Wonder 2018

Summation Calculations for Reactor Antineutrino Spectra, Decay Heat and Delayed Neutron Fractions Involving New TAGS Data and Evaluated Databases

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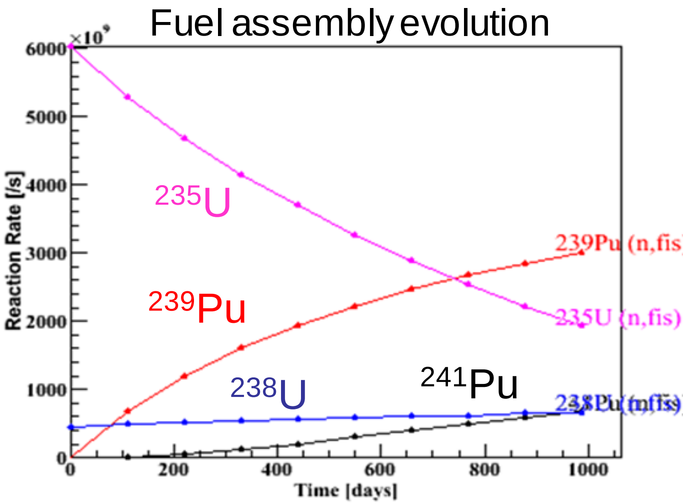
Aix-en-Provence - October 8.-12. 2018

Outline

- General Physics Motivations & Experimental Issues
- Reactor Antineutrinos
- Decay Heat
- Delayed Neutrons
- Conclusions and Perspectives

General Physics Motivation & Experimental Issues

Reactors and Beta Decay


Fuel assembly evolution

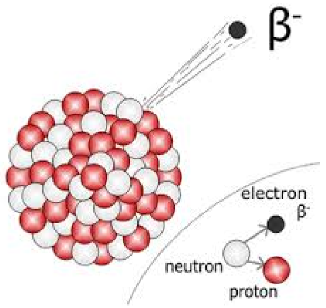
- In Pressurized Water Reactors, thermal power mainly induced by 4 isotopes:
  - ❑ Burn-up effect => unit GWd/t
- Fission process gives thermal energy:

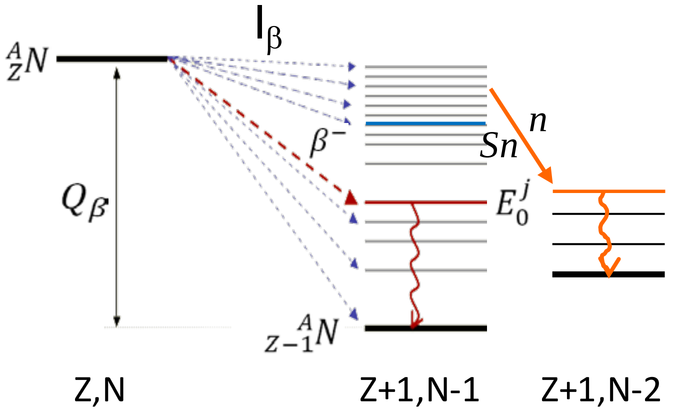
$$n + {}^{235}U \rightarrow {}^{236}U^{*} \rightarrow \mathrm{FP1} + \mathrm{FP2} + \mathrm{neutrons}\,(200MeV)$$

- The fission products (FP) after the fissions are neutron-rich nuclei undergoing β and β-n decays:

$${}^{A}_{Z}X \rightarrow {}^{A}_{Z+1}Y^{*} + e^{-} + \overline{\nu}_{e}$$

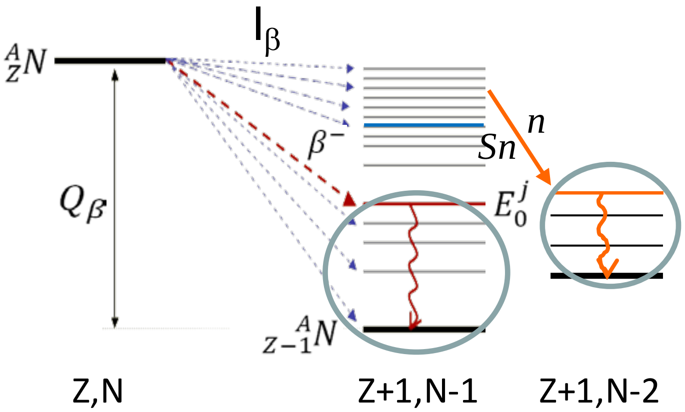
$$\llcorner\ {}^{A}_{Z+1}Y^{*} \rightarrow {}^{A}_{Z+1}Y \ + \gamma s$$





Beta Decay for Present and Future Reactors

- The exploitation of the products of the beta decay is threefold:
  - ☐ The **antineutrinos** escape and can be detected → reactor monitoring, potential non-proliferation tool and essential for fundamental physics
  - ☐ The released γ and β contribute to the "**decay heat**" → critical for reactor safety and economy
  - ☐ β-n emitters: **delayed neutron fractions** → important for the operation and control of the chain reaction of reactors



γ Measurement Caveat

- Before the 90s, conventional detection techniques: high resolution γ-ray spectroscopy
  - ❑ Excellent resolution but efficiency which strongly decreases at high energy
  - ❑ Danger of overlooking the existence of β-feeding into the high energy nuclear levels of daugther nuclei (especially with decay schemes with large Q-values)

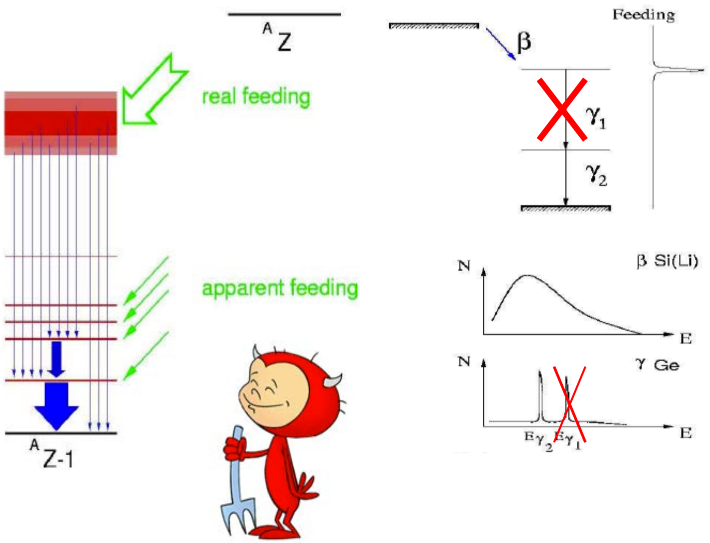
- Incomplete decay schemes: overestimate of the high-energy part of the FP β spectra

- Phenomenon commonly called « pandemonium effect** » by J. C Hardy in 1977

** J.C.Hardy et al., Phys. Lett. B, 71, 307 (1977)

➡ Strong potential bias in nuclear databases and all their applications (indirect effect on neutrino energy spectra computation)

Picture from A. Algora



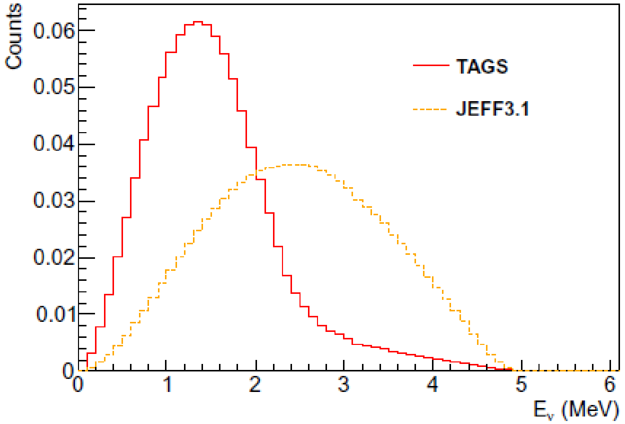
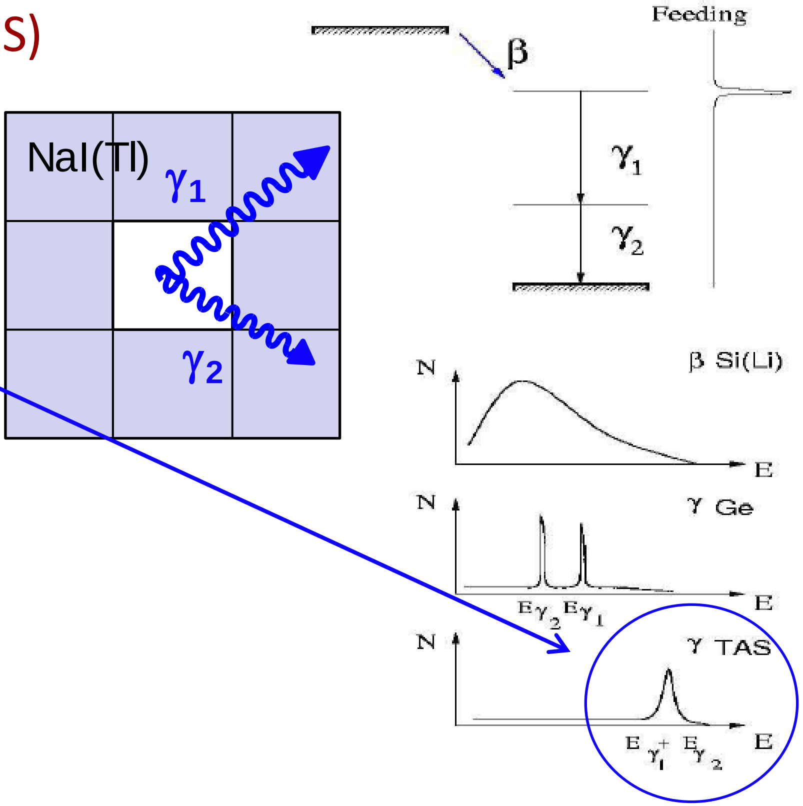


FIG. 1. Illustration of the pandemonium effect on the ^{105}Mo nucleus anti-ν energy spectrum presents in the JEFF3.1 data base and corrected in the TAS data.

TAGS: a Solution to the Pandemonium Effect

- Total absorption γ-ray spectroscopy (TAGS)
  - ❑ A TAS is a calorimeter
  - ❑ It contains big crystals covering 4π
  - ❑ Instead of detecting the individual gamma rays, absorbs the full gamma energy released by the gamma cascades in the β-decay process



- First TAS developed in the 70's but too small detectors to be efficient. Development of the TAGS method efficient and systematic since the 90's (Greenwood & al.)

- Calculation of level energy feeding through the resolution of the inverse problem by deconvolution
  - ❑ R_{ij} = matrix detector response
  - ❑ d_i = measured data
  - ❑ Extract f_j the level feeding by deconvolution

$$d_i = \sum_{j=1}^{m} R_{ij} \,.\, f_j \Rightarrow \boxed{I_i = \frac{f_i}{\sum_k f_k}}$$

J. L. Tain & D. Cano-Ott,
NIMA 571 (2007) 728

2 TAS Campains at IGISOL Jyväskylä in 2009 and 2014

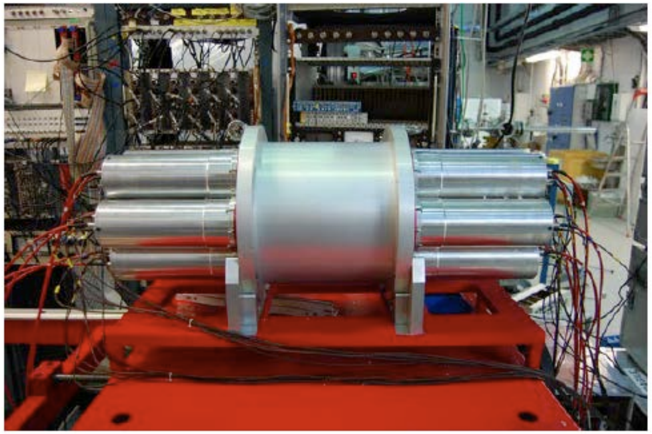
- IGISOL@Jyväskylä:
  - ☐ Proton induced fission ion-guide source
  - ☐ Mass separator magnet
  - ☐ Double Penning trap system to clean the beams

B. Rubio, J. L. Tain, A. Algora et al., Proceedings of the Int. Conf. For nuclear Data for Science and technology (ND2013)

- 2 (segmented) TAS campains :

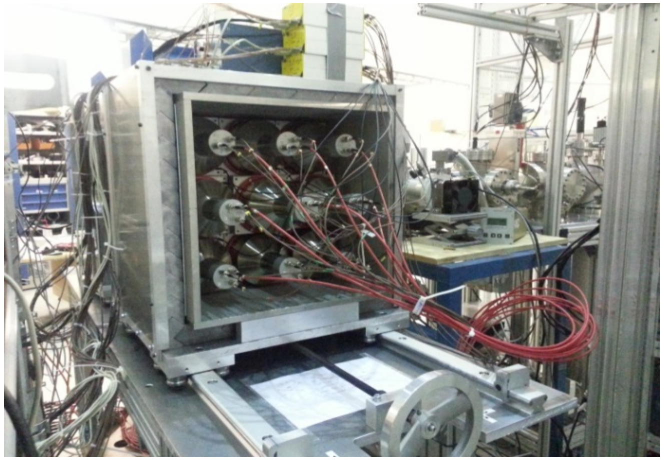
J.L. Tain et al., NIMA 803 (2015) 36
V. Guadilla et al., submitted to NIMA (2018)

☐ ROCINANTE (IFIC Valencia/Surrey):



- ✓ 12 BaF_2 covering 4π
- ✓ Detection efficiency of γ ray cascade >80% (up to 10 MeV)
- ✓ Coupled with a Si detector for β
- ✓ 7 nuclei (4 delayed neutron emitters) measured (6 for DH and 2 for anti-ν)

☐ DTAS (IFIC Valencia):



- ✓ 18 NaI(Tl) crystals of 15cm×15cm×25 cm
- ✓ Individual crystal resolutions: 7-8%
- ✓ Total efficiency: 80-90%
- ✓ Coupled with plastic scintillator for β
- ✓ 12 nuclei for anti-ν measured & 11 for DH

30 Measured Nuclei of Interest to DH, Anti-ν and DN

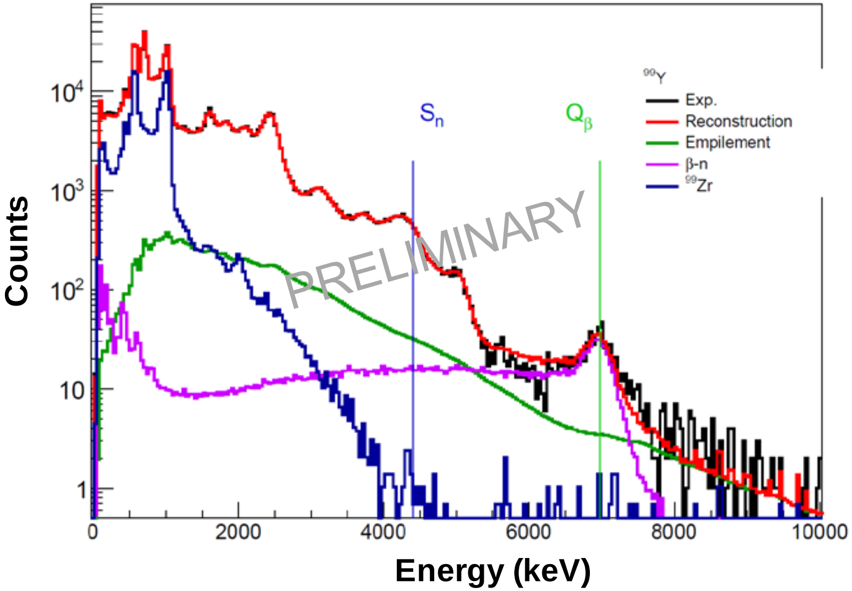
- 2014 campain (23 nuclei):

IAEA Report INDC(NDS) 0676 (2015)

Nuclide	Priority U/Pu	Priority Th/U	Priority $\overline{\nu}_e$	Nuclide	Priority U/Pu	Priority Th/U	Priority $\overline{\nu}_e$
^{95}Rb	1	2		^{102m}Nb	-	1	-
^{95}Sr	-	-	1	^{103}Tc	1	2	-
^{95}Y	-	-	1	^{103}Mo	1	2	-
^{96gs}Y	2	2	1	^{108}Tc	-	-	-
^{96m}Y	-	1	-	^{108}Mo	-	-	-
^{99}Y	-	-	1	^{137}Xe	1	3	-
^{99}Zr	2	1	-	^{138}Xe	-	1	-
^{98gs}Nb	1	1	1	^{137}I	1	2	1
^{98m}Nb	-	-	-	^{138}I	-	-	2
^{100gs}Nb	1	1	1	^{140}Cs	-	-	1
^{100m}Nb	-	1	-	^{142}Cs	3	-	1
^{102gs}Nb	2	2	1				

So far, 15 nuclei analyzed and under the process of publication in V. Guadilla's PhD thesis (9 nuclei Valencia) and L. Le Meur's PhD thesis (3 nuclei Subatech) + 6/7 nuclei being analyzed in Subatech.

An Example: the Case of ^{99}Y



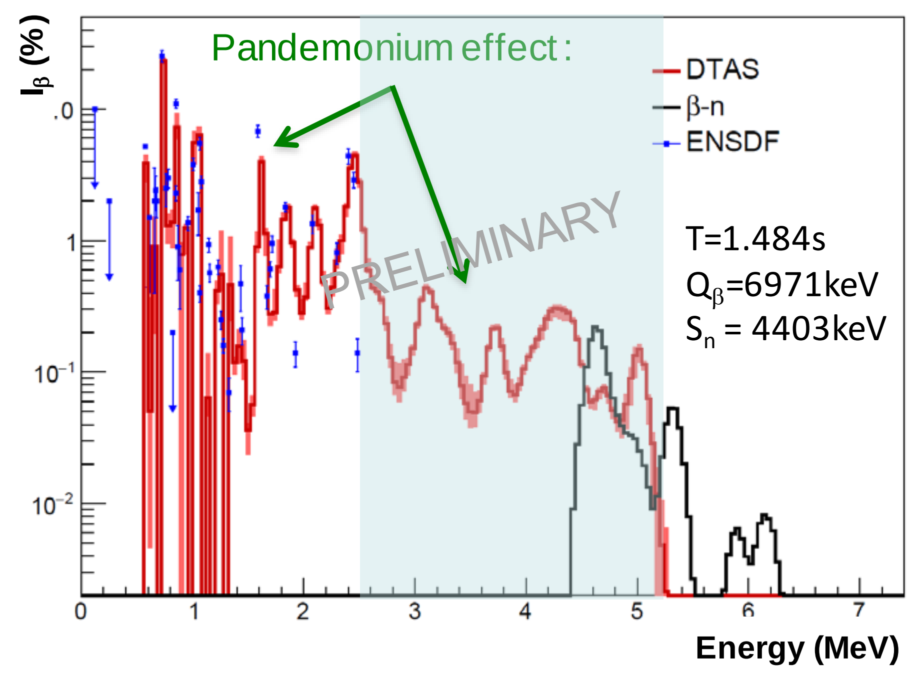
- ^{99}Y $\xrightarrow{\beta^-}$ ^{99}Zr ($T_{1/2}$ = 1.484s)
- Experimental spectrum β tagged -> cleaned from background
- Daughter contaminant ^{99}Zr subtracted
- Comparison to ENSDF
  - ☐ Clear pandemonium case

PhD Thesis work:
Loïc Le Meur (Subatech, Nantes)

$$d_i = \sum_j R_{ij} f_j \quad \Rightarrow \quad \mathbf{R_j} = \sum_{k=0}^{j-1} b_{jk} \mathbf{g_{jk}} \otimes \mathbf{R_k}$$

Deconvolution (Inverse problem) algorithms

Monte Carlo simulations
+
Nuclear statistical model



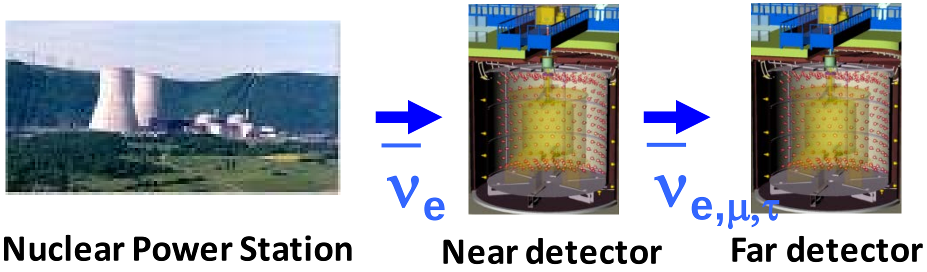
Pandemonium effect :

T=1.484s
Q_β=6971keV
S_n = 4403keV

Beta Decay and Applications: Reactor Antineutrinos

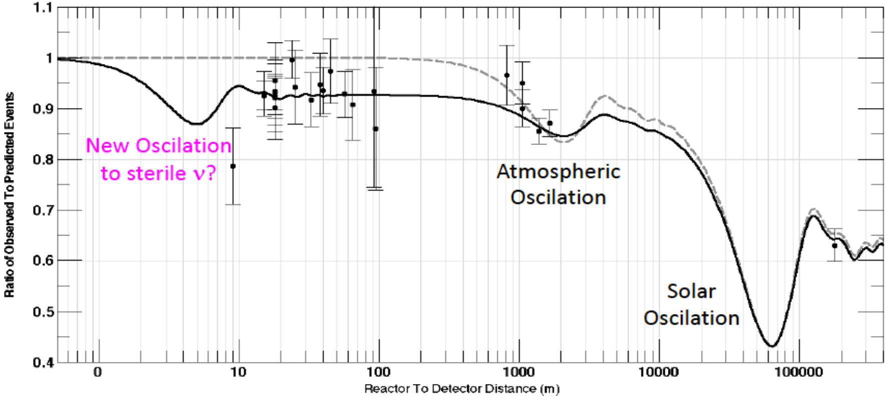
Reactor Antineutrinos & Fundamental Physics

- Measurement of the θ_{13} oscillation param by Double Chooz, Daya Bay, Reno
  - ☐ Independent computation of the anti-ν spectra using nuclear DB: conversion method



- Sterile neutrino measurement to explain the “reactor anomaly”
  - ☐ 6% deficit of the absolute value of the measured flux compared to the best prediction ILL data
  - ☐ Bump (spectrum distortion) in the full spectrum (btw 4.8-7.3 MeV)
  - ☐ Daya Bay PRL points-out a pb in the converted antineutrino spectra from ^{235}U measured beta spectrum @ILL

G. Mention *et al*. Phys. Rev. D83, 073006 (2011)



- Next generation reactor neutrino experiments like JUNO or background for other multipurpose experiment

➡ Putting integral beta measurement of ^{235}U of Scheckenbach *et al.* and sterile neutrinos into question.

➡ Growing interest in summation method to calculate anti-ν spectra

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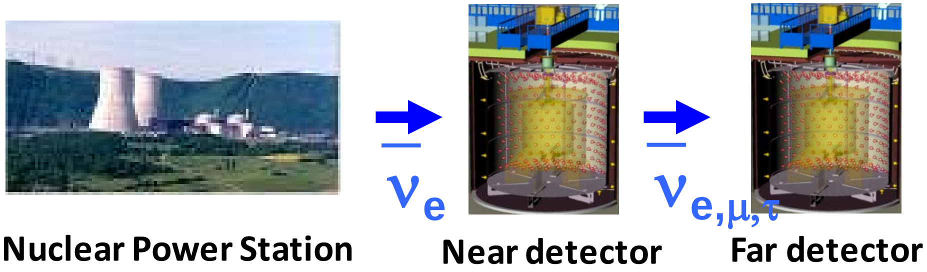

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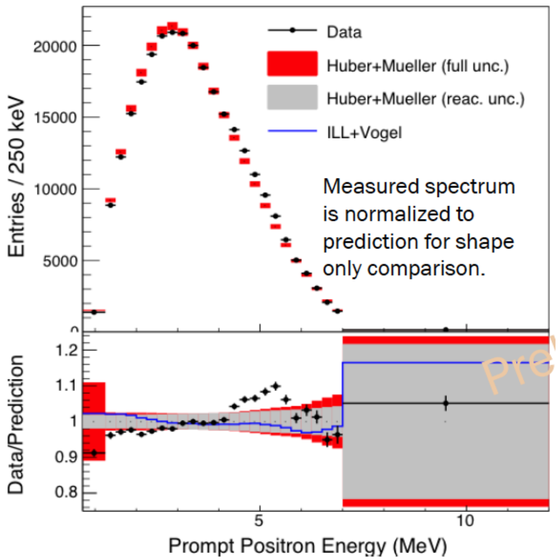
- Absolute shape comparison of data and prediction: $\chi^2/ndf = 41.8/21$



- Next generation reactor neutrino experiments like JUNO or background for other multipurpose experiment

➡ Putting integral beta measurement of ^{235}U of Scheckenbach *et al.* and sterile neutrinos into question.

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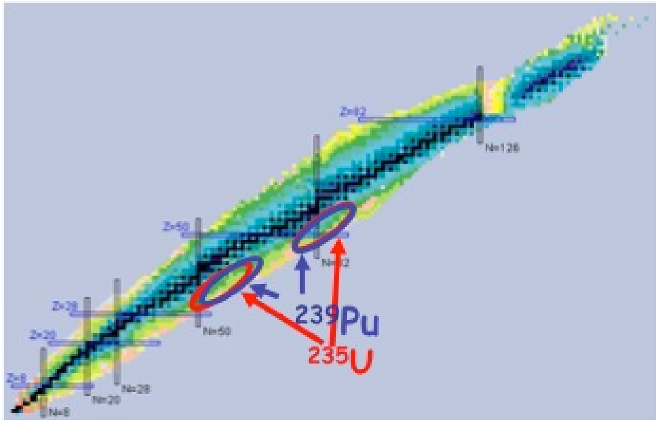
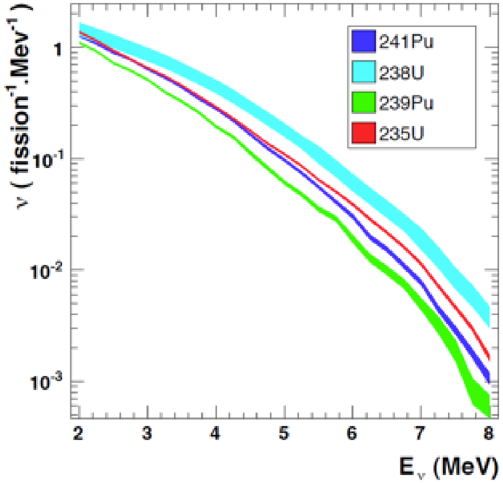
Antineutrinos for Peace





About 6 antineutrinos emitted per fission

→ About 10^{21} antineutrinos/s emitted by a 1 GW_e reactor

- Use the discrepancy between antineutrino flux and energies from U and Pu isotopes to infer reactor fuel isotopic composition and power
  - ☐ Reactor monitoring, non-proliferation and interest for the IAEA IAEA Report SG-EQGNRL-RP-0002 (2012)
  - ☐ Idea born in the 70s, demonstrated in the 80s/90s but developed lately

- The IAEA Nuclear Data section includes the measurements for reactor antineutrino spectra in their Priority lists (CRP meetings, TAGS consultant meetings…)

Anti-ν Spectra: the Summation Method

- SM first developed in 2011 (Mueller et al.) and improved in 2012 (Fallot et al.)
- Computation of antineutrino spectra for which no beta spectrum was measured. The SM is independent from the integral beta mesurement of Schreckenbach et al. @ ILL
- One of the only alternatives to ILL data!

- Reactor anti-ν energy spectrum can be computed with the SM as the DH, for one isotope k:

$$S_k(Z,A,E) = \sum_{fp=1}^{N_{fp}} A_{fp} \times \boxed{\sum_{b=1}^{N_b} I_{\beta_{fp}}^{\;b}} \times S_{fp}^{\;b} \boxed{\left(Z_{fp}, A_{fp}, E_{0\,fp}^{\;b}, E\right)}) \longleftarrow \boxed{\text{NUCLEAR DB}}$$

- S_{fp} theoretically computable but our approach different:
  - ☐ Exploit all data available on β-branches in modern nuclear databases in order to reduce the input of nuclear models
  - ☐ Strongly depends on nuclear databases: the choice of the DB cocktail is essential

- Coupled with reactor simulations: predictions for innovative fuels (Th, MA, …) and future reactor designs

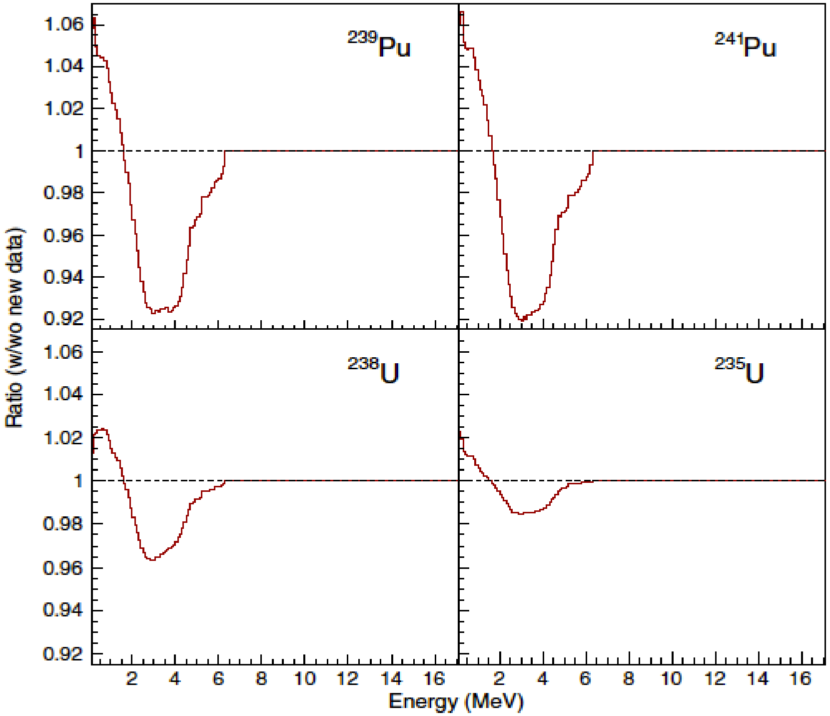
- Propagation of uncertainties under study
  - ☐ Necessity to have the covariance matrices of the fission yields, and possibly of decay data …

Anti-ν Spectra: the Summation Method

An example of summation calculation from Phys. Rev. Lett. 109,202504 (2012) Taking into consideration the TAS data of the $^{102;104–107}$Tc, ^{105}Mo, and ^{101}Nb isotopes measured @ Jyväskylä

- ❑ ~850 nuclei included
- ❑ Noticeable deviation from unity (1.5 to 8% decrease)
- ❑ Change in the flux (presented later)

M. Fallot *et al.*, PRL 109, 202504 (2012)



Relative Effects of the 2012 TAS data on the Antineutrino Spectra

A Reduced List of Important Contributors

A.-A. Zakari-Issoufou, PRL 115, 102503 (2015)

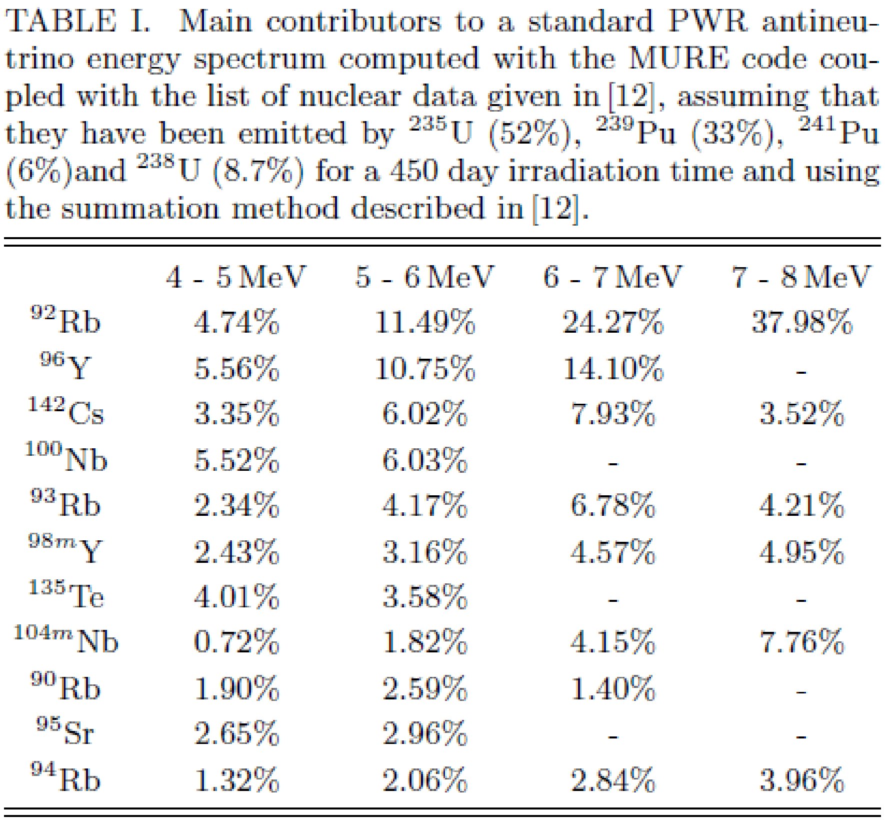


TABLE I. Main contributors to a standard PWR antineutrino energy spectrum computed with the MURE code coupled with the list of nuclear data given in [12], assuming that they have been emitted by ^{235}U (52%), ^{239}Pu (33%), ^{241}Pu (6%)and ^{238}U (8.7%) for a 450 day irradiation time and using the summation method described in [12].

	4 - 5 MeV	5 - 6 MeV	6 - 7 MeV	7 - 8 MeV
^{92}Rb	4.74%	11.49%	24.27%	37.98%
^{96}Y	5.56%	10.75%	14.10%	-
^{142}Cs	3.35%	6.02%	7.93%	3.52%
^{100}Nb	5.52%	6.03%	-	-
^{93}Rb	2.34%	4.17%	6.78%	4.21%
^{98m}Y	2.43%	3.16%	4.57%	4.95%
^{135}Te	4.01%	3.58%	-	-
^{104m}Nb	0.72%	1.82%	4.15%	7.76%
^{90}Rb	1.90%	2.59%	1.40%	-
^{95}Sr	2.65%	2.96%	-	-
^{94}Rb	1.32%	2.06%	2.84%	3.96%

- Summation calculations give the following priority list of nuclei, with a large contribution to the PWR antineutrino spectrum in the high energy bins

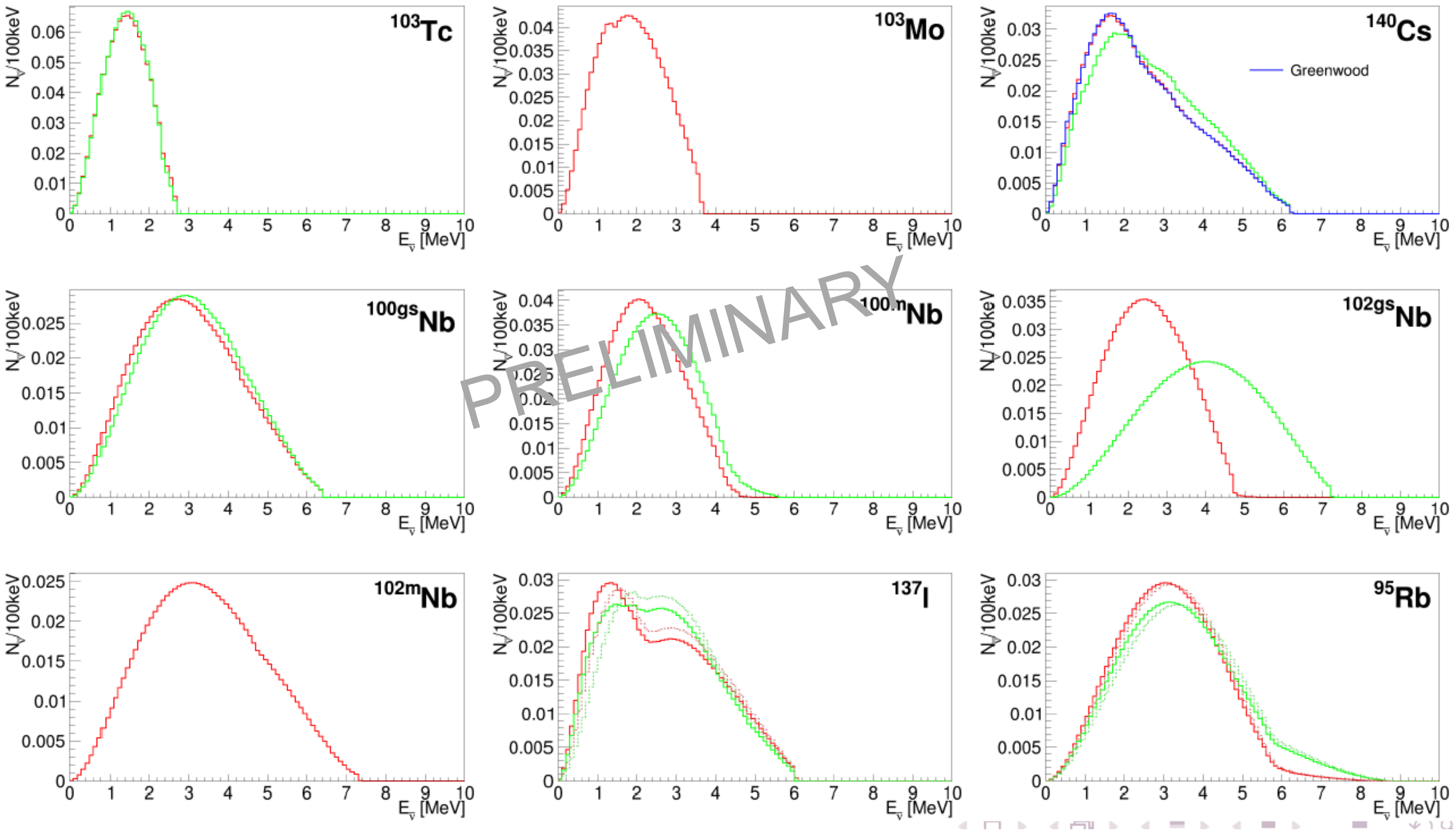
The number of contributors in these bins is small enough to give the hope to produce summation calculations with reduced systematic errors due to decay data at a relatively short time scale

- Involvement in IAEA TAGS' Consultant meetings for priority list of nuclei identification

Individual Anti-ν Energy Spectra: DTAS vs ENSDF

- Comparison of the individual antineutrino energy spectra between DTAS and ENSDF.
  - Strong pandemonium bias in previous nuclear databases
  - Impact the total antineutrino spectrum
  - First measurement of ^{102m}Nb

V. Guadilla's PhD thesis

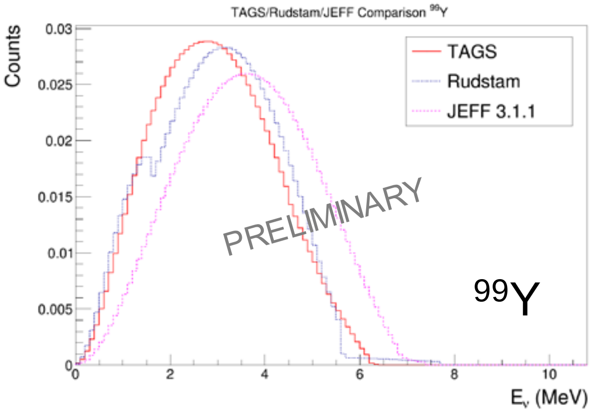


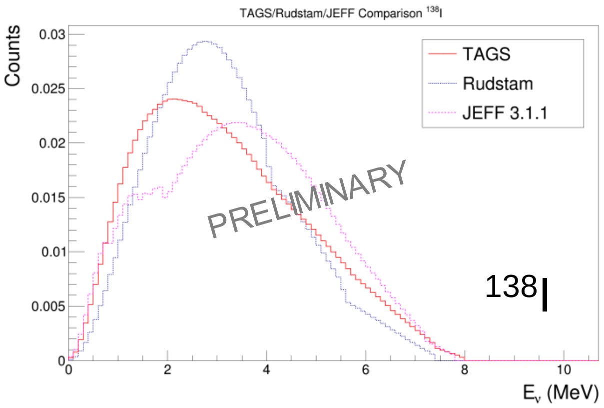
Individual Anti-ν Energy Spectra: ^{99}Y, ^{138}I and ^{142}Cs

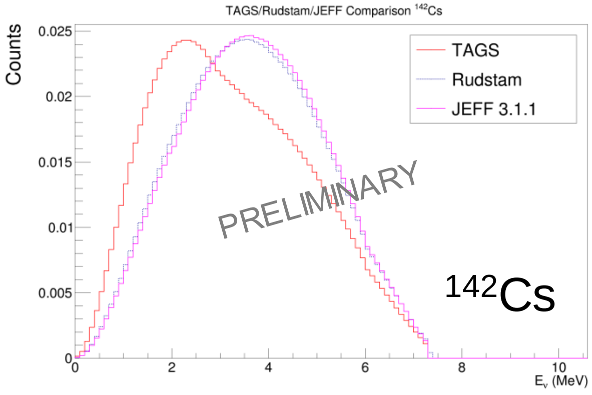
- Comparison of the individual antineutrino energy spectra between DTAS, the preferred nuclear database that was used for our previous calculation and JEFF 3.1.1.
  - ☐ Non pandemonium free data in JEFF 3.1.1
  - ☐ Rudstam β spectra converted
  - ☐ Shift towards low energy in TAS: apparent biases in Rudstam measurement but large error bars

➡ Impact the total antineutrino spectrum

Data: L. Le Meur’s PhD thesis

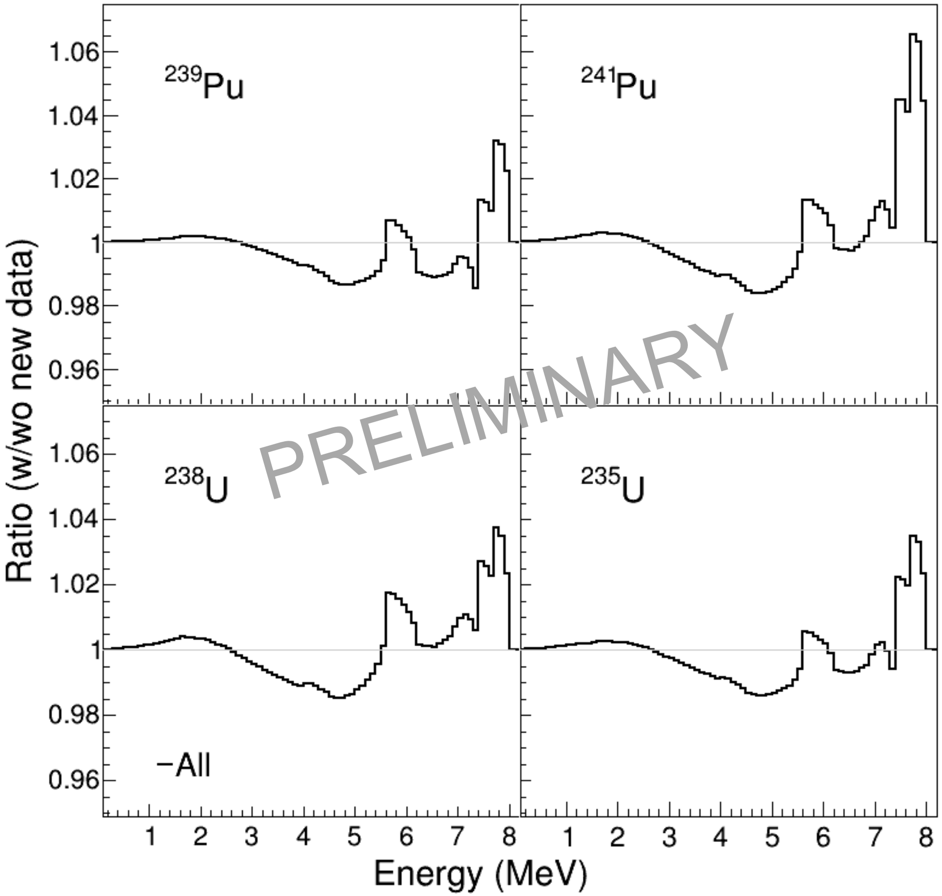






Associated Summation Calculation

- Summation calculation including the 3 nuclei compared to our previous preferred databases.
- Ratio of summation antineutrino spectra including the new TAS data over the same spectra but with the previous preferred database cocktail.
- Impact on the spectrum
  - ☐ Deviation up to 6% for ^{241}Pu and between 3 and 4% for the 3 other nuclei



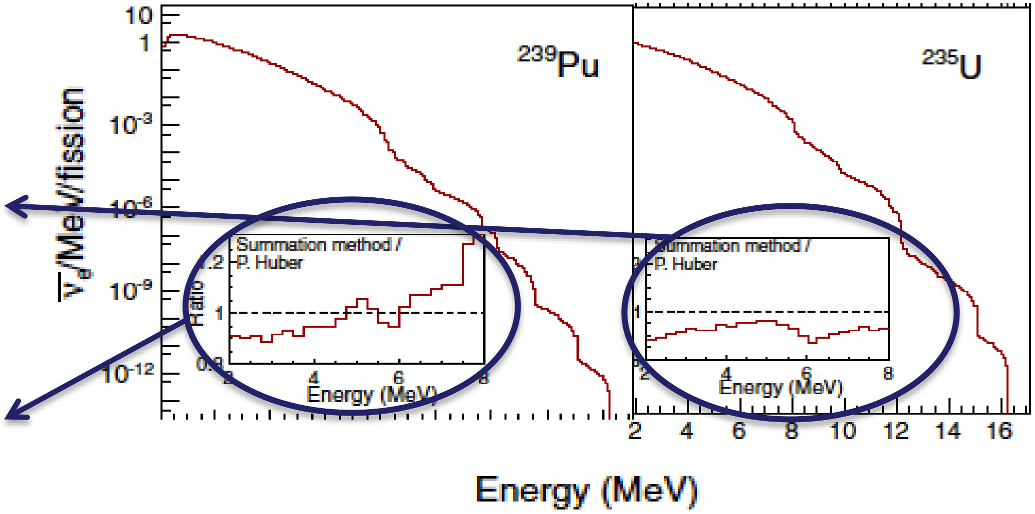
Data: L. Le Meur's PhD thesis

Comparison to the ILL Reference

- 2012 Ratio between spectra calculated with summation method and converted spectra from ILL measurements

❑ For ^{235}U: the summation is 5 to 10% below the conversion. Goes in the direction of Daya Bay's new 2017 result on the reactor anomaly: pb is in the ^{235}U spectrum!!!

❑ Summation spectra still not pandemonium free requiring new TAS measurements.



M. Fallot *et al.*, PRL 109, 202504 (2012)

Beta Decay and Applications: Decay Heat

Reactor Decay Heat (DH)

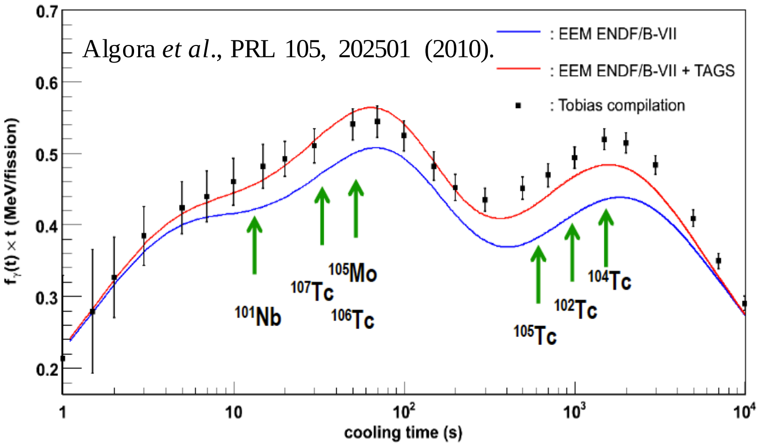
- Definition: following the shut-down of the chain reaction in a reactor, the nuclear fuel continues to release energy called decay heat.
- Emitters: essentially made up of the radiactive decays of FP and actinides
  - DH: residual power of 6-12% of the nominal power of the reactor just after its shut-down
- Why studying it? Evaluation of the reactor safety (design, operation, shielding, management of radioactive waste products, etc.) as well as various economic aspects of nuclear power generation requires a good knowledge of the DH

- Estimate through the only predictive method for futur reactors: the « summation method »
- Summation of all the fission product and actinide contributions inventoried for specific conditions of reactor operation and subsequent cooling period:

$$\mathbf{f}(t) = \sum_i \underbrace{(\bar{E}_{\beta,i} + \bar{E}_{\gamma,i})}_{\beta,\gamma\text{ decay}} \underbrace{\boldsymbol{\lambda}_i \mathbf{N}_i(t)}_{\text{Total decay constant (half-life) and Fission Yield}} \longrightarrow \left\{ \begin{array}{l} \bar{E}_\gamma = \sum_i I_\beta(E_i) E_i \\ \bar{E}_\beta = \sum_i I_\beta(E_i) < E_{\beta i} > \end{array} \right.$$

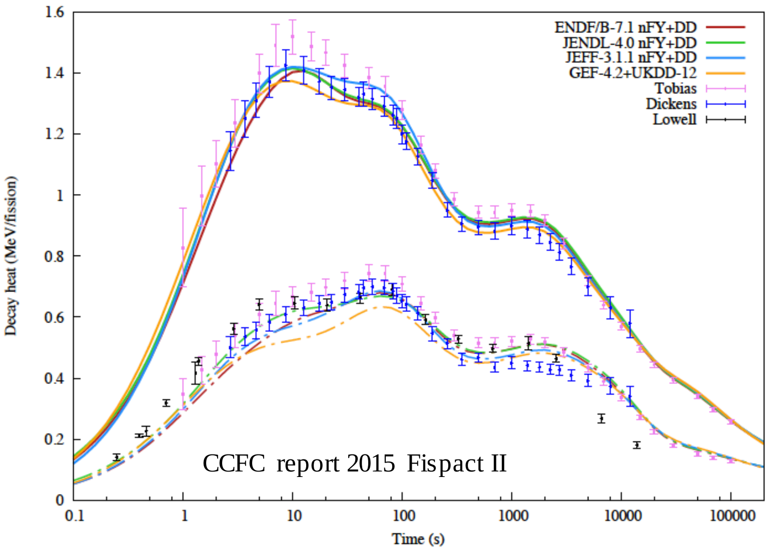
Decay Heat: Issues with Nuclear Data

- In the 1970s: important discrepancies observed comparing DH calculation and benchmark experiments ("pandemonium effect")
- Inclusion of mean beta and gamma energies derived from Gross Theory of Beta Decay to compensate the missing beta-strenghs
- Since the 1990s: temporary solution step by step replaced by the use of measured data with total absorption spectrometers TAGS



Impact of the results for ^{239}Pu: electromagnetic component

- Conclusion from CCFC report on total and γ DH:
  - ☐ Integral measurements not in agreement for several cooling times of the most well-known nuclide
  - ☐ Discrepancy between data and simulations using different DB for γ heat
  - ☐ Total heat better than γ heat => probably comes from a compensation on ELP and EEM but still not fully understood



Total (solid) and gamma (dash) decay heat from thermal pulse on ^{235}U

Nuclear Science NEA/WPEC-25 (2007), Report INDC(NDS)-0577 (2009), Report INDC(NDS-0551, Report INDC(NDS)-0676 (2015), CCFC report 2015 Fispact II

Average Energies of the Fission Products

- Mean gamma and bêta energies from bêta intensities:

$$\bar{E}_\gamma = \sum_i I_\beta(E_i) E_i$$

$$\bar{E}_\beta = \sum_i I_\beta(E_i) < E_{\beta i} >$$

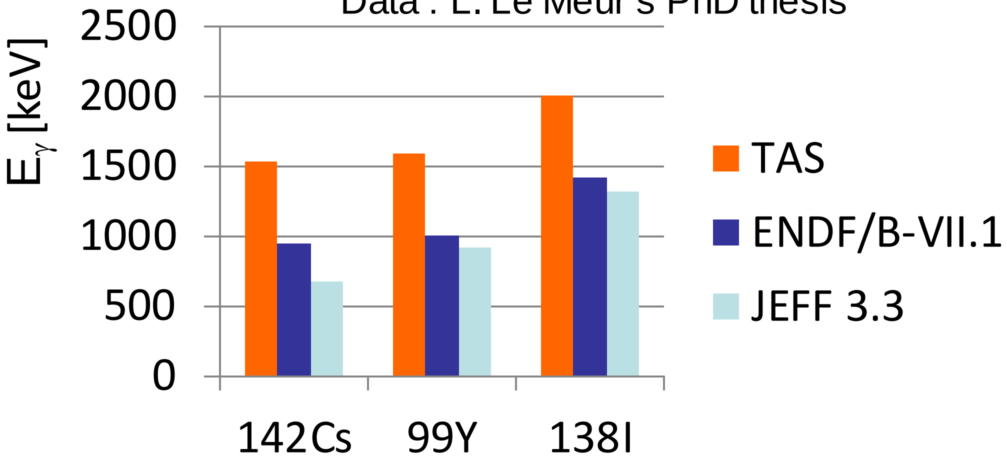
- Comparison to nuclear data bases ENDF-B-VII.1 and JEFF3.3 pandemonium biased

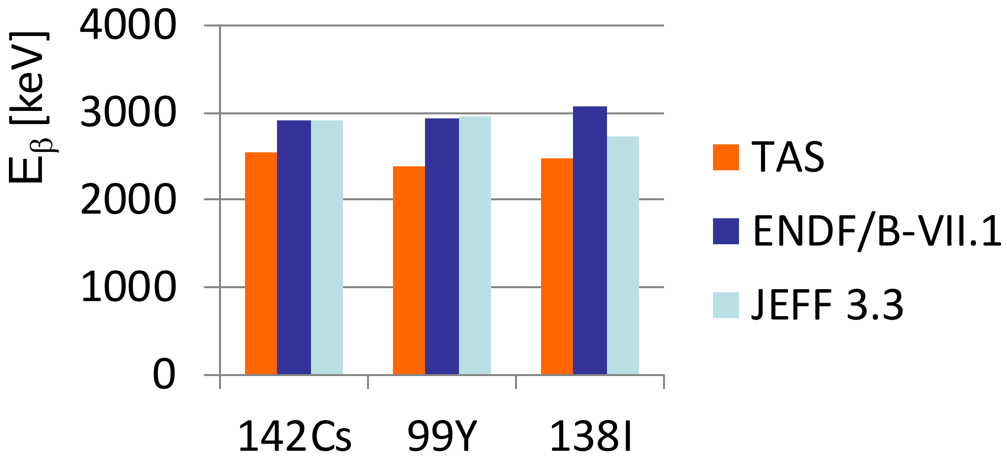
- Issue with uncertainties in nuclear data bases:
  - Example of ^{138}I:

ΔE_β (JEFF3.3-TAS) ~ 250keV (~2x σ_E(JEFF))

ΔE_β (ENDF/B7-TAS) ~ 600 keV (~2x σ_E(ENDF))

Data : L. Le Meur's PhD thesis

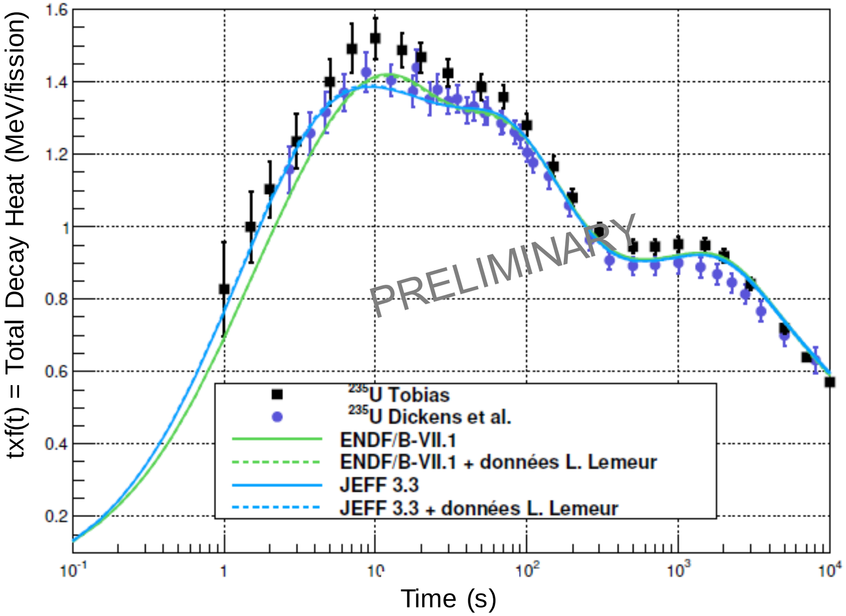




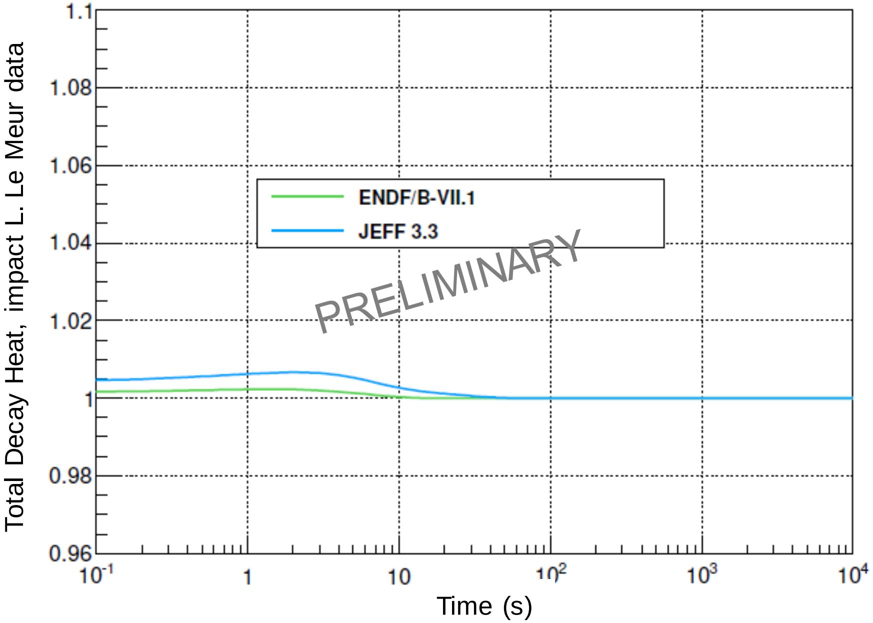
Collaboration with the CEA Cadarache to study the impact of reevaluated uncertainties in nuclear databases based on the differences observed between TAGS and JEFF 3.1.1 on the total uncertainty associated to DH

Impact of ^{99}Y, ^{138}I and ^{142}Cs on Total DH

^{235}U fission pulses



Calculations: giot@subatech.in2p3.fr
Data : L. Le Meur's PhD thesis



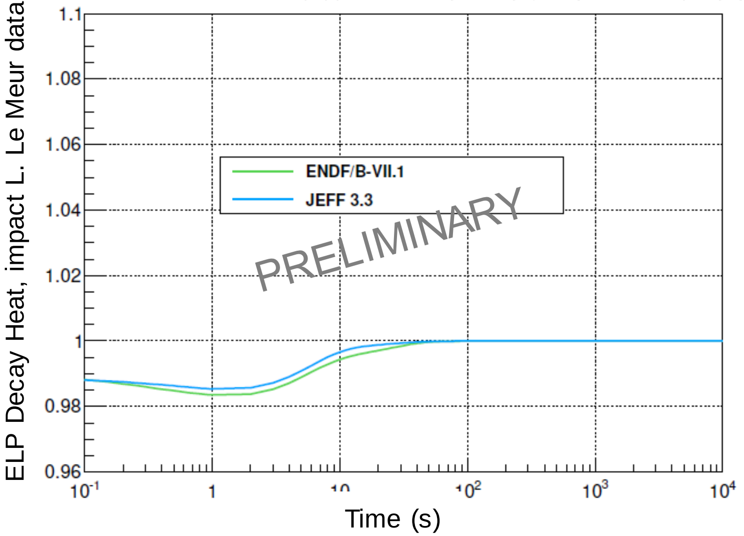
- Summation calculations performed by L. Giot using Serpent 2 code.
- Impact of the sum of the 3 nuclei lower than 1% in both DB on the total DH below 10s

Impact of ^{99}Y, ^{138}I and ^{142}Cs on ELP/EEM DH

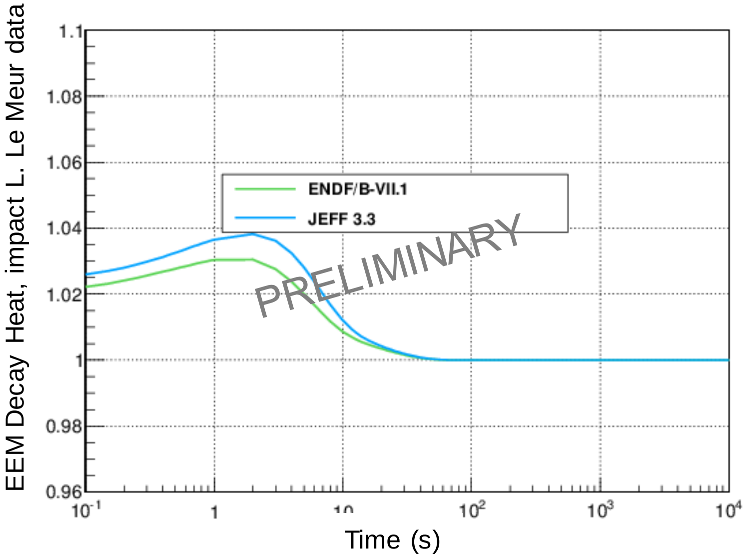
^{235}U fission pulses

Calculations: giot@subatech.in2p3.fr
Data : L. Le Meur's PhD thesis

- The 2 ELP and EEM compensate
- 1% to 2% impact on ELP
- 2% to 3% (ENDF) / 4% (JEFF3.3) on EEM
- ^{142}Cs (not shown) which was of Priority 3 in the IAEA list of priority nuclei to measure for DH contributes to ~2% on EEM in both NDB
- ^{99}Y (not shown) not listed as priority has also a not negligeable impact of ~1.5%



Light particle component

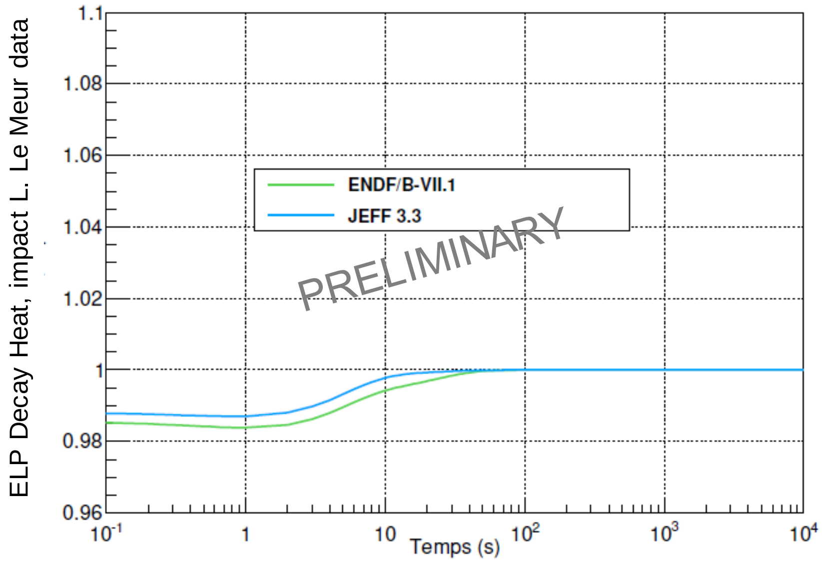


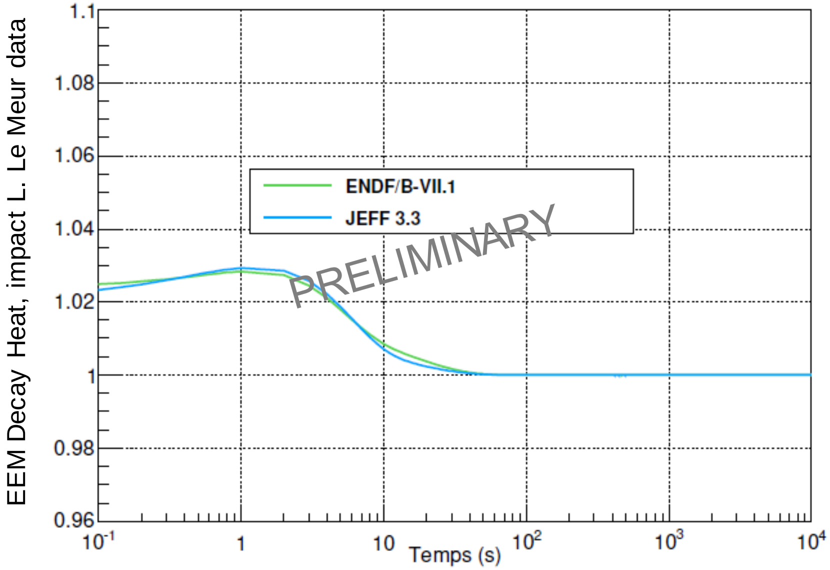
Electro-magnetic component

Impact of ^{99}Y, ^{138}I and ^{142}Cs on Total DH

^{239}Pu fission pulses

Calculations: giot@subatech.in2p3.fr
Data : L. Le Meur's PhD thesis





- Similar behaviour than for ^{235}U fission pulses comparing JEFF3.3 and ENDF-BVII for both the total DH and the EEM and ELP.
- Impact less than 0.5% on total DH for both libraries which comes from the compensation of EEM and ELP
- Similar impacts of ^{142}Cs and ^{99}Y on ELP (~0.5%) and EEM (~1-1.5%) in both DB

Knowing that these nuclei were not of priority 1 for DH in our list, these results are encouraging and the effort to measure and study the impact of TAS data has to be sustained

Beta Decay and Applications: bDN

IAEA and β-Delayed Neutron (bDN) Emission

Paraskevi (Vivian) Dimitriou
Nuclear Data Section,
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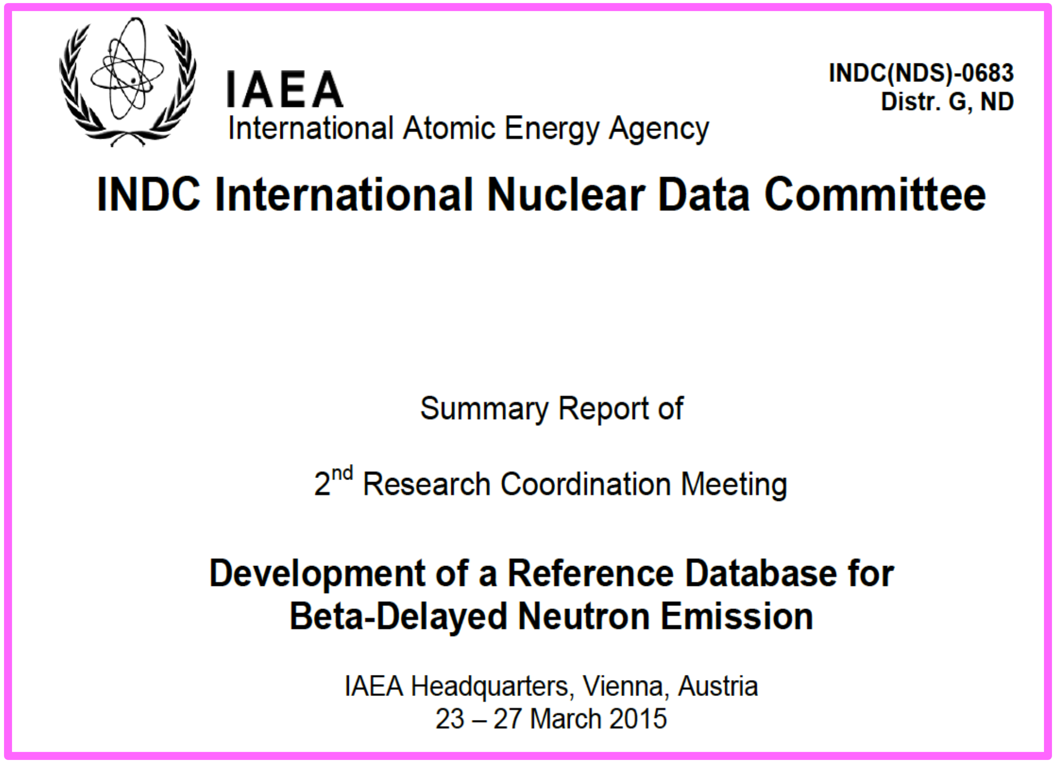
International Atomic Energy Agency,
Vienna, Austria



IAEA
International Atomic Energy Agency

INDC(NDS)-0683
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INDC International Nuclear Data Committee

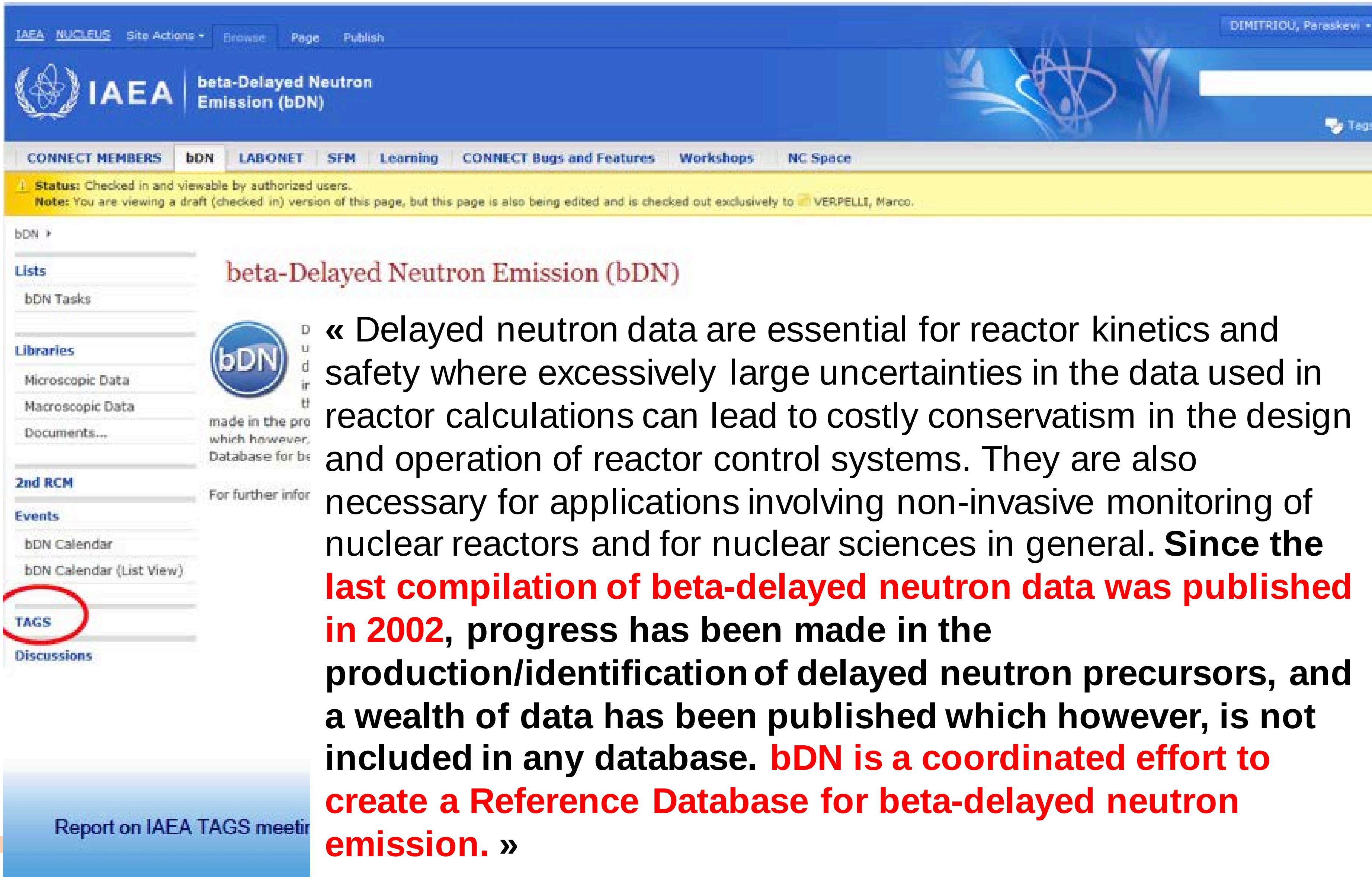
Summary Report of

2nd Research Coordination Meeting

Development of a Reference Database for Beta-Delayed Neutron Emission

IAEA Headquarters, Vienna, Austria
23 – 27 March 2015

IAEA CONNECT SharePoint platform for collaboration within bDN CRP and TAGS meeting participants



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beta-Delayed Neutron Emission (bDN)

made in the pro
which however,
Database for be

For further infor

Report on IAEA TAGS meetir

« Delayed neutron data are essential for reactor kinetics and safety where excessively large uncertainties in the data used in reactor calculations can lead to costly conservatism in the design and operation of reactor control systems. They are also necessary for applications involving non-invasive monitoring of nuclear reactors and for nuclear sciences in general. **Since the last compilation of beta-delayed neutron data was published in 2002, progress has been made in the production/identification of delayed neutron precursors, and a wealth of data has been published which however, is not included in any database. bDN is a coordinated effort to create a Reference Database for beta-delayed neutron emission.** »

AIEA CRP for delayed neutron interests

- With the summation calculation tools developed, we contribute to the development of a reference database for beta-delayed neutron emission
  - ☐ Inter-comparison of summation calculations between groups for the 4 major actinides
  - ☐ Total delayed neutron yield $\bar{\nu}_d$ and associated uncertainties
  - ☐ Most important contributors to $\bar{\nu}_d$ for thermal and fast neutron-induced fission
  - ☐ Evaluated databases compared: JEFF3.1.1, ENDF/B-VIII.0 and JENDL/DDF-2015
  - ☐ …

To be published in Nuclear Data Sheets, P. Dimitriou et al.

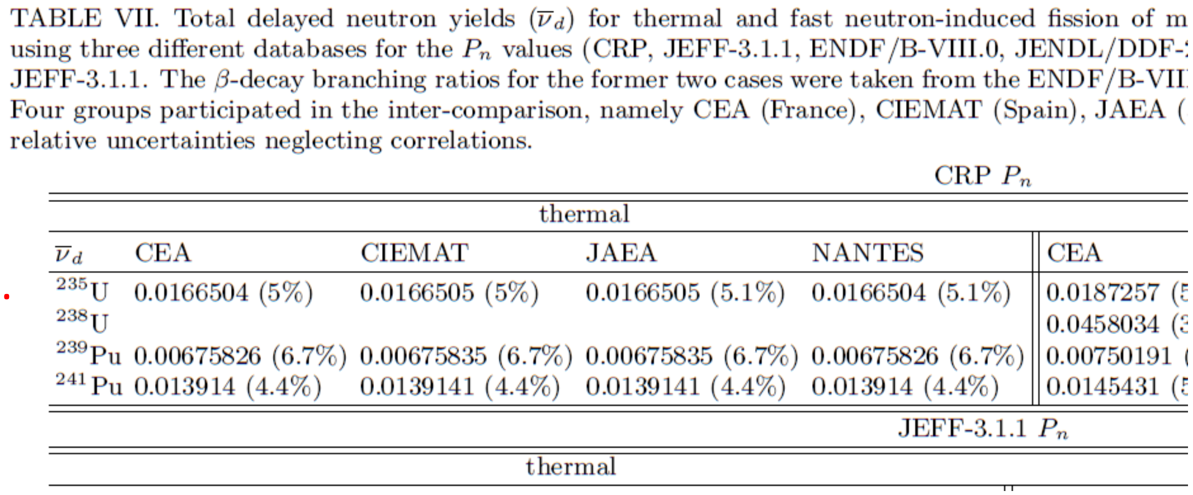

TABLE VII. Total delayed neutron yields ($\overline{\nu}_d$) for thermal and fast neutron-induced fission of m
using three different databases for the P_n values (CRP, JEFF-3.1.1, ENDF/B-VIII.0, JENDL/DDF-
JEFF-3.1.1. The β-decay branching ratios for the former two cases were taken from the ENDF/B-VII
Four groups participated in the inter-comparison, namely CEA (France), CIEMAT (Spain), JAEA (
relative uncertainties neglecting correlations.

CRP P_n

thermal

$\overline{\nu}_d$	CEA	CIEMAT	JAEA	NANTES	CEA
^{235}U	0.0166504 (5%)	0.0166505 (5%)	0.0166505 (5.1%)	0.0166504 (5.1%)	0.0187257 (
^{238}U					0.0458034 (
^{239}Pu	0.00675826 (6.7%)	0.00675835 (6.7%)	0.00675835 (6.7%)	0.00675826 (6.7%)	0.00750191
^{241}Pu	0.013914 (4.4%)	0.0139141 (4.4%)	0.0139141 (4.4%)	0.013914 (4.4%)	0.0145431 (

JEFF-3.1.1 P_n

thermal

See D. Foligno's presentation for more details about bDN !

Conclusion/Perspectives

- TAS technique: a solution to the Pandemonium effect

- Measurements of many nuclei of interest for nuclear database, anti-ν, DH and a few β-delayed n emitters during the past years => we are going in the right direction!

- The Summation method allows to identify the nuclei of interest and to quantify their impacts both on DH and anti-n from reactors.

- Inclusion of recent TAS data in antineutrino spectrum and DH calculations. It shows again non negligeable effects

- Next steps:
  - ☐ Continue to Assess impact of new TAS experimental results (end 2018) ;
  - ☐ New TAS experiment programmed in 2019 in Jyväskylä ;
  - ☐ Propagation of uncertainties including FY correlations, work on Decay Data correlations… ;
  - ☐ Future outcomes from β-shape measurements, impact on DH & anti-ν > 2019…

TAS COLLABORATION

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Thank you !