1.26322E+01	1.17435E+01
5.00000E-03	1.75172E+01	5.98889E+00	1.15283E+01
1.00000E-02	1.58838E+01	4.55133E+00	1.13325E+01
2.00000E-02	1.46751E+01	3.66517E+00	1.10099E+01
4.00000E-02	1.36734E+01	3.19591E+00	1.04775E+01
7.00000E-02	1.28627E+01	3.03899E+00	9.81379E+00

Calculations ARE NOT evaluations

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What is
Nuclear Data Evaluation?

A properly weighted combination (usually by GLSQ model) of selected experimental data (and nuclear reaction modelling results if needed).

Both to decay & reaction data evals

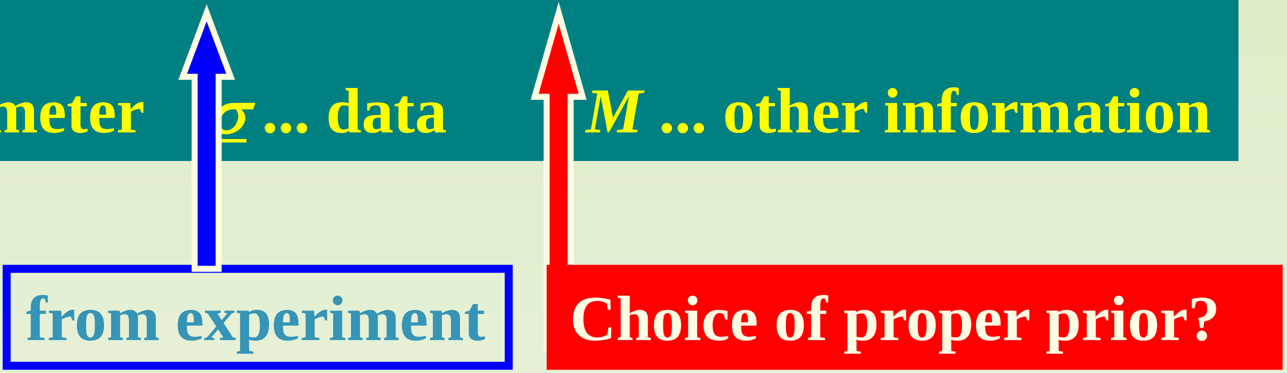
Bayes theorem: A proper combination of data and model

Bayes Theorem (1763):

$$p(\underline{x}|\underline{\sigma}M) \;=\; p(\underline{\sigma}|\underline{x}M)\quad p(\underline{x}|M) \;/\; p(\underline{\sigma}|M)$$

posterior = likelihood x prior / evidence

$\underline{x}$ ... model parameter **$\underline{\sigma}$ ... data** **M ... other information**



from experiment

Choice of proper prior?

From D. Neudecker, S. Gundacker, H. Leeb *et al.*, ND2010, Jeju Isl., Korea

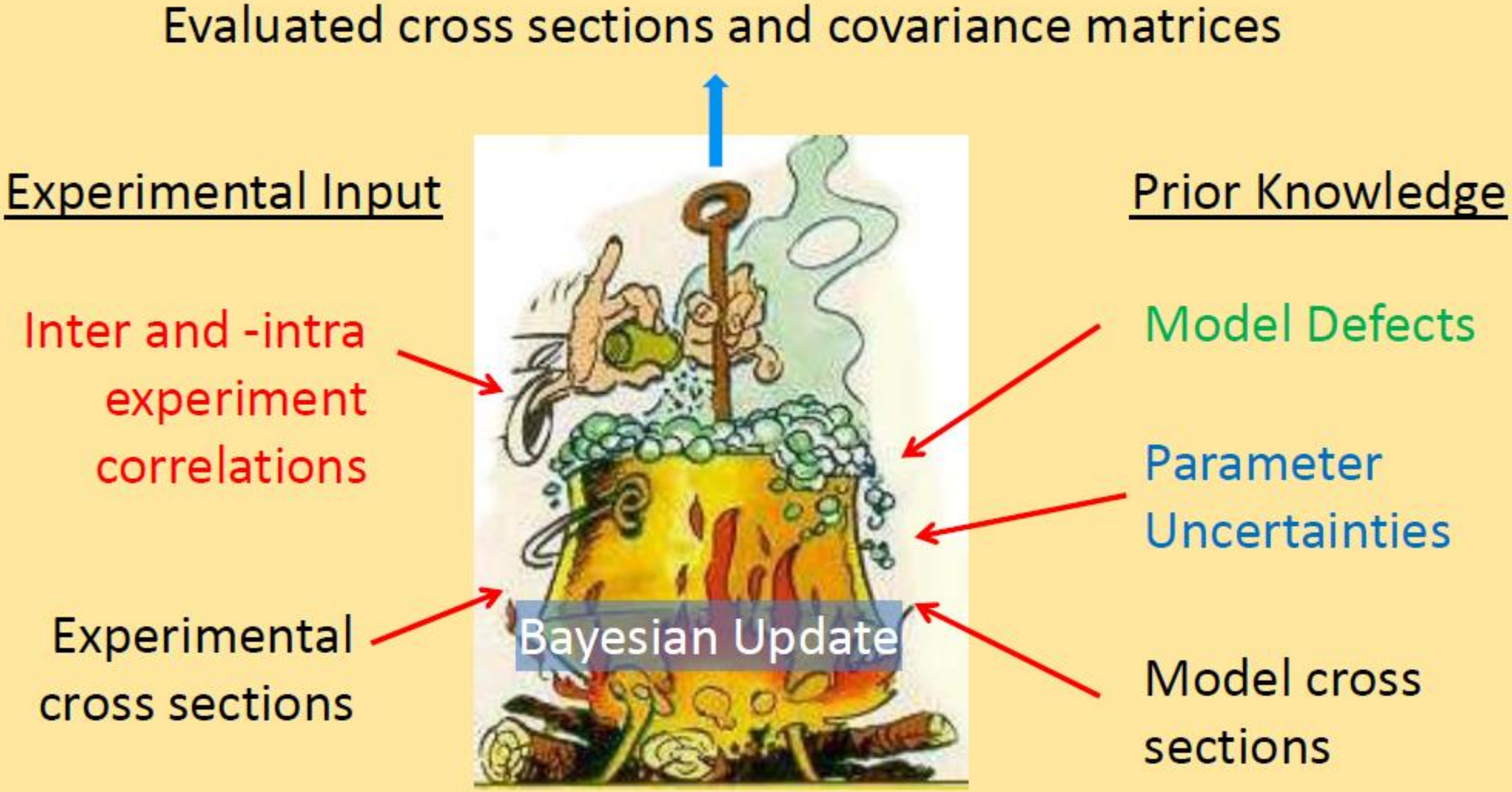
Definition of (ND) Evaluation

A properly weighted combination (usually by GLSQ fit) of selected experimental data (and nuclear reaction modelling results).

Bayesian approaches **(properly weighted)**:

- ☐ "Non-model" GLSQ fit: Neutron standards
- ☐ Model prior + GLSQ fit (in XS space)
- ☐ Parameter prior + Parameter fit of data (in parameter space)

ND Evaluation



Evaluated cross sections and covariance matrices

Experimental Input

Inter and -intra experiment correlations

Experimental cross sections

Bayesian Update

Prior Knowledge

Model Defects

Parameter Uncertainties

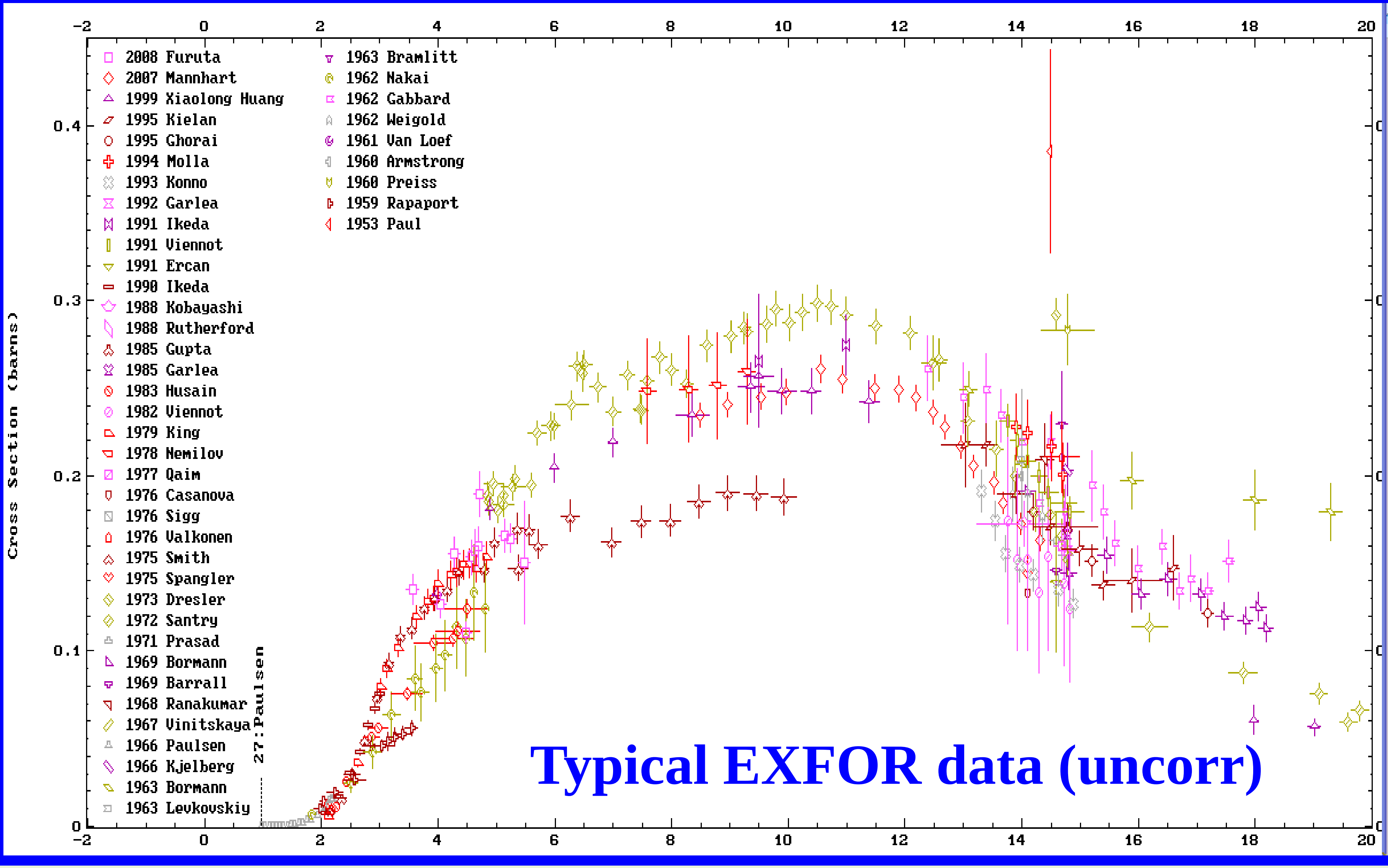
Model cross sections

From D. Neudecker, S. Gundacker, H. Leeb *et al.*, ND2010, Jeju Isl., Korea

Experimental uncertainties:

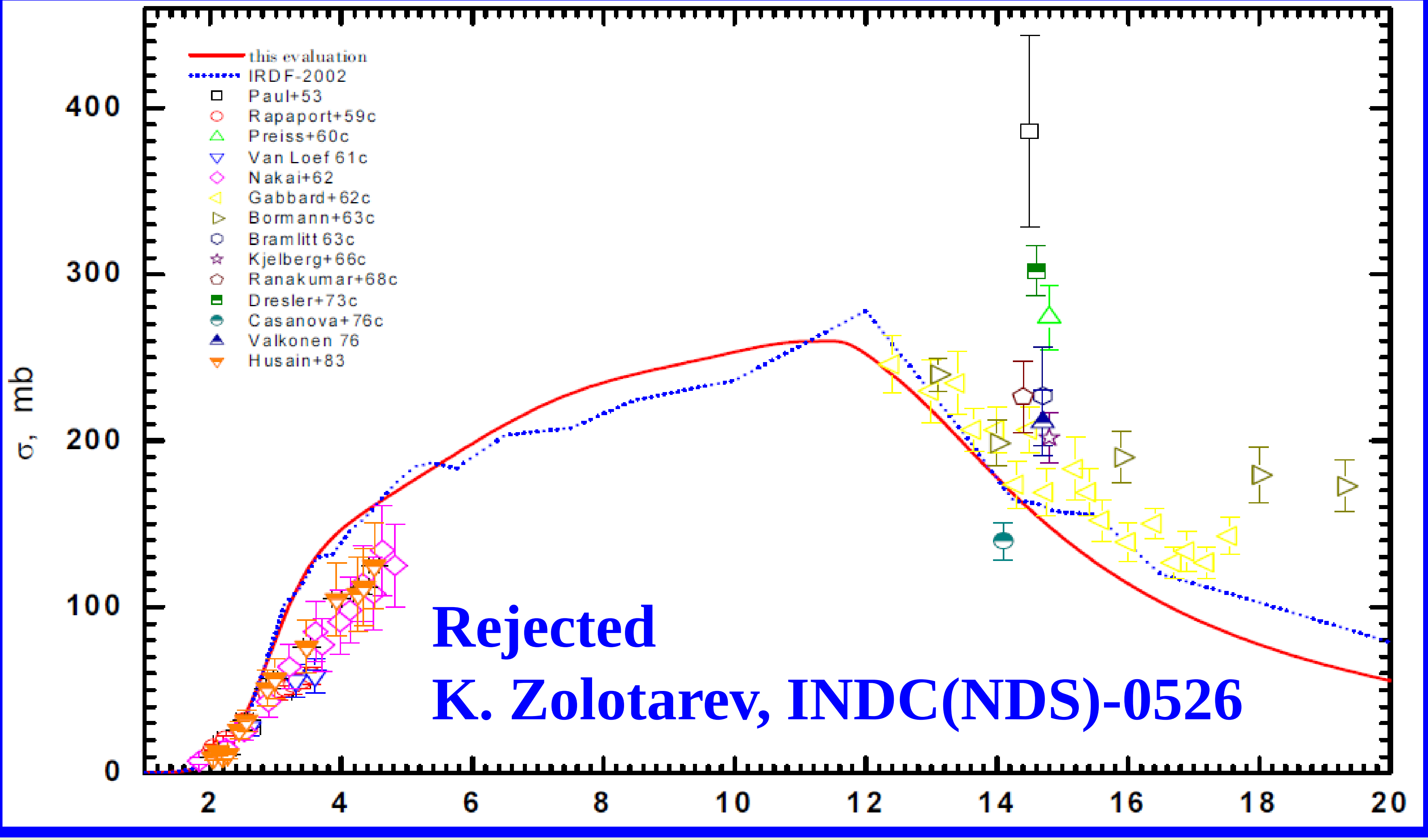
This is a critical component of high-quality evaluations defining the posterior uncertainty

Selection of experimental data (1)



Typical EXFOR data (uncorr)

Selection of experimental data (2)



Rejected

K. Zolotarev, INDC(NDS)-0526

http://www-nds.iaea.org/reports-new/indc-reports/indc-nds/indc-nds-0526.pdf

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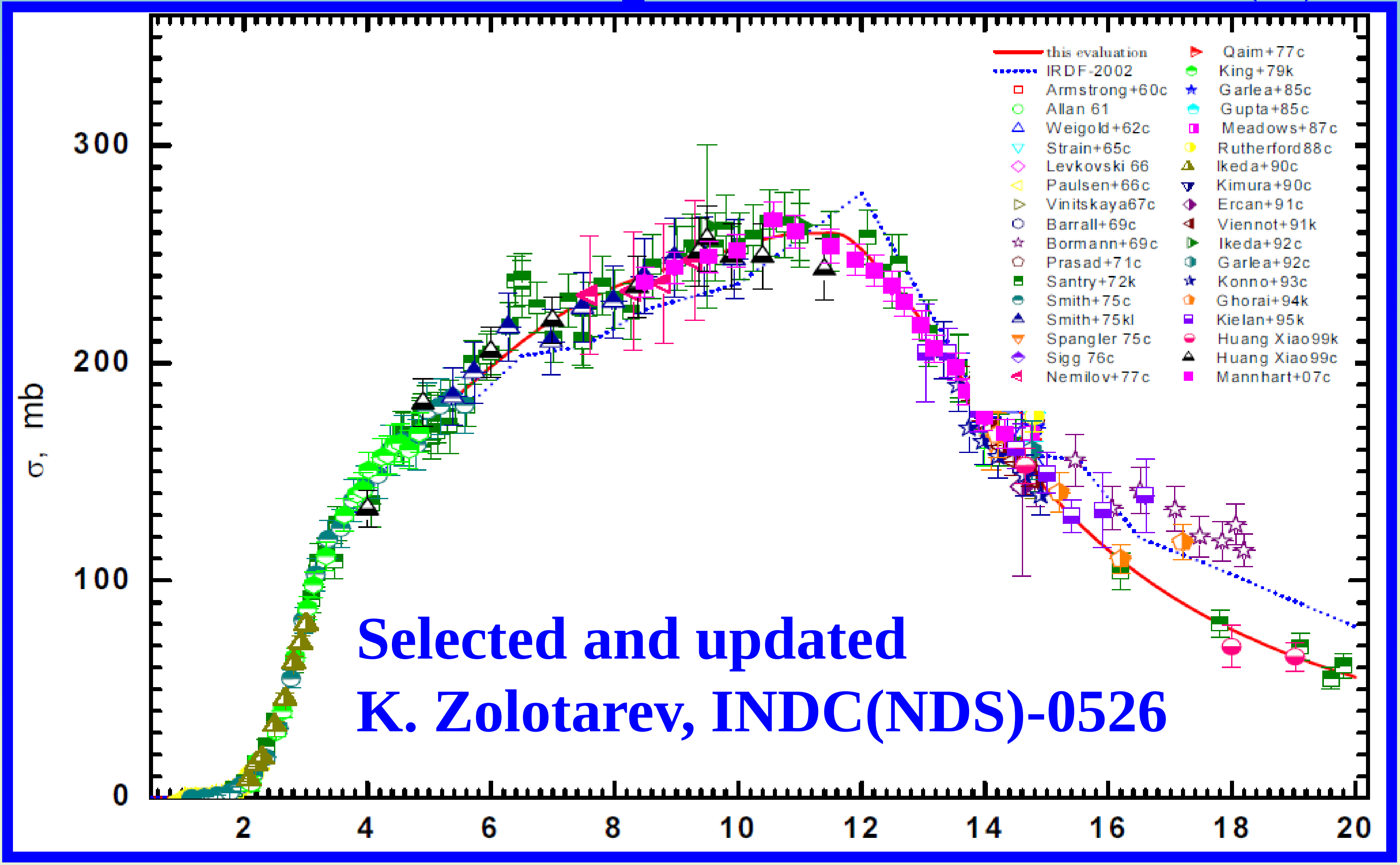
Selection of experimental data (3)



Selected and updated
K. Zolotarev, INDC(NDS)-0526

http://www-nds.iaea.org/reports-new/indc-reports/indc-nds/indc-nds-0526.pdf

Experimental correlations

"Evaluation of measurement data - Guide to the expression of uncertainty in measurement"

Joint Committee for Guides in Metrology,

JCGM 100:2008, www.bipm.org (2008)

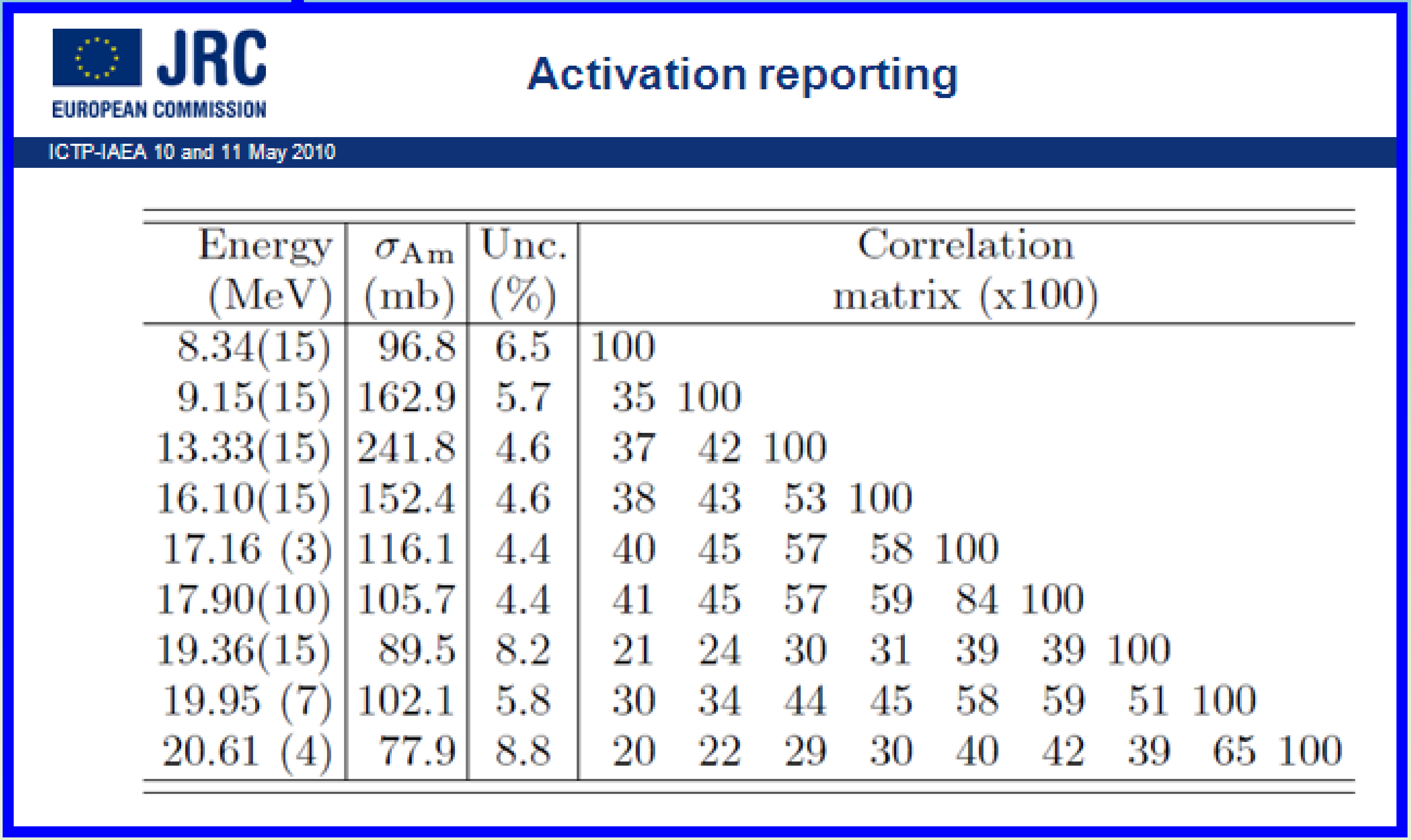
- ☐ Intra experiments correlations:

  Short and long term correlations within a single experiment can and should be estimated

  (statistical and systematic uncertainty)
- ☐ Inter-experiments correlations

  (very often neglected, default zero !!!)

Experimental uncertainties



JRC
EUROPEAN COMMISSION

Activation reporting

ICTP-IAEA 10 and 11 May 2010

Energy (MeV)	σ_{Am} (mb)	Unc. (%)	Correlation matrix (x100)								
8.34(15)	96.8	6.5	100								
9.15(15)	162.9	5.7	35	100							
13.33(15)	241.8	4.6	37	42	100						
16.10(15)	152.4	4.6	38	43	53	100					
17.16 (3)	116.1	4.4	40	45	57	58	100				
17.90(10)	105.7	4.4	41	45	57	59	84	100			
19.36(15)	89.5	8.2	21	24	30	31	39	39	100		
19.95 (7)	102.1	5.8	30	34	44	45	58	59	51	100	
20.61 (4)	77.9	8.8	20	22	29	30	40	42	39	65	100

A. Plompen, ND workshop, ICTP, Trieste 2010

Model uncertainties

Model parameter uncertainties

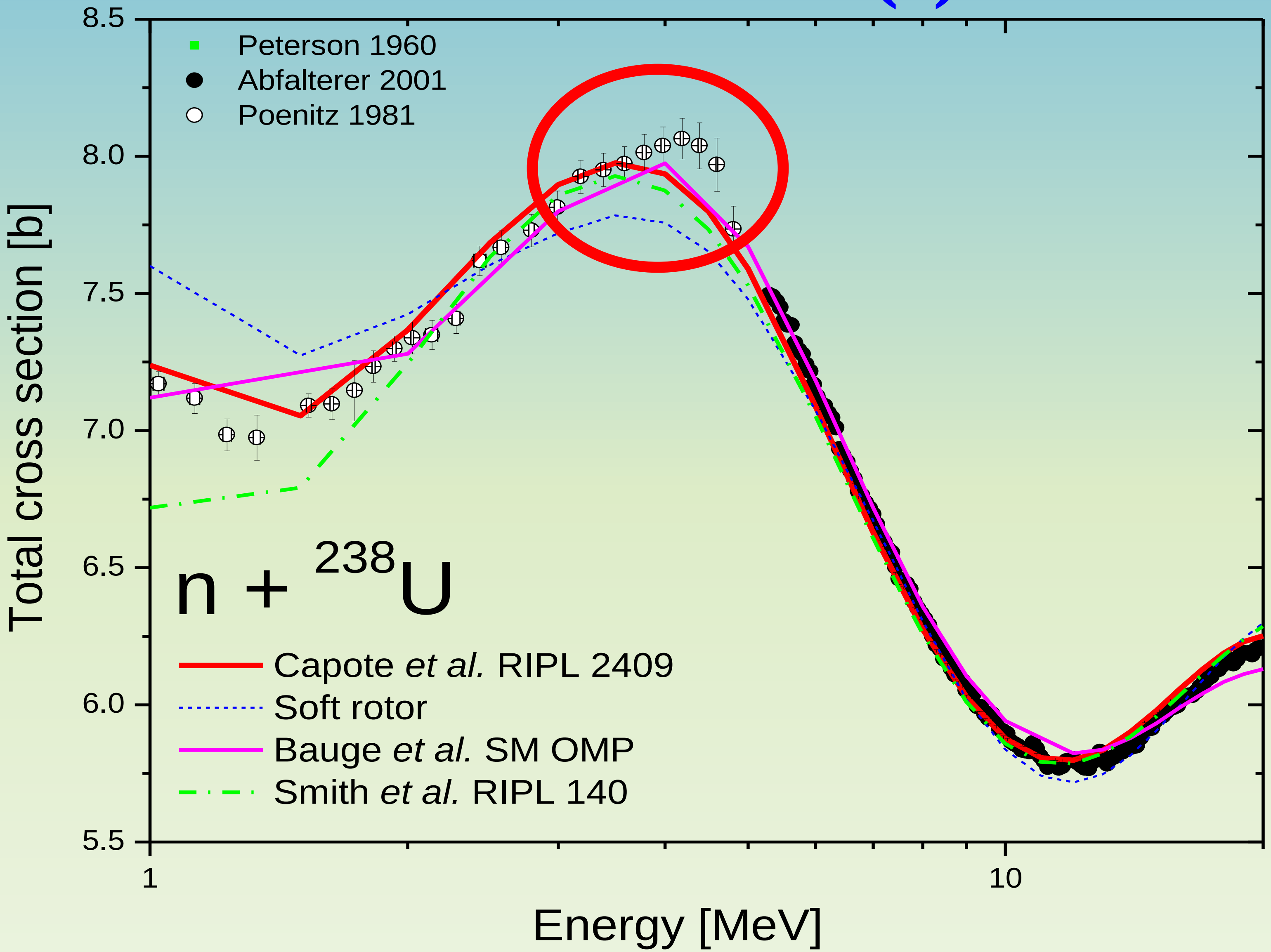
D.L. Smith, “Covariance Matrices for Nuclear Cross-Sections Derived from Nuclear Model Calculations”. Report **ANL/NDM-159**, Argonne National Laboratory, 2005

$$\overline{\sigma}_i = \frac{1}{K}\sum_{k=1}^{K}\sigma_{ik} \qquad V_{ij} = \overline{\sigma_i\sigma_j} - \overline{\sigma_i}\times\overline{\sigma_j}$$

i,j - energy indexes

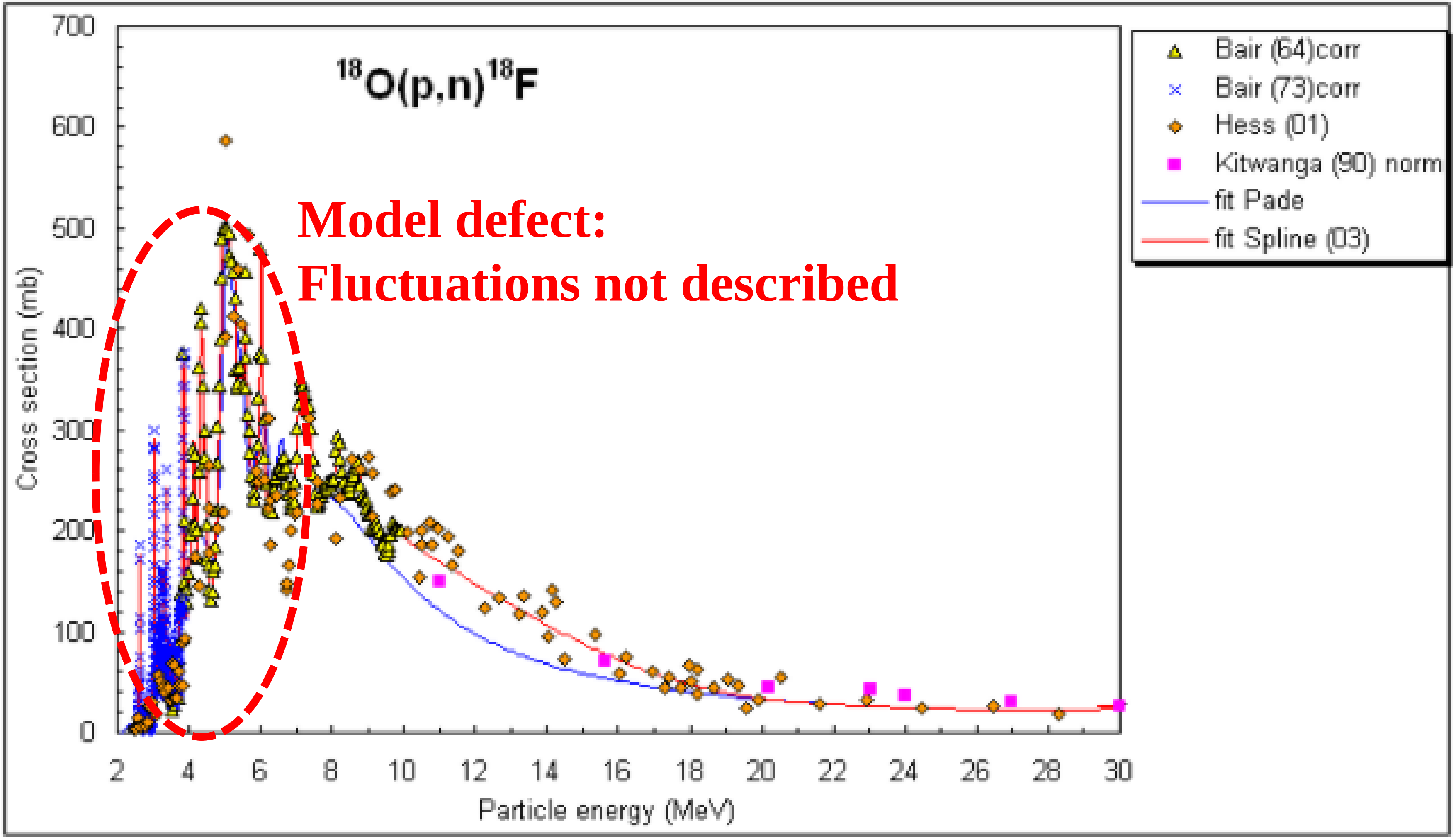
Monte Carlo calculation of covariance first tested by A. Koning

Model defects (I)



F-18 production in small cyclotrons

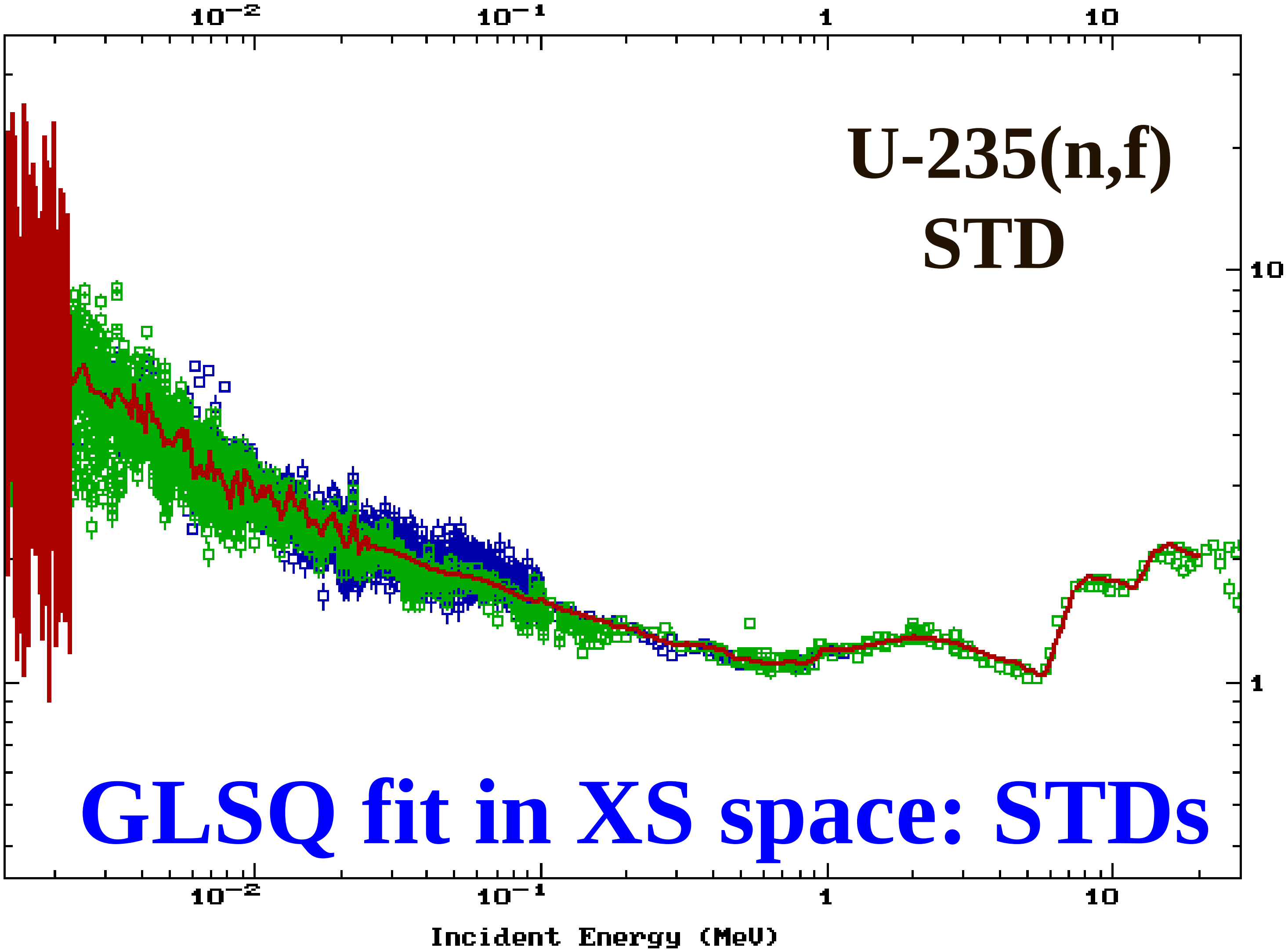
Selected experimental data for $^{18}O(p,n)^{18}F$ reaction



Model defect:
Fluctuations not described

Evaluation methods

“Non model” GLSQ fit : STDs



U-235(n,f)
STD

GLSQ fit in XS space: STDs

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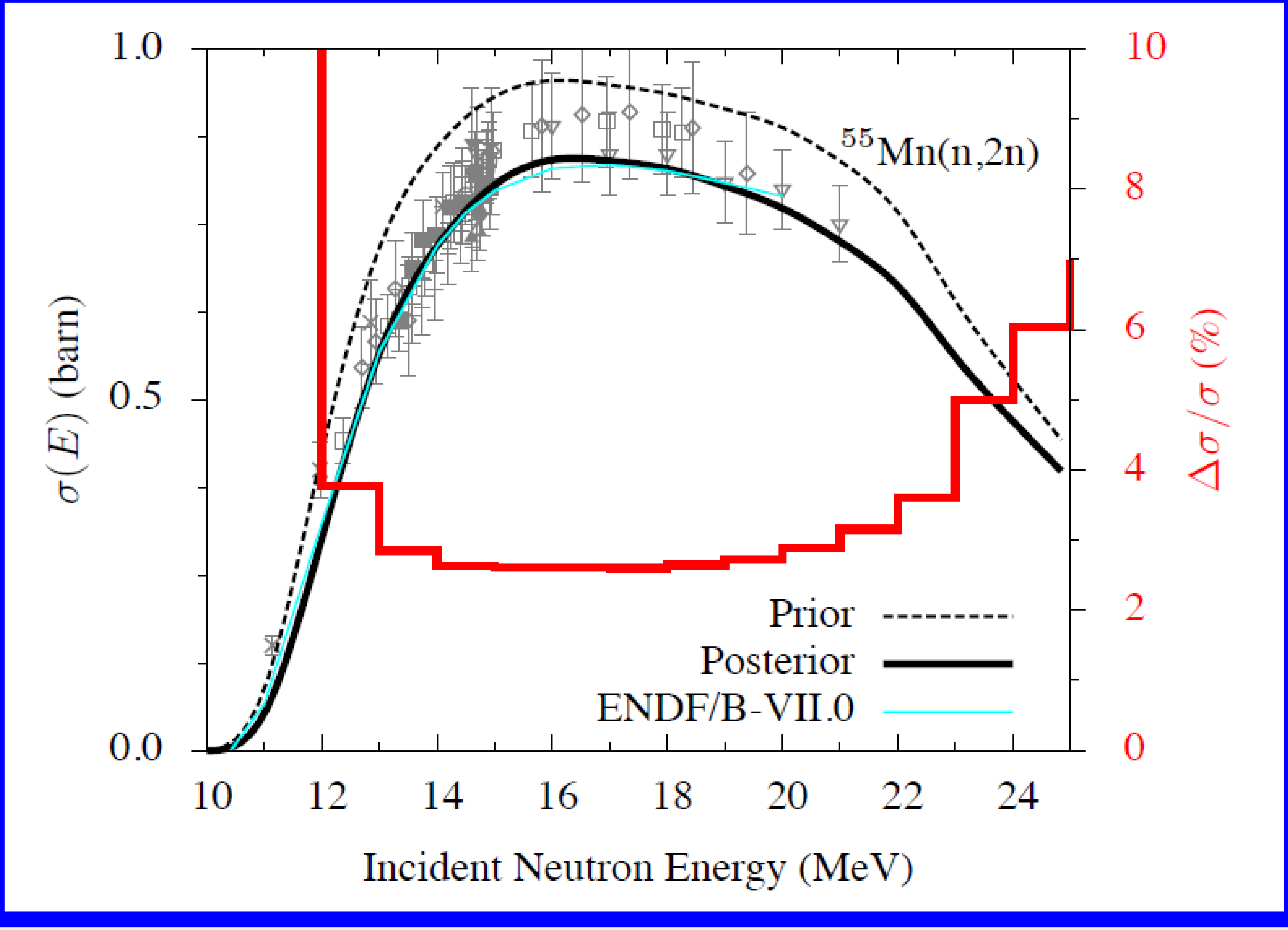
Typical situation (see above 21 MeV)



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Web: http://www-nds.iaea.org

Model prior +GLSQ fit (XS space)

Monte Carlo prior

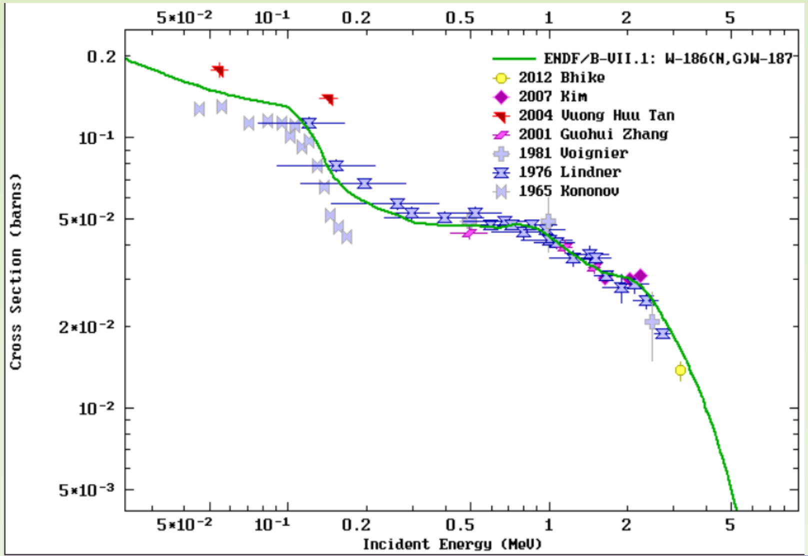
+

GANDR *(GLSQ)*

A. Trkov and R. Capote, “Cross-Section Covariance Data”, Th-232 evaluation for ENDF/B-VII.0 (MAT=9040 MF=1 MT=451); Pa-231 and Pa-233 evaluations for ENDF/B-VII.0 (MAT=9133 and 9137 MF=1 MT=451), National Nuclear Data Center, BNL (http://www.nndc.bnl.gov), 15 December 2006.

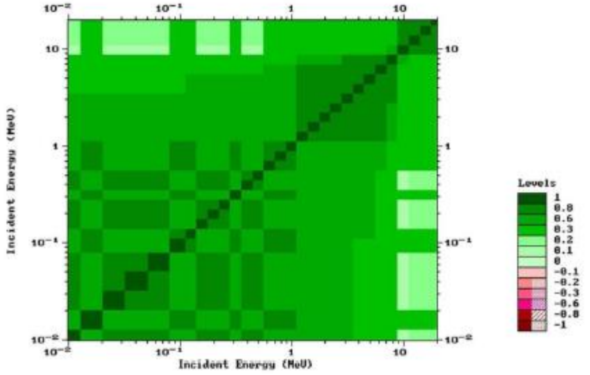
D.W. Muir, ***GANDR*** *project (IAEA)*, Online at www-nds.iaea.org/gandr/
GANDR includes a GUI

A.Trkov, RC, et al
Nuclear Data Sheets 112 (2011) 3098–3119



EVALUATION PHILOSOPHY: GANDR

MC MODEL cov. prior



Posterior cov. Prior incl. exp. data

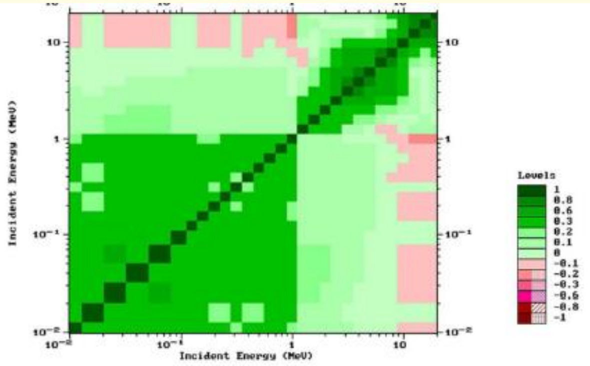
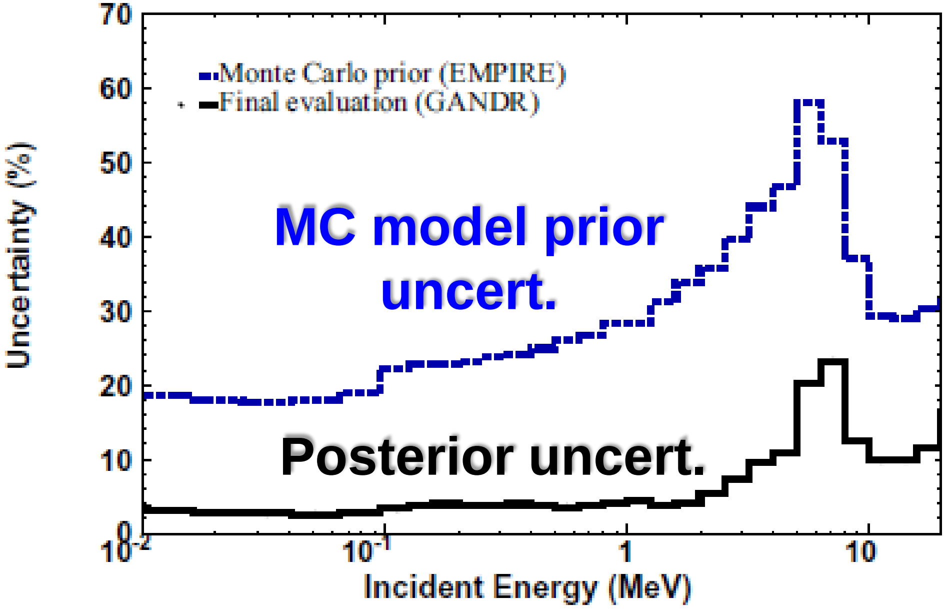


FIG. 2: Correlation matrix of the $^{186}W(n,\gamma)$ after the GANDR fit.



MC model prior uncert.

Posterior uncert.

FIG. 3: Uncertainty of the $^{186}W(n,\gamma)$ reaction in the fast neutron region: Monte Carlo calculated prior with EMPIRE (dashed line) is compared with the evaluation derived by GANDR result (solid line).

Evaluated uncertainties reduced due to lower exp. uncertainties AND strong model correlations

ND evaluation

Challenge #1: Decay data and model/data consistency

Decay data issues in activation experiments: Different DD values to deal with - DDEP vs ENSDF = f(t)

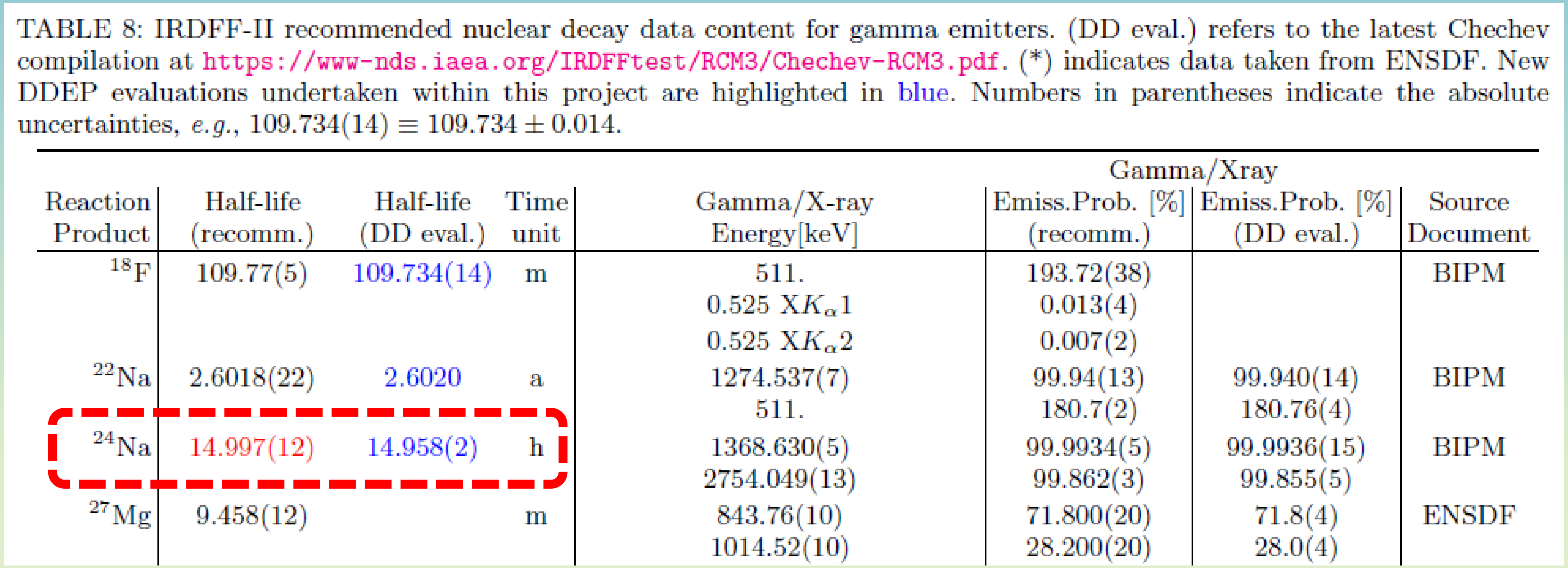


TABLE 8: IRDFF-II recommended nuclear decay data content for gamma emitters. (DD eval.) refers to the latest Chechev compilation at `https://www-nds.iaea.org/IRDFFtest/RCM3/Chechev-RCM3.pdf`. (*) indicates data taken from ENSDF. New DDEP evaluations undertaken within this project are highlighted in blue. Numbers in parentheses indicate the absolute uncertainties, *e.g.*, 109.734(14) ≡ 109.734 ± 0.014.

					Gamma/Xray		
Reaction Product	Half-life (recomm.)	Half-life (DD eval.)	Time unit	Gamma/X-ray Energy[keV]	Emiss.Prob. [%] (recomm.)	Emiss.Prob. [%] (DD eval.)	Source Document
^{18}F	109.77(5)	109.734(14)	m	511.	193.72(38)		BIPM
				0.525 $XK_{\alpha}1$	0.013(4)		
				0.525 $XK_{\alpha}2$	0.007(2)		
^{22}Na	2.6018(22)	2.6020	a	1274.537(7)	99.94(13)	99.940(14)	BIPM
				511.	180.7(2)	180.76(4)	
^{24}Na	14.997(12)	14.958(2)	h	1368.630(5)	99.9934(5)	99.9936(15)	BIPM
				2754.049(13)	99.862(3)	99.855(5)	
^{27}Mg	9.458(12)		m	843.76(10)	71.800(20)	71.8(4)	ENSDF
				1014.52(10)	28.200(20)	28.0(4)	

$T_{1/2}$=14.997 (12) h –R. Firestone et al, Nucl. Data Sheets 108 (2007) 2317

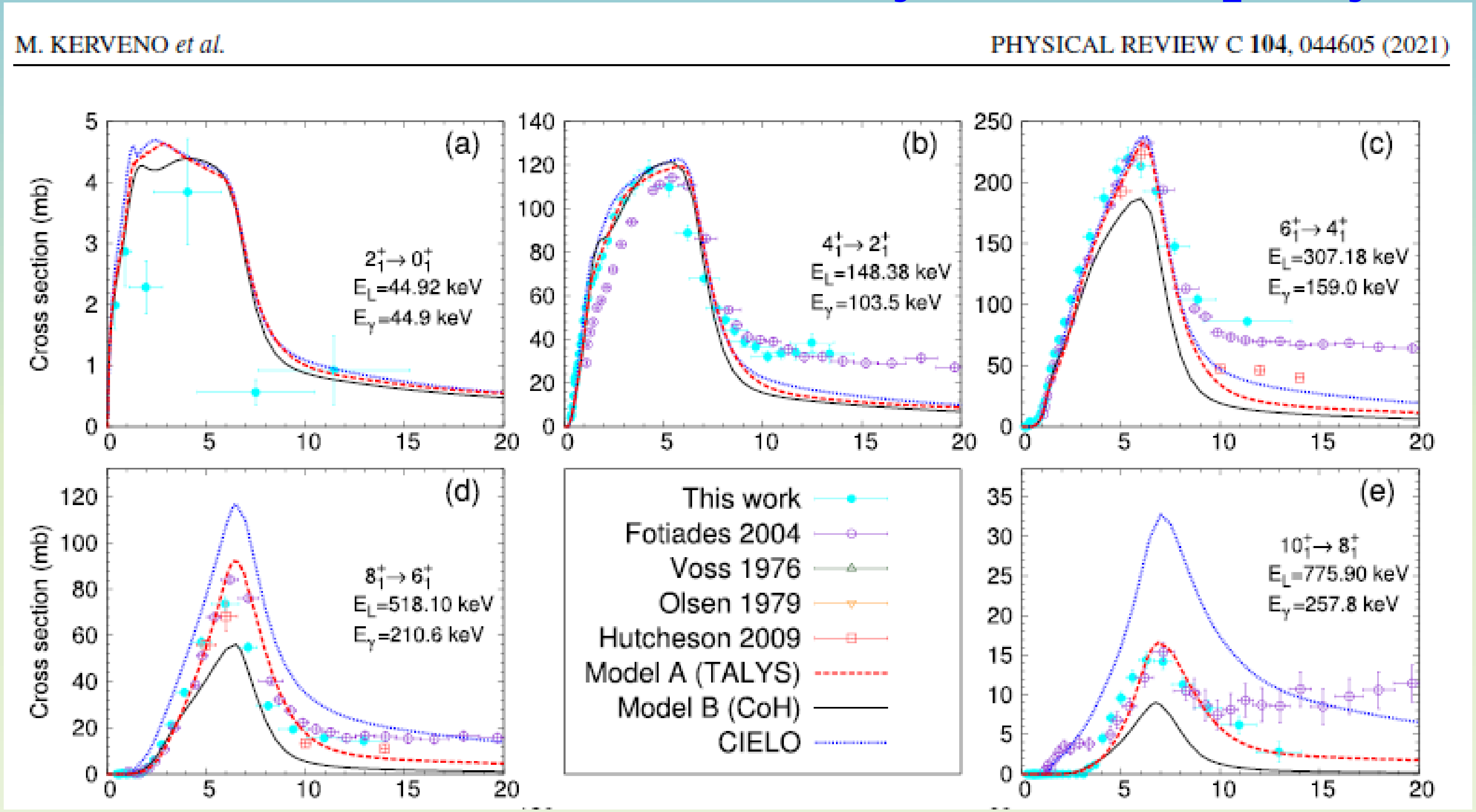
$T_{1/2}$=14.958 (2) h – V.P. Chechev, N.K. Kuzminov, 06/2014,

IAEA IRDFF evaluation adopted for DDEP.

✓ **~~From users: A long standing issue, we need time stamp and archive !~~**

A. Trkov et al, IRDFF-II: A new neutron metrology library
Nuclear Data Sheets 163, 1-111 (2020); arXiv 1909.03336 (2019)

Experimental data statistical consistency
Reaction model and decay data adequacy



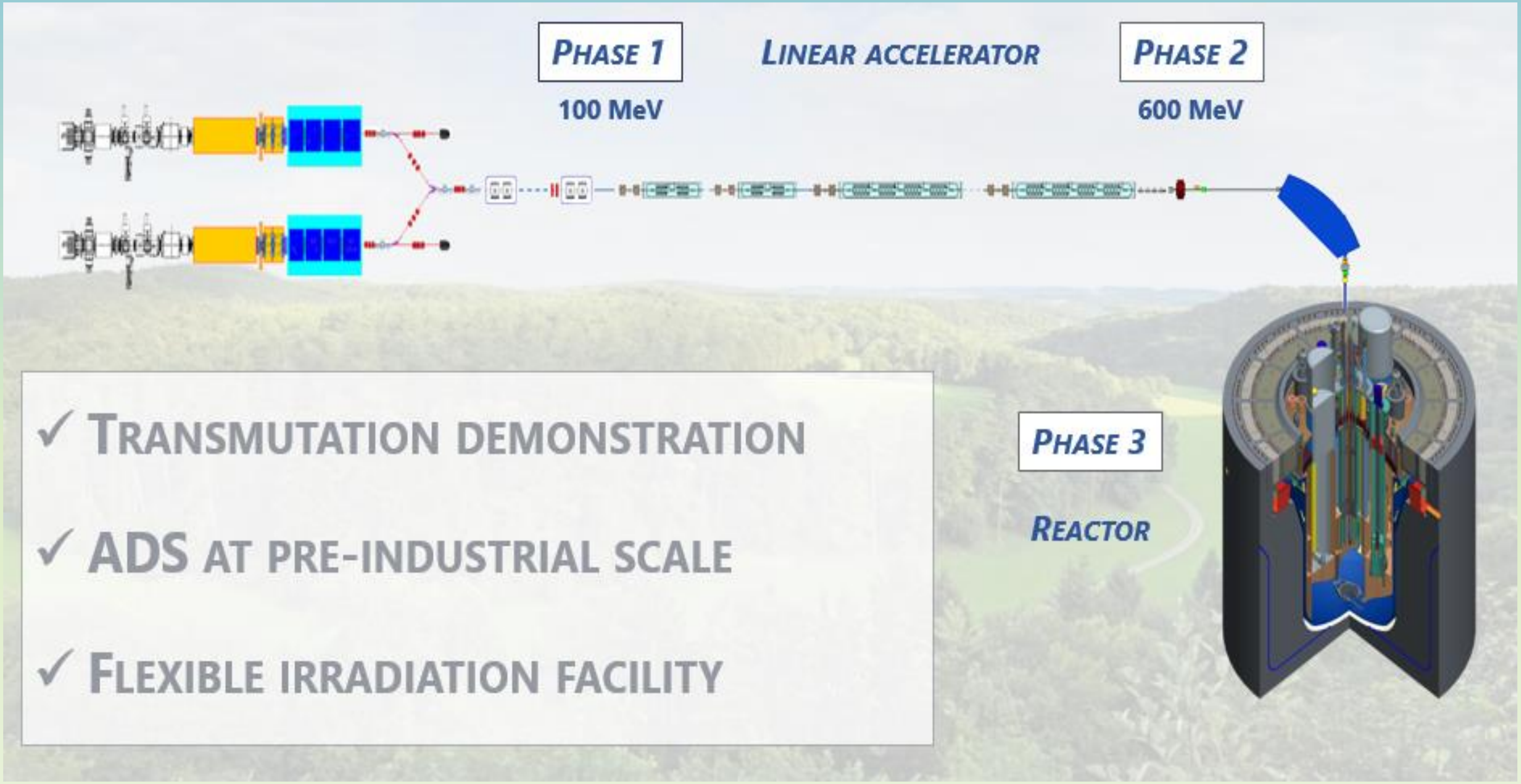
M. KERVENO *et al.* PHYSICAL REVIEW C **104**, 044605 (2021)

Measurement of ^{238}U(n, $n'\gamma$) cross section data and their impact on reaction models

ND evaluation

Challenge #2:
Pu-239 needs for ADS

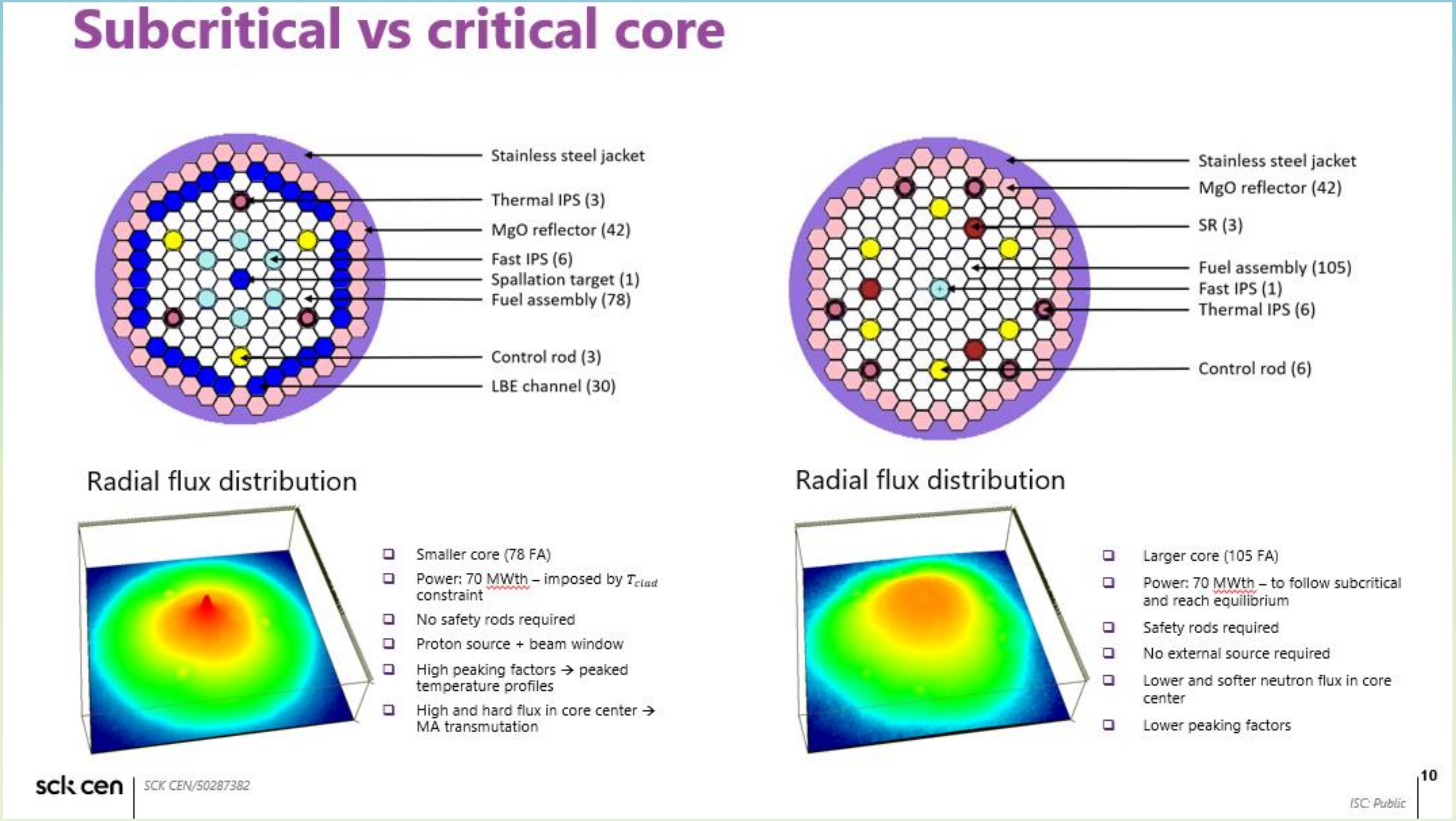
MYRRHA: accel. driven system



- ✓ TRANSMUTATION DEMONSTRATION
- ✓ ADS AT PRE-INDUSTRIAL SCALE
- ✓ FLEXIBLE IRRADIATION FACILITY

**Taken from A. Stankovsky, SCK CEN
@IAEA TM INDEN, 08/2022**

MYRRHA reactor



Subcritical vs critical core

Stainless steel jacket
Thermal IPS (3)
MgO reflector (42)
Fast IPS (6)
Spallation target (1)
Fuel assembly (78)
Control rod (3)
LBE channel (30)

Stainless steel jacket
MgO reflector (42)
SR (3)
Fuel assembly (105)
Fast IPS (1)
Thermal IPS (6)
Control rod (6)

Radial flux distribution

- ☐ Smaller core (78 FA)
- ☐ Power: 70 MWth – imposed by T_{clad} constraint
- ☐ No safety rods required
- ☐ Proton source + beam window
- ☐ High peaking factors → peaked temperature profiles
- ☐ High and hard flux in core center → MA transmutation

Radial flux distribution

- ☐ Larger core (105 FA)
- ☐ Power: 70 MWth – to follow subcritical and reach equilibrium
- ☐ Safety rods required
- ☐ No external source required
- ☐ Lower and softer neutron flux in core center
- ☐ Lower peaking factors

sck cen | SCK CEN/50287382

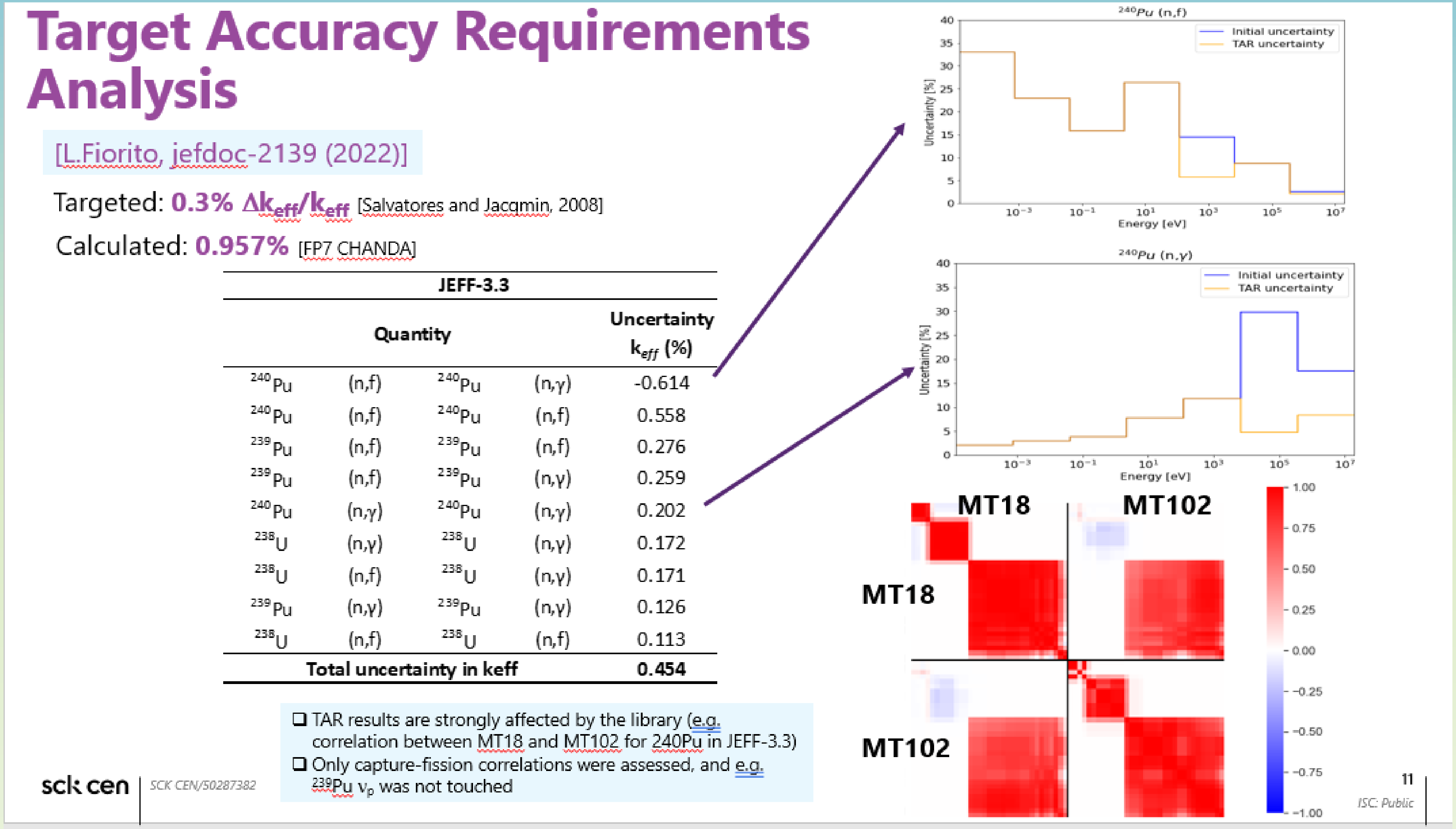
10

ISC: Public

^{239,240}Pu, Mg, O, SS (Fe)!!

Taken from A. Stankovsky, SCK CEN @IAEA TM INDEN, 08/2022

MYRRHA reactor



Target Accuracy Requirements Analysis

[L.Fiorito, jefdoc-2139 (2022)]

Targeted: **0.3% $\Delta k_{eff}/k_{eff}$** [Salvatores and Jacqmin, 2008]

Calculated: **0.957%** [FP7 CHANDA]

JEFF-3.3				
Quantity				**Uncertainty k_{eff} (%)**
^{240}Pu	(n,f)	^{240}Pu	(n,γ)	-0.614
^{240}Pu	(n,f)	^{240}Pu	(n,f)	0.558
^{239}Pu	(n,f)	^{239}Pu	(n,f)	0.276
^{239}Pu	(n,f)	^{239}Pu	(n,γ)	0.259
^{240}Pu	(n,γ)	^{240}Pu	(n,γ)	0.202
^{238}U	(n,γ)	^{238}U	(n,γ)	0.172
^{238}U	(n,f)	^{238}U	(n,γ)	0.171
^{239}Pu	(n,γ)	^{239}Pu	(n,γ)	0.126
^{238}U	(n,f)	^{238}U	(n,f)	0.113
Total uncertainty in keff				**0.454**

- ☐ TAR results are strongly affected by the library (e.g. correlation between MT18 and MT102 for 240Pu in JEFF-3.3)
- ☐ Only capture-fission correlations were assessed, and e.g. ^{239}Pu ν_p was not touched

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11

ISC: Public

^{239,240}Pu, Mg, O, SS (Fe)!!

Taken from A. Stankovsky, SCK CEN
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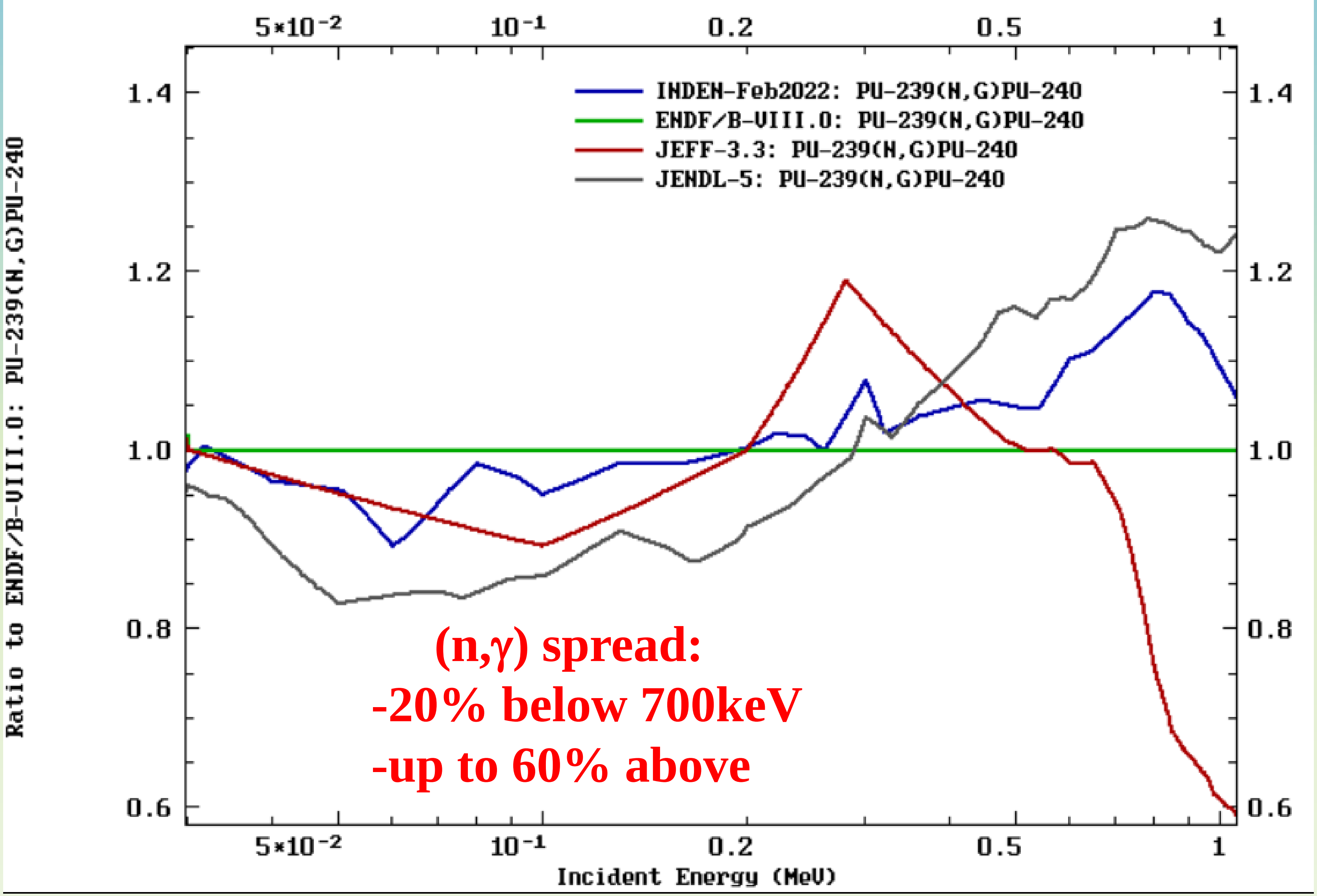
^{239}Pu(n,γ) overview in libraries



(n,γ) spread:

-20% below 700keV

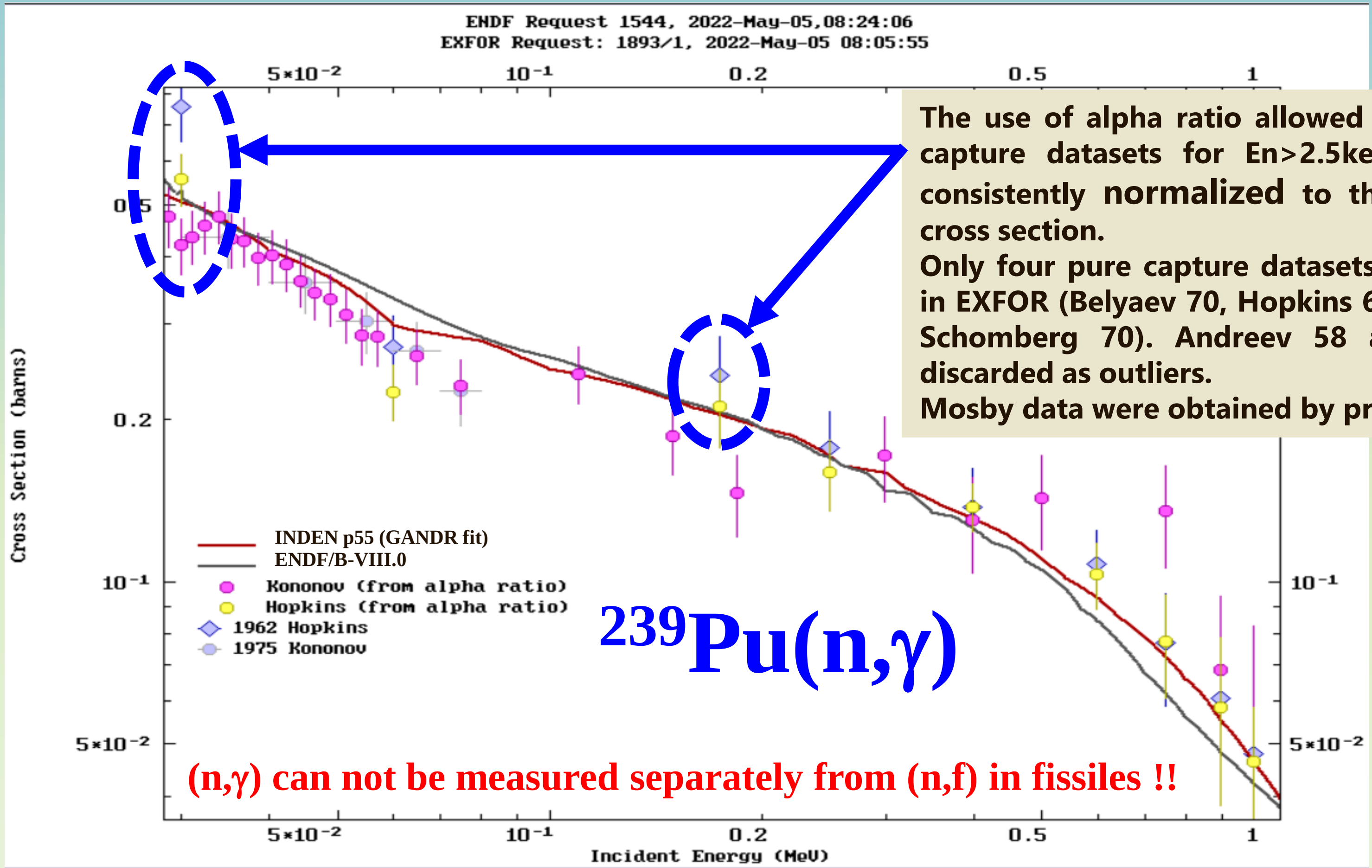
-up to 60% above

INDEN Pu-239 evaluation: (n,γ) in the fast region

- 11 capture datasets derived from alpha=σ_c/σ_f data using the latest Neutron Standard (n,f) cross sections.
- New capture evaluation is based on a GANDR generalized least squares fit of experimental data. Empire model calculation with large uncertainties is used as a model prior in GANDR fit.
- We find significant differences with existing evaluations (!?)
- Uncertainty of the GLSQ (GANDR) evaluated capture cross sections is reduced by ~40% compared to the ENDF/B-VIII.0 assessed uncertainty

Importance of σ_c/σ_f ratio data:

Renormalization to updated fission standard XS



The use of alpha ratio allowed to generate 11 capture datasets for En>2.5keV, which were consistently **normalized** to the same fission cross section.

Only four pure capture datasets were available in EXFOR (Belyaev 70, Hopkins 62, Kononov 75, Schomberg 70). Andreev 58 and Spivak 56 discarded as outliers.

Mosby data were obtained by priv. comm.

^{239}Pu(n,γ)

(n,γ) can not be measured separately from (n,f) in fissiles !!

Derived from α ratio Hopkins shows improved agreement with evals (e.g., @0.03MeV)

Kononov 75 data derived from α ratio contains many more points than capture data

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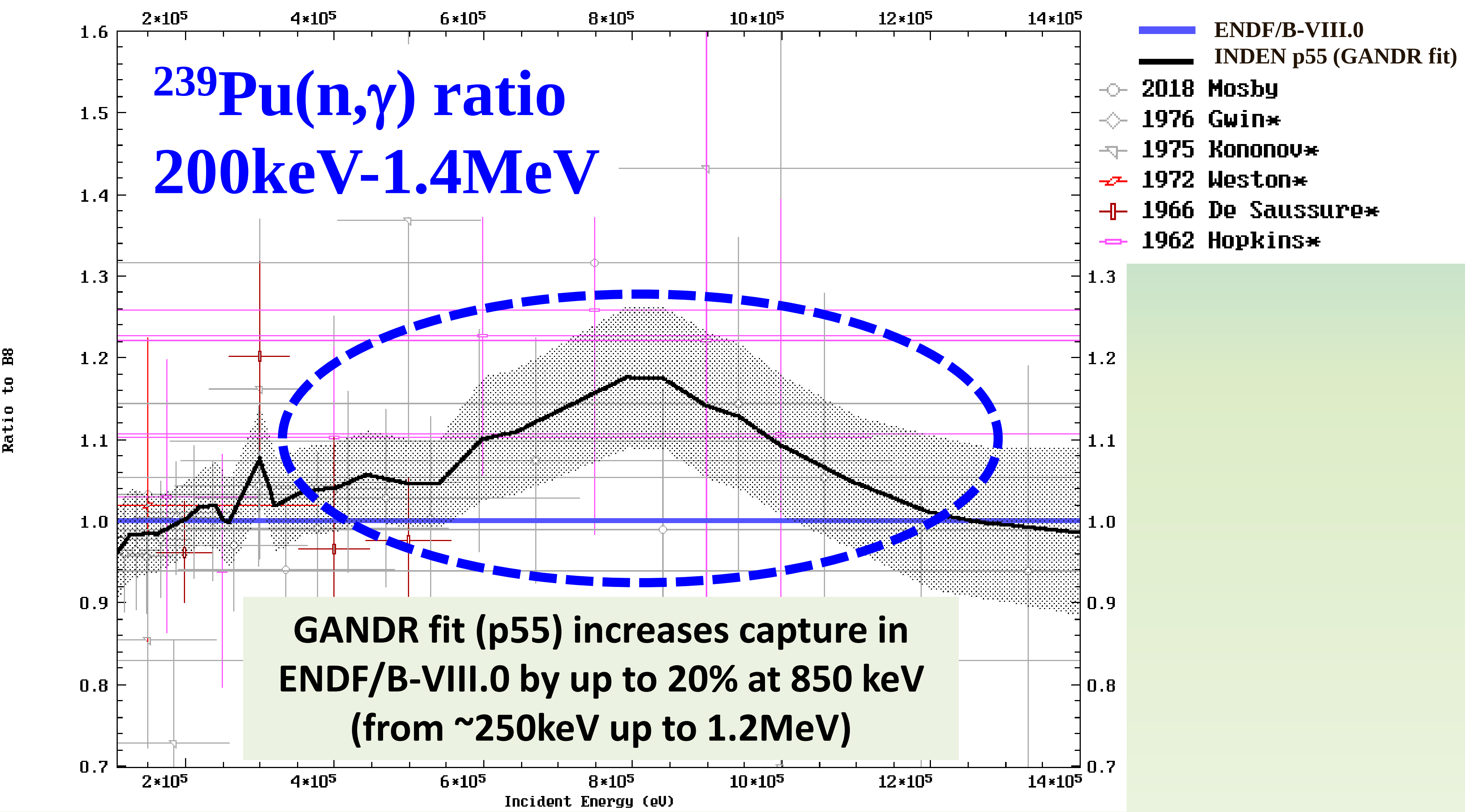
(n,γ) exper. data vs GANDR fit (p55) vs ENDF/B-VIII.0

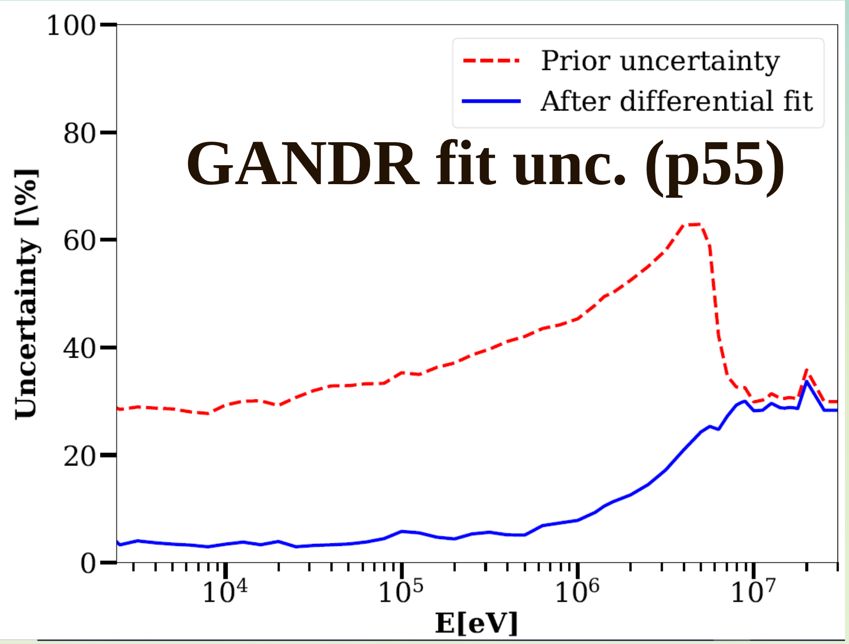


GANDR fit (p55) increases capture in ENDF/B-VIII.0 by up to 20% at 850 keV (from ~250keV up to 1.2MeV)

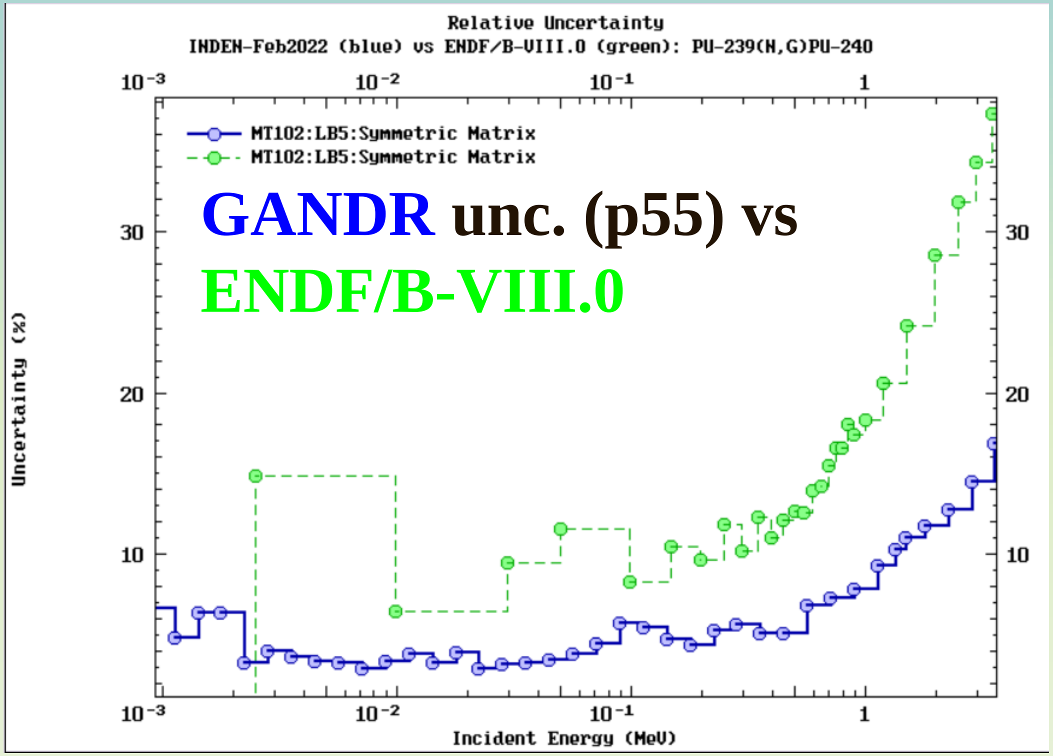
^{239}Pu(n,γ) GANDR fit uncertainty

Uncertainty 0.1-1 MeV ~ 5-8%

Significant uncertainty reduction from ENDF/B-VIII.0



GANDR fit unc. (p55)



GANDR unc. (p55) vs ENDF/B-VIII.0

INDEN p55 uncertainties = GANDR fit uncertainties

Comprehensive data reduces the need of models !

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ND evaluation

Challenge #3:
Capture and inelastic gammas

PROBLEMS IN EVALUATED GAMMA SPECTRA

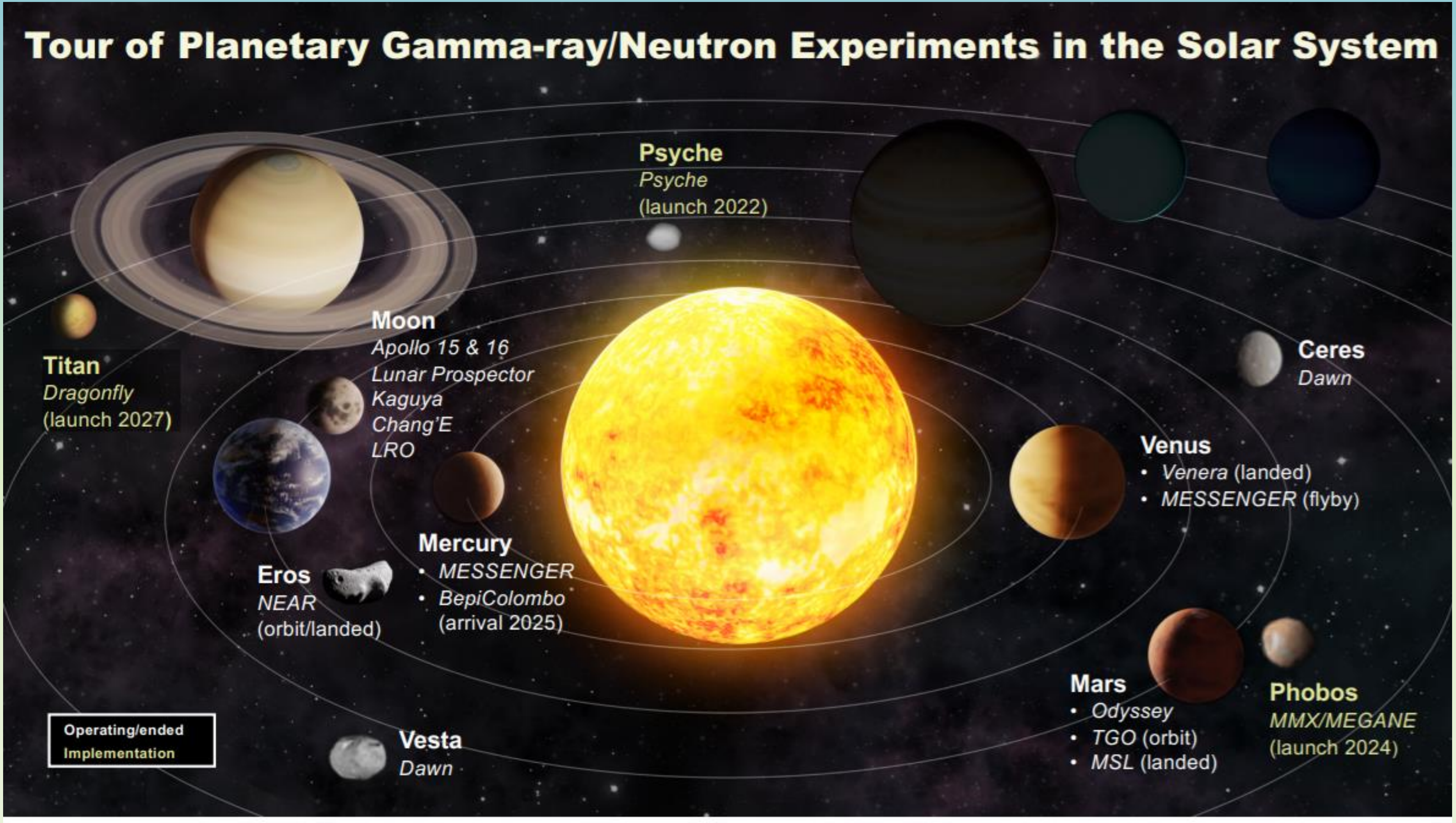
Nuclear data needs of gamma spectra (D. Lawrence & P. Peplowski)

- ☐ Active neutron interrogation (C. Romano et al)
- ☐ Non-proliferation applications (D. Matters)
- ☐ Planetary science (T. Prettyman et al)
- ☐ Subsurface exploration (M.-L.Mauborgne et al)
- ☐ Shielding and criticality safety (Miller et al.)

see WANDA 2021 presentations at:

https://conferences.lbl.gov/event/504/

ND needs in planetary gamma spectroscopy



Taken from D. Lawrence and P. Peplowski, @WANDA2021

Problems identified in inelastic/capture gammas of many ENDF/B-VIII.0 evaluations

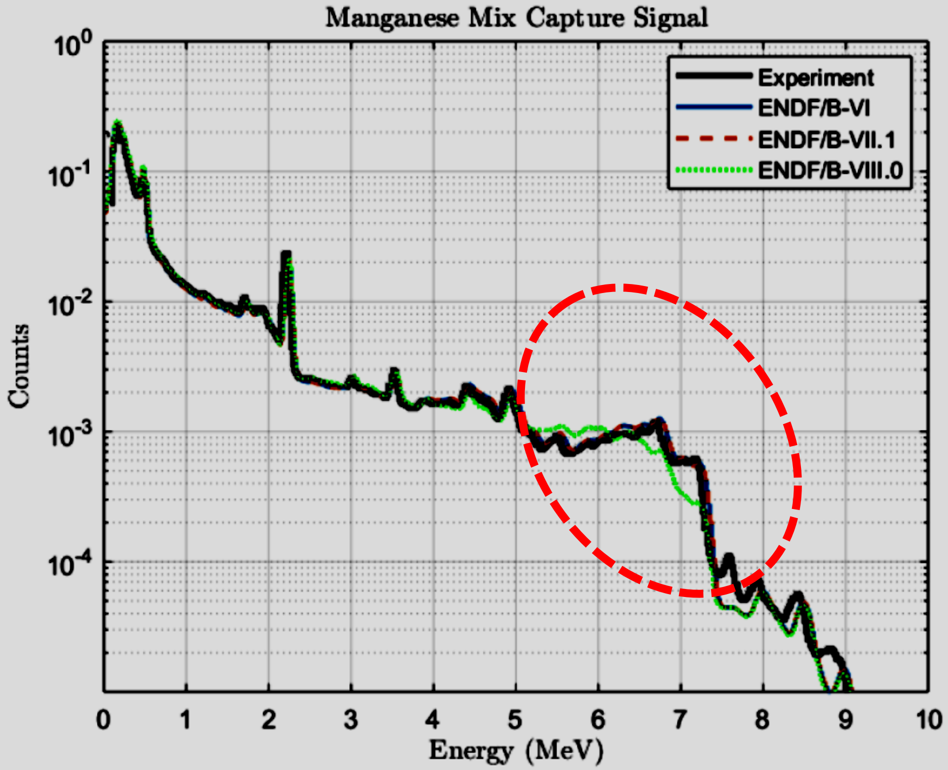


Fig. 9: Comparison of capture γ-ray spectra from the manganese mix from experiment and modeled with various libraries

Similar problems:
Si(n,n'γ),
Fe(n,γ), Fe(n,n'γ),
Mg(n,γ), Mg(n,n'γ),
Ti(n,γ), Ti(n,n'γ)

Hint:
ENDF/B-VI.8
was better for gammas

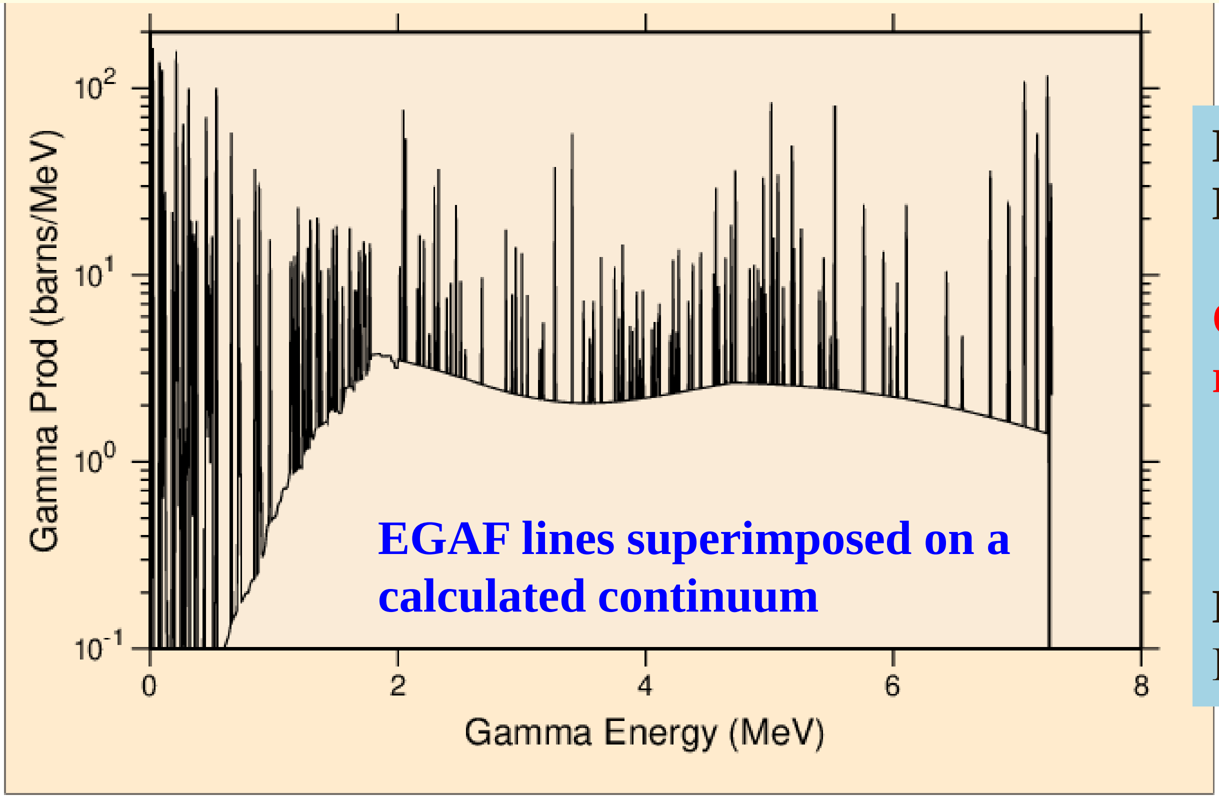
Marie-Laure Mauborgne et al, CSWEG 2019 & EPJ WoC **239** (2020) 20007 (ND2019)

^{55}Mn update of thermal (n,g) gammas

- ☐ Mn-55 evaluation in ENDF/B-VIII.0 was criticized for poor prediction of capture gamma spectra (Marie-Laure Mauborgne, CSEWG-2019, EPJ WoC **239**, 20007 (2020).
- ☐ The data are important for oil-well exploration.
- ☐ Using the information in the EGAF library and EMPIRE nuclear model calculations the gamma production data were improved. High resolution energy bins (~5 keV/bin)
- ☐ Good performance of updated file **mn55e80p** on proprietary benchmark was confirmed by Marie-Laure Mauborgne
- ☐ Documented in INDC(NDS)-0810 on "Evaluation of thermal capture gamma spectra"

^{55}Mn update of thermal (n,γ) gammas

Thermal capture photon spectrum updated



EGAF lines superimposed on a calculated continuum

EGAF IAEA database:
Measured thermal capture γ

Quite challenging to reproduce via modelling

⬇

But very well measured ☺, Let’s use it.

See description in INDC(NDS)-0810, performance restored

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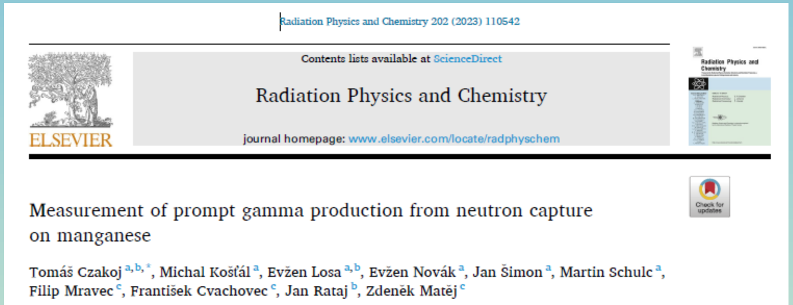
Validation of INDEN Mn-55



Radiation Physics and Chemistry 202 (2023) 110542

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

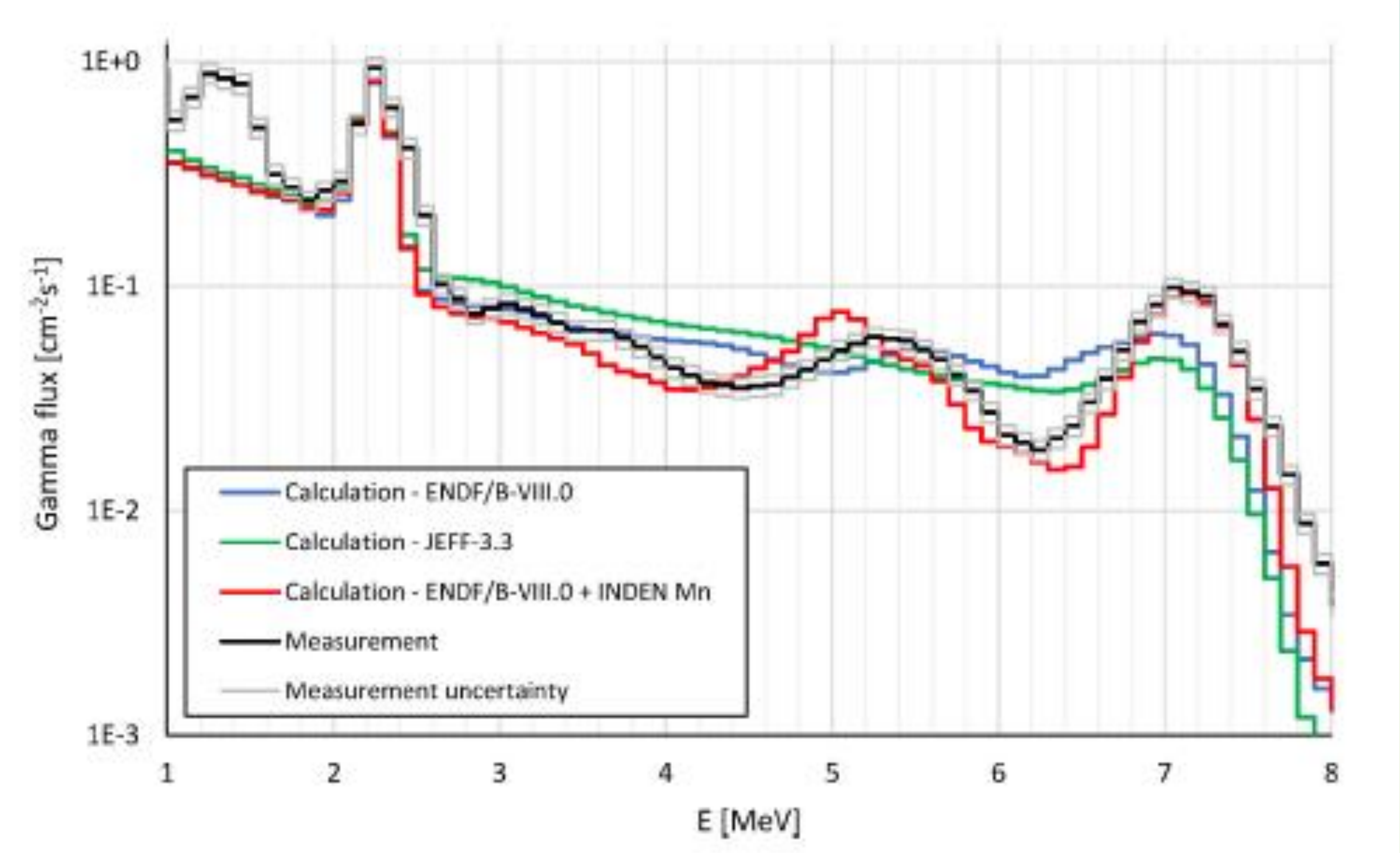
journal homepage: www.elsevier.com/locate/radphyschem

Measurement of prompt gamma production from neutron capture on manganese

Tomáš Czakoj [a,b,*], Michal Košťál [a], Evžen Losa [a,b], Evžen Novák [a], Jan Šimon [a], Martin Schulc [a],
Filip Mravec [c], František Cvachovec [c], Jan Rataj [b], Zdeněk Matěj [c]

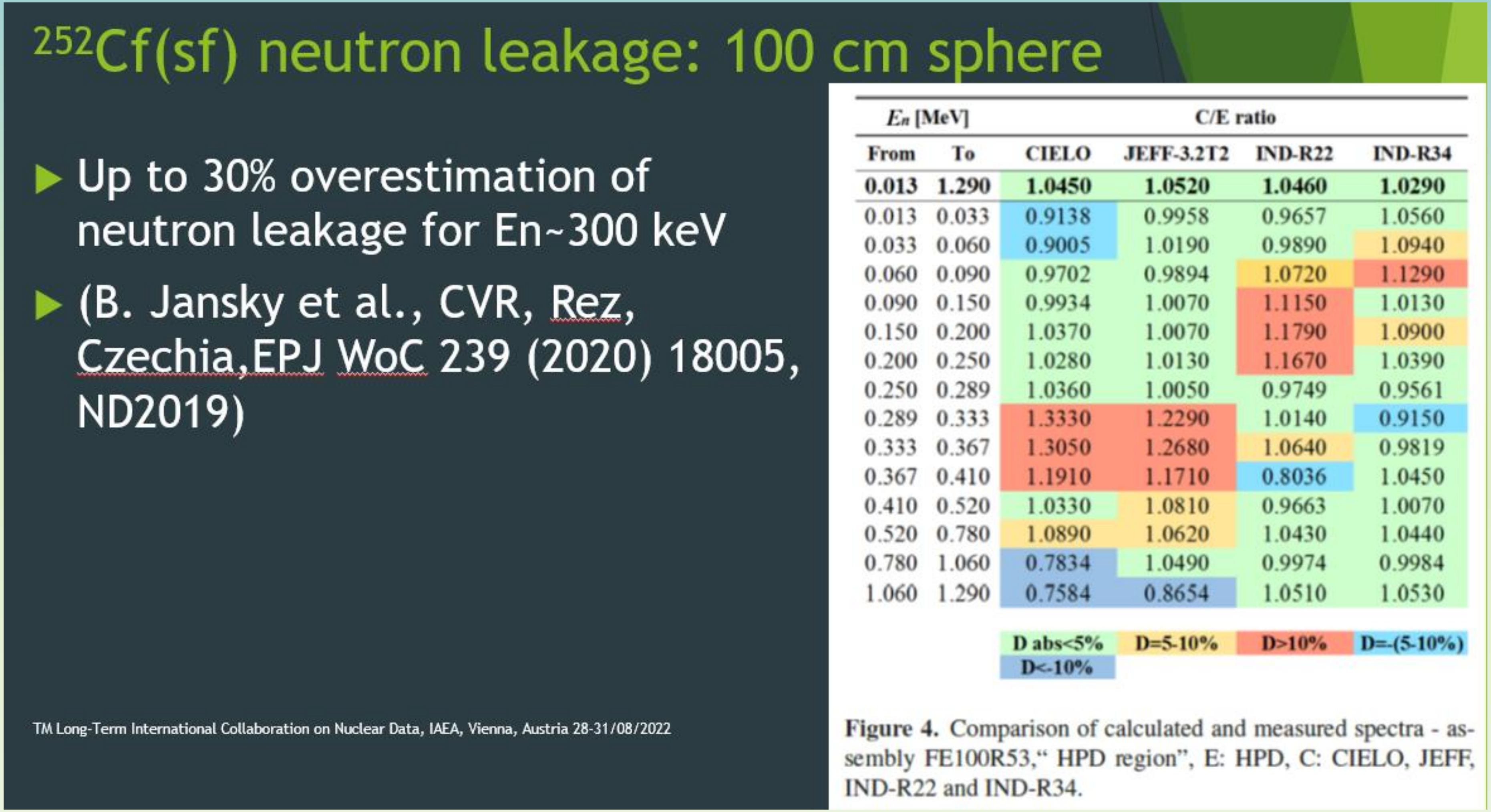
INDEN Mn-55:
INDC(NDS)-810

https://www-nds.iaea.org/publications/indc/indc-nds-0810.pdf



Challenge #3:
How to use validation experiments and evaluations?

Iron ND problem identified in Cf-neutron leakage measurement (int. experiment)



^{252}Cf(sf) neutron leakage: 100 cm sphere

- Up to 30% overestimation of neutron leakage for En~300 keV
- (B. Jansky et al., CVR, Rez, Czechia,EPJ WoC 239 (2020) 18005, ND2019)

En [MeV]		C/E ratio			
From	To	CIELO	JEFF-3.2T2	IND-R22	IND-R34
0.013	**1.290**	**1.0450**	**1.0520**	**1.0460**	**1.0290**
0.013	0.033	0.9138	0.9958	0.9657	1.0560
0.033	0.060	0.9005	1.0190	0.9890	1.0940
0.060	0.090	0.9702	0.9894	1.0720	1.1290
0.090	0.150	0.9934	1.0070	1.1150	1.0130
0.150	0.200	1.0370	1.0070	1.1790	1.0900
0.200	0.250	1.0280	1.0130	1.1670	1.0390
0.250	0.289	1.0360	1.0050	0.9749	0.9561
0.289	0.333	1.3330	1.2290	1.0140	0.9150
0.333	0.367	1.3050	1.2680	1.0640	0.9819
0.367	0.410	1.1910	1.1710	0.8036	1.0450
0.410	0.520	1.0330	1.0810	0.9663	1.0070
0.520	0.780	1.0890	1.0620	1.0430	1.0440
0.780	1.060	0.7834	1.0490	0.9974	0.9984
1.060	1.290	0.7584	0.8654	1.0510	1.0530

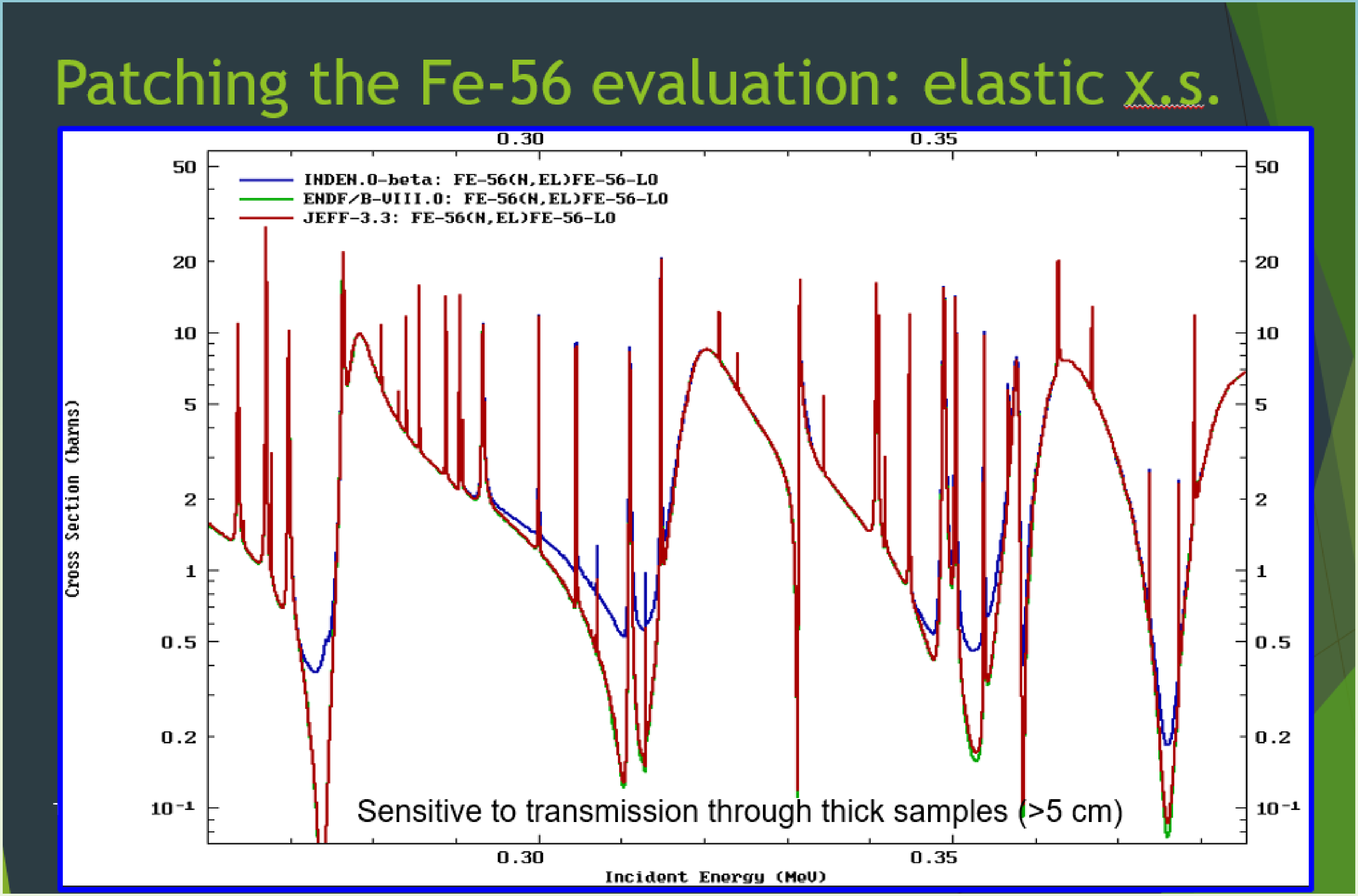
D abs<5% **D=5-10%** **D>10%** **D=-(5-10%)**
D<-10%

Figure 4. Comparison of calculated and measured spectra - assembly FE100R53,“ HPD region”, E: HPD, C: CIELO, JEFF, IND-R22 and IND-R34.

TM Long-Term International Collaboration on Nuclear Data, IAEA, Vienna, Austria 28-31/08/2022

The case of iron evaluation

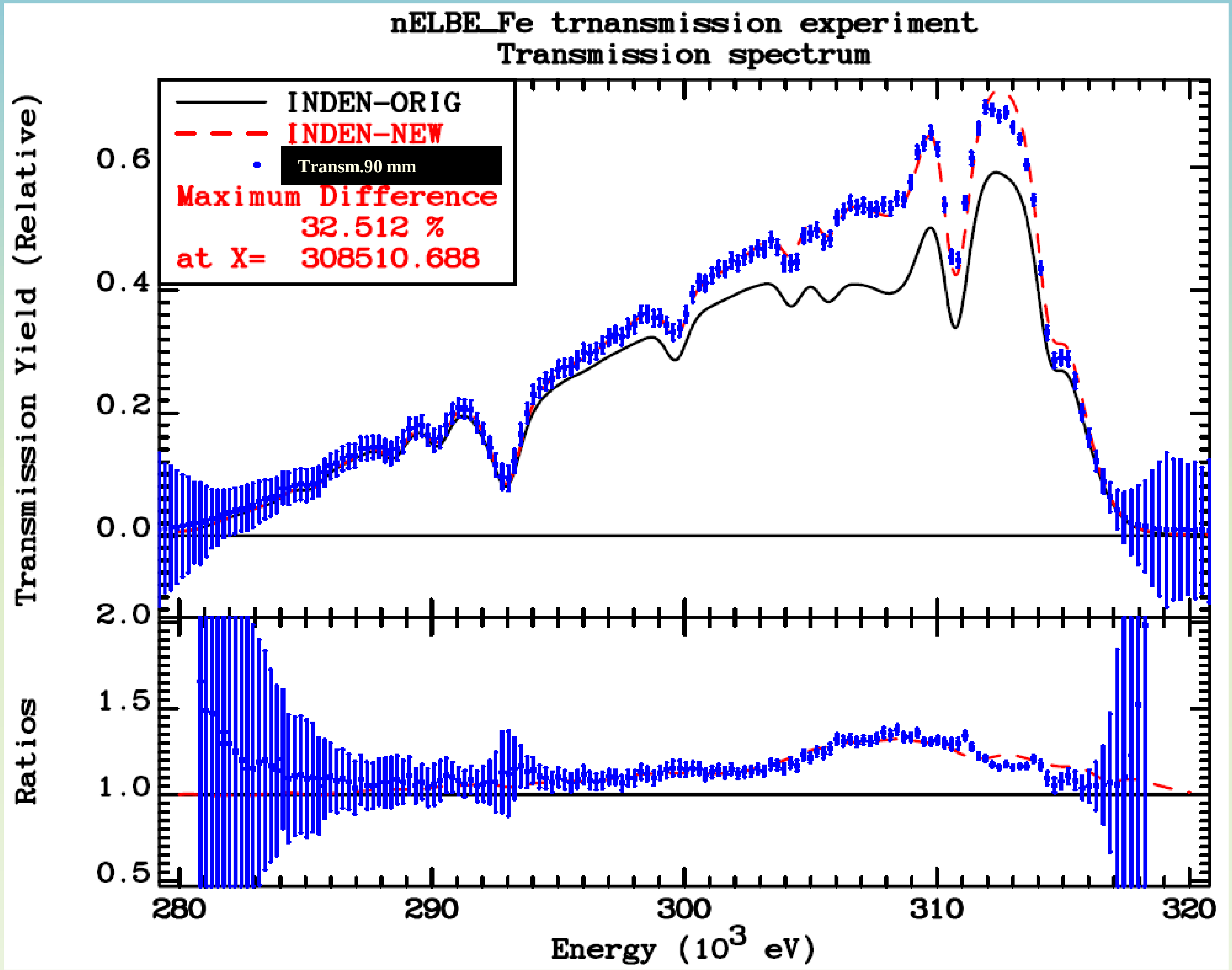
Solution proposed, ^{56}Fe eval. updated



Patching the Fe-56 evaluation: elastic x.s.

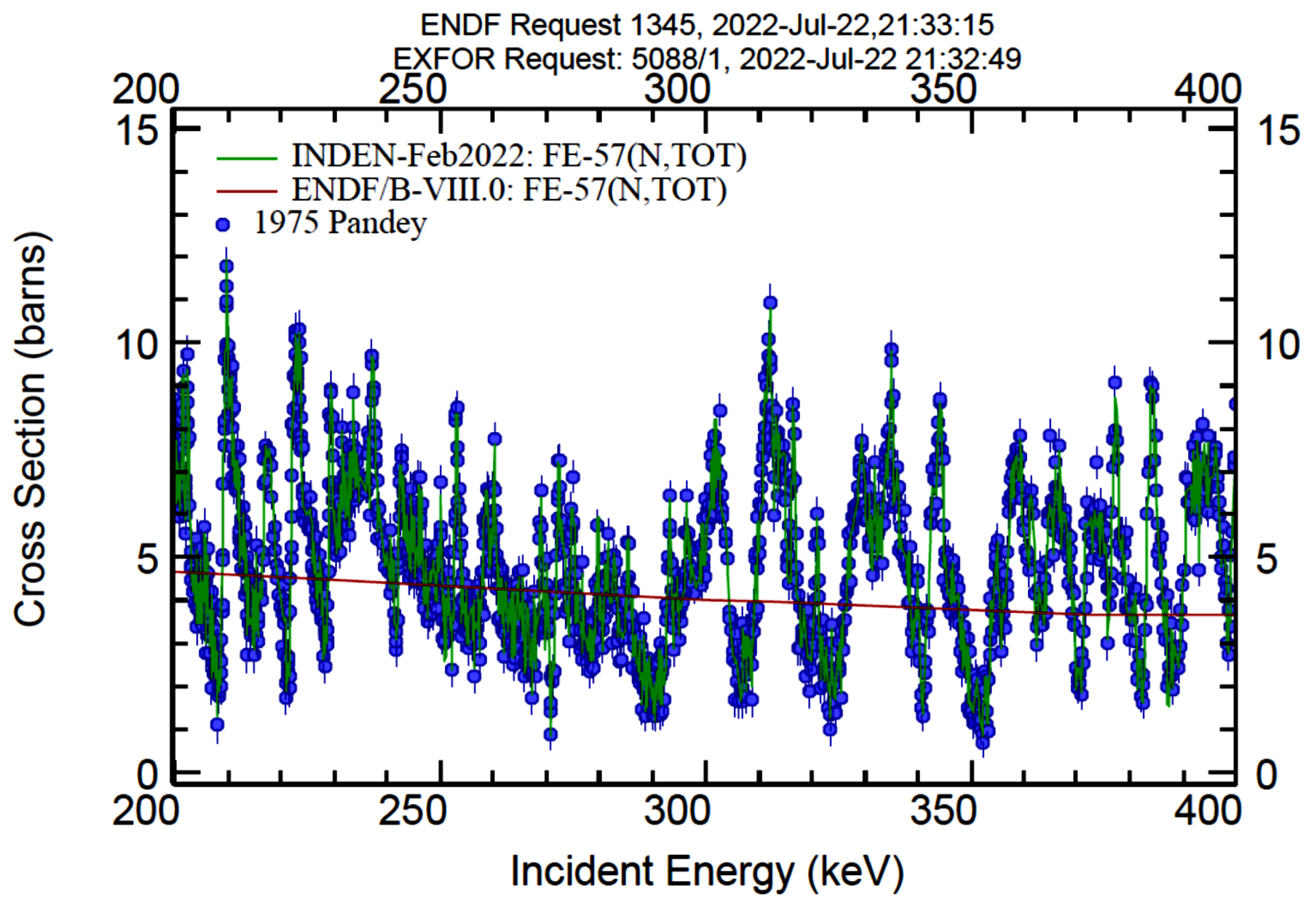
Sensitive to transmission through thick samples (>5 cm)

9cm target transmission measur. at nELBE



Experiment showed the patch to be wrong (black line)

Real problem found in RR of Fe-57: Evaluation corrected



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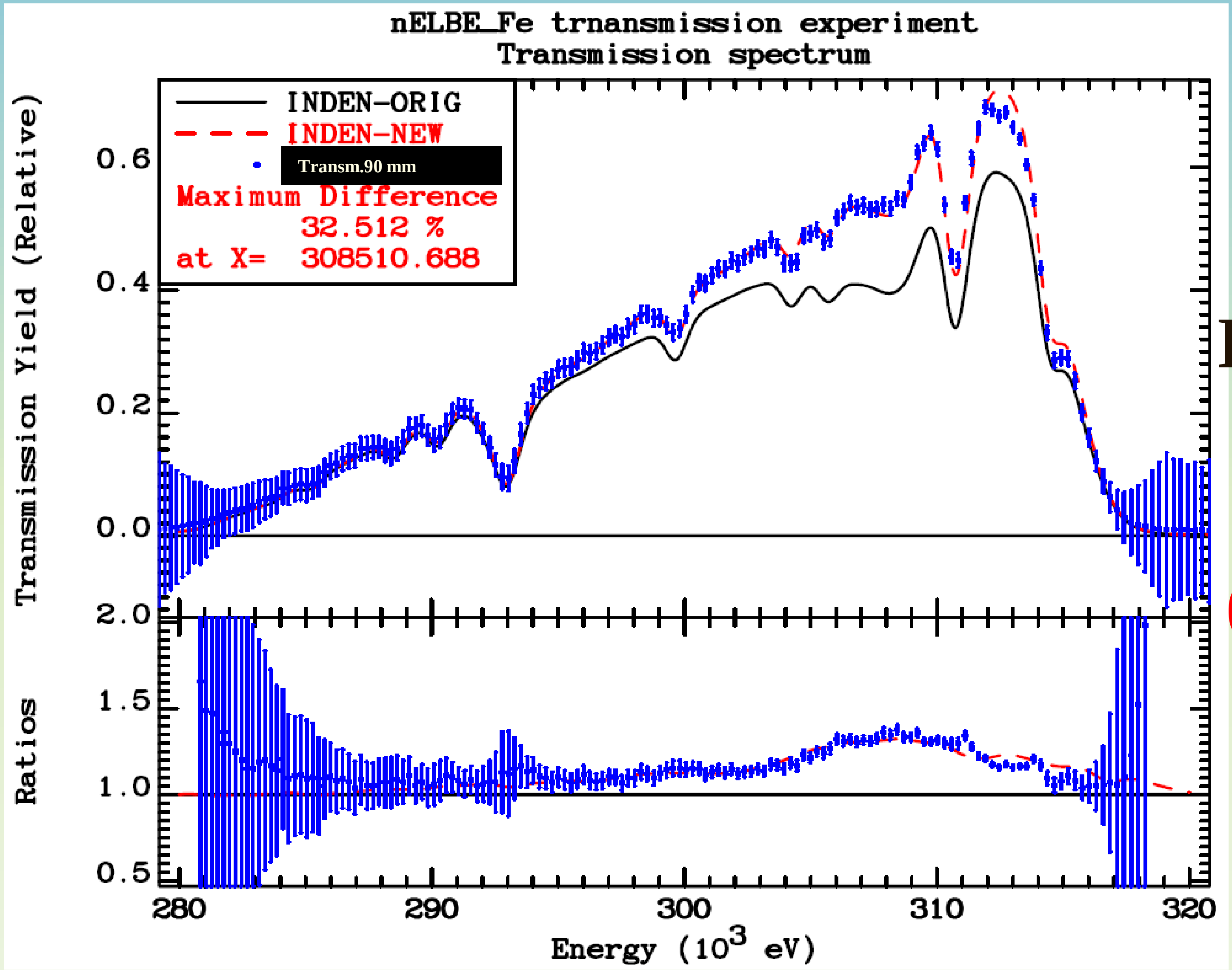
Thick target transmission measur. at nELBE



Fe-57 update solved the issue (dashed red)

Summary

- A non-comprehensive (and biased) selection of nuclear data challenges have been presented as a motivation for nuclear data research
- Applications like innovative reactors, planetary exploration, geology and oil logging have been used to highlight associated nuclear data needs
- Examples on how to address some of these challenges in Pu, Mn, and Fe were shown

Take home message

Evaluation: A properly weighted combination (usually by GLSQ fit) of selected experimental data (and nuclear reaction modelling results).

Bayesian approaches

❑ “Non-model” GLSQ fit (standards)
❑ Model prior + GLSQ fit (working horse)
❑ Other Bayesian methods

Experimental data analysis and extended UQ

(templates) are a critical step