D. Rochman :: NES :: Paul Scherrer Institut

From nuclear data to spent fuel storage optimization

NES colloquium, PSI, January 15, 2020

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Summary

1. <u>General information on nuclear data</u>
   - Context
   - What are we talking about, who's a user ?
   - Can we predict a cross section ?
   - International efforts, Nuclear data lifecycle

2. <u>Application and examples</u>
   - Astrophysics, medical
   - Reactor physics
   - Spent nuclear fuel

3. <u>Current and Future developments</u>
   - Better nuclear data
   - Better QA
   - Better integration for optimization
   - Better measurements

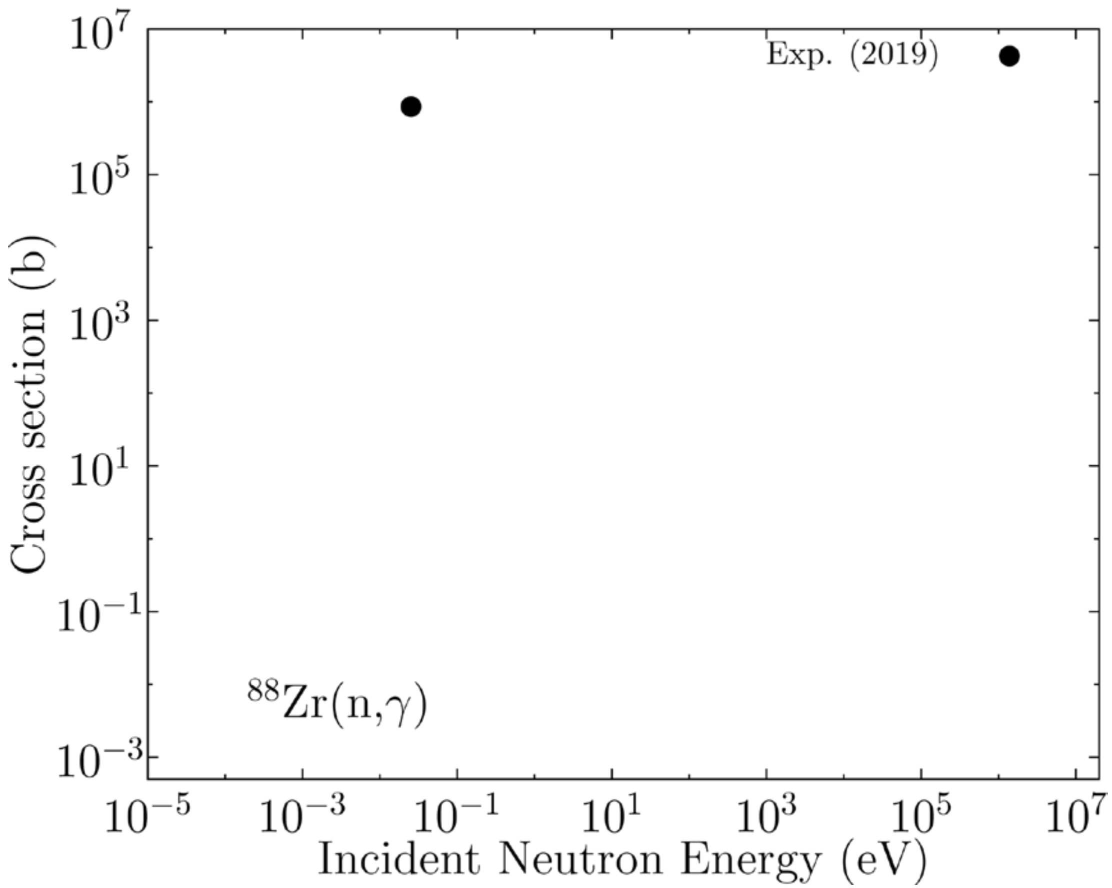
4. <u>Conclusion</u>

(*no deep details will be provided in this presentation, but specific points can be discussed later*)

1. General information on nuclear data

Context 1: A representative case: $^{88}Zr(n,\gamma)$

- The thermal $^{88}Zr(n,\gamma)$ cross section was recently measured, see Nature (2019).
- Its value is 861 000 barns ± 8 %
- The last time such a large cross section was "discovered" was in the 1940s.



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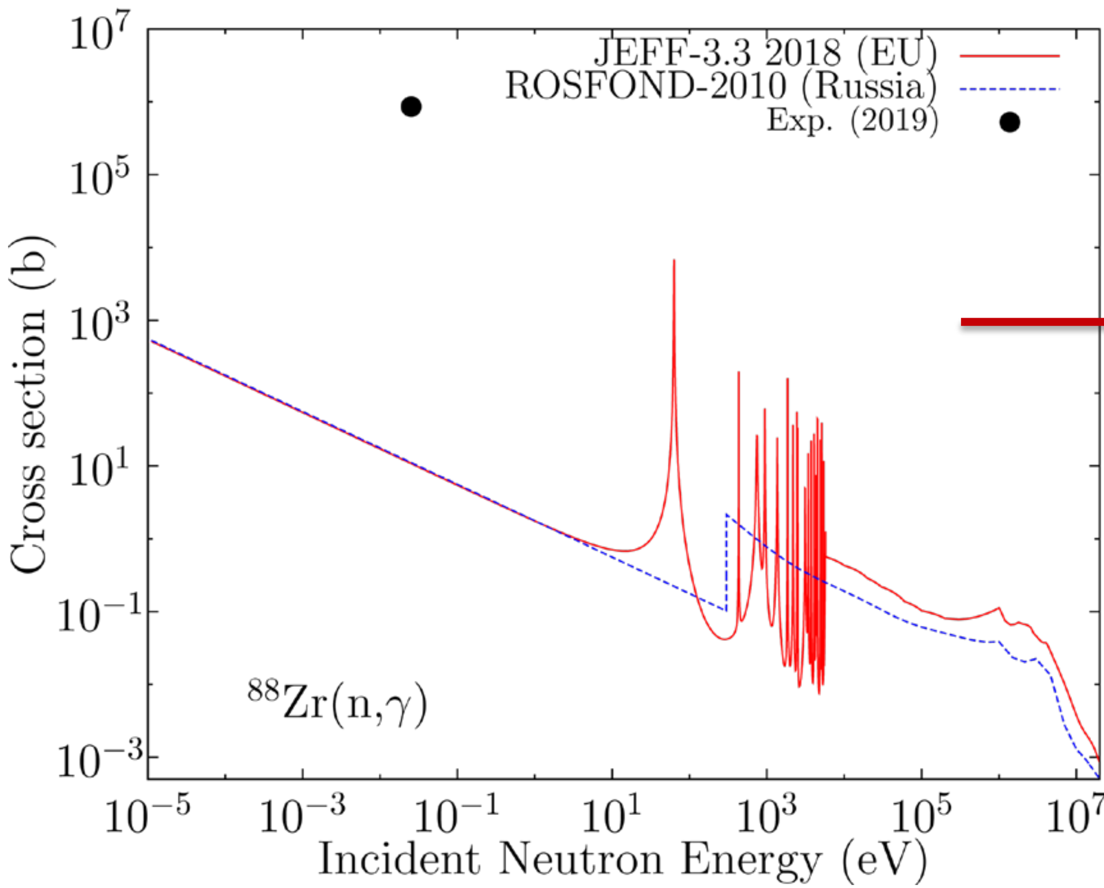
- The last time such a large cross section was “discovered” was in the 1940s.

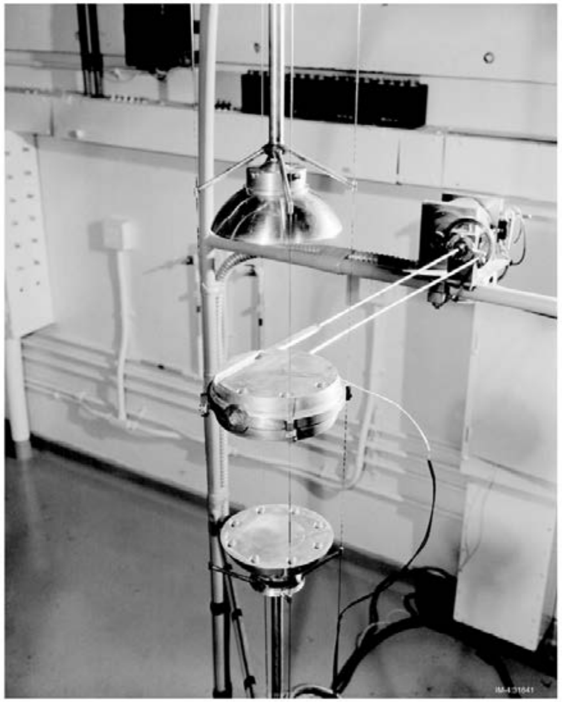
- Before the measurement, the prediction from the “nuclear data libraries” was

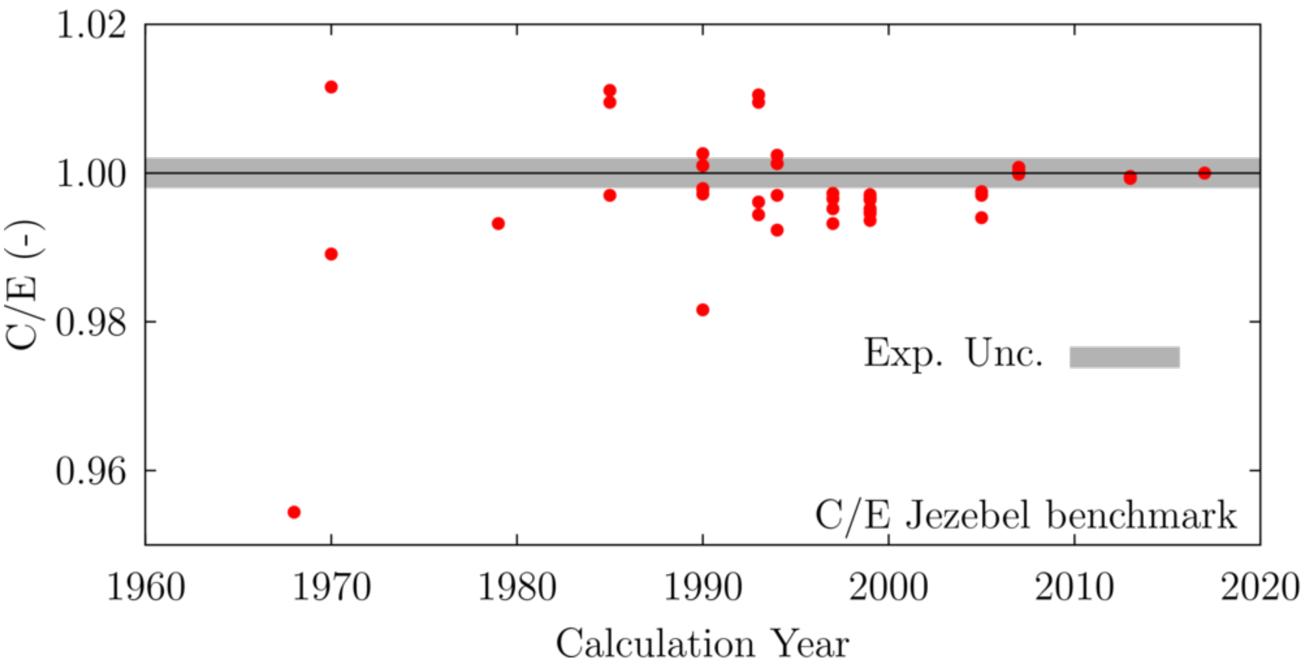


No predictive power

Context 2: fit well, but still uncertain

- Measured cross section: nominal well known, but uncertainty beyond engineering expectations
- Example on the Jezebel benchmark (≈16 kg of Pu239), performed in 1954-1955





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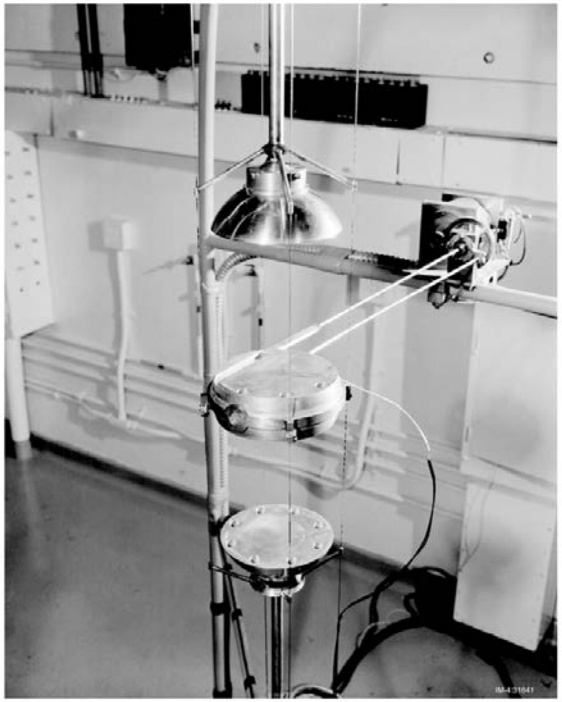

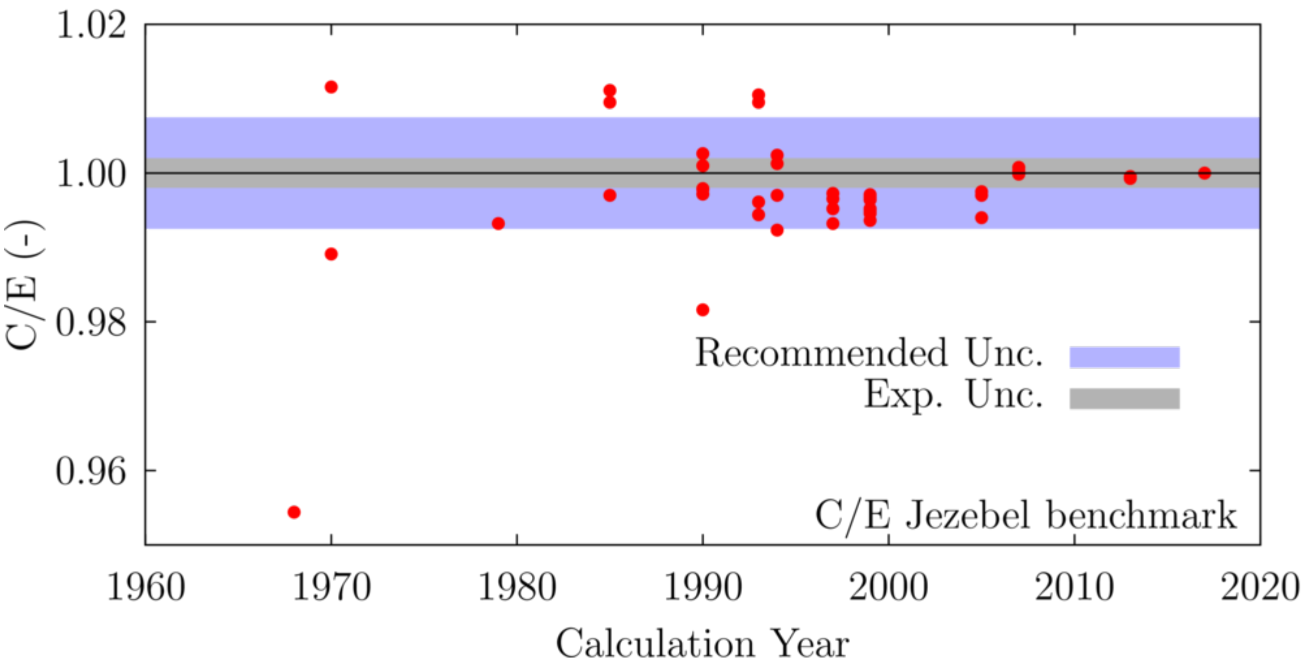

→ The “good fit” is not translated in “confidence interval”

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Nuclear data on the news

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U.S. NEWS MARCH 22, 2011 / 9:45 AM

U.S., Russia exchange nuclear data

(0)

President Dimitri Medvedev (L) of Russia and U.S. President Barack Obama hold a bilateral meeting at the United Nations in New York on September 23, 2009. UPI/Olivier Douliery/Pool | License Photo

WASHINGTON, March 22 (UPI) -- A senior U.S. official confirmed Russia and the United States have begun exchanging nuclear stockpile information under the new arms reduction treaty.

Middlebury Institute *of* International Studies at Monterey
James Martin Center for Nonproliferation Studies
1955

ABOUT | RESEARCH | EVENTS | EDUCATION

Decreasing the Risk of Iran's 5% Enriched Uranium Hexafluoride Stockpile

Dr. F. Dalnoki-Veress
March 3, 2015

Read the full report on a new option for Iran's uranium stockpile:
A Crazy idea? Isotopic Denaturing of Iran's 5% UF6 Stockpile with Reprocessed Uranium

The Nuclear Negotiations

The most challenging problem in the negotiations between Iran and the P5+1 is dealing with the scope of the enrichment program envisaged by Iran. Iran has stated that it requires enough enrichment capacity to be able to provide fuel for the Bushehr reactor requiring at least 100,000 SWU/year, which is an order of magnitude more capacity than Iran currently has installed. However, the P5+1 are likely to insist on an annual production limit a factor of 20 times less. Another challenge is the quantity of low enriched UF_6 (uranium hexafluoride) that currently exists in Iran's stockpile or may be produced in the future, which could be re-enriched to weapons grade in a breakout scenario.

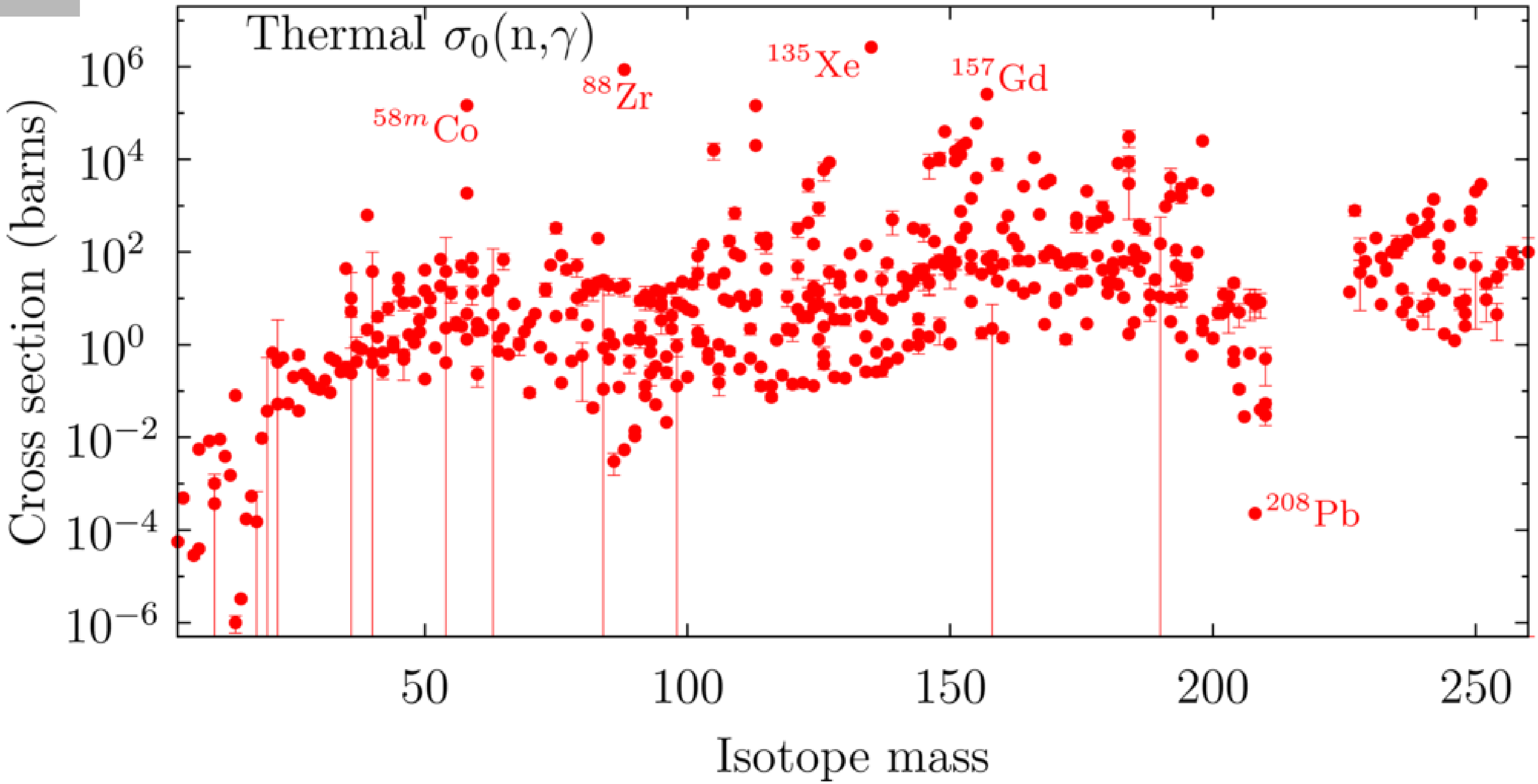
Worker at Uranium Conversion Facility in Iran, Source: state.gov

follow the following steps (sorry, this is a bit technical).

- Derive expression for the masses and concentrations for a 2-step blended material based on the initial feed mass of 7952.9 kg near-5% U-235 mass.
- Calculate neutron production as a function of the concentration of U-232 due to alpha particle activity from U-232 daughters interacting through the (α,n) reaction on assumed impurities present if the UF_6 is converted to metal form (assumed composition of metal as on the Novosibirsk Chemical Concentrate Plant website). We use SRIM 2008 to calculate the α-particle energy loss, TENDL-2013 for the cross-sections of the impurities, and ORNL's Scale 6 to calculate the quantity of U-232 daughters after an assumed 1 year of decay.

What are nuclear data ?

- Neutron-induced reactions:
  - Microscopic cross sections, thermal scattering data, from 0 to ≈200 MeV
  - Angular distributions, emitted spectra, fission yields…



- But also charged particle-induced reaction

What are nuclear data ?

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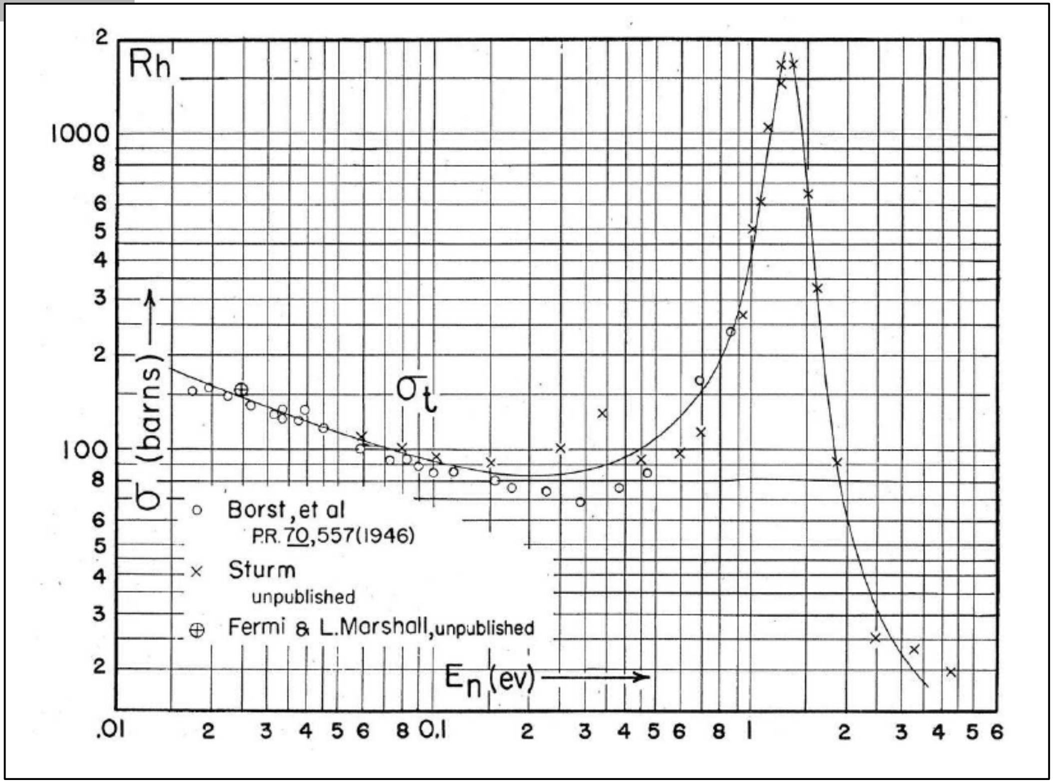

What are nuclear data ?

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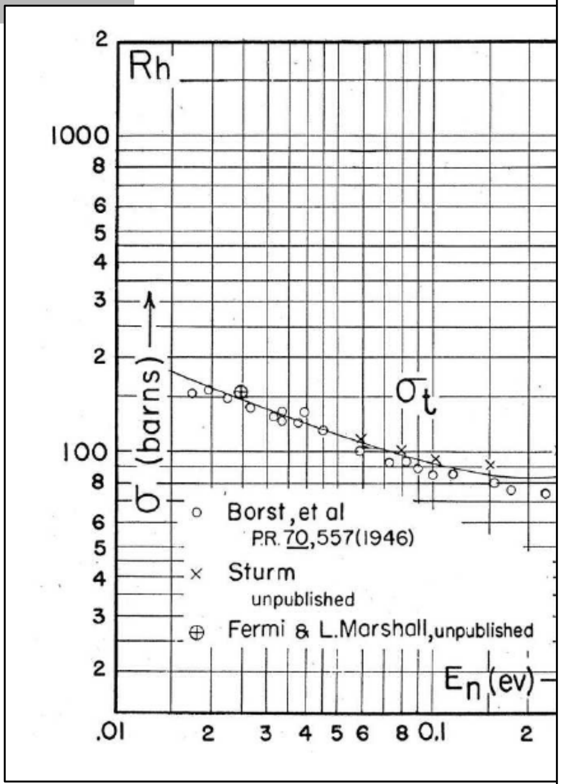
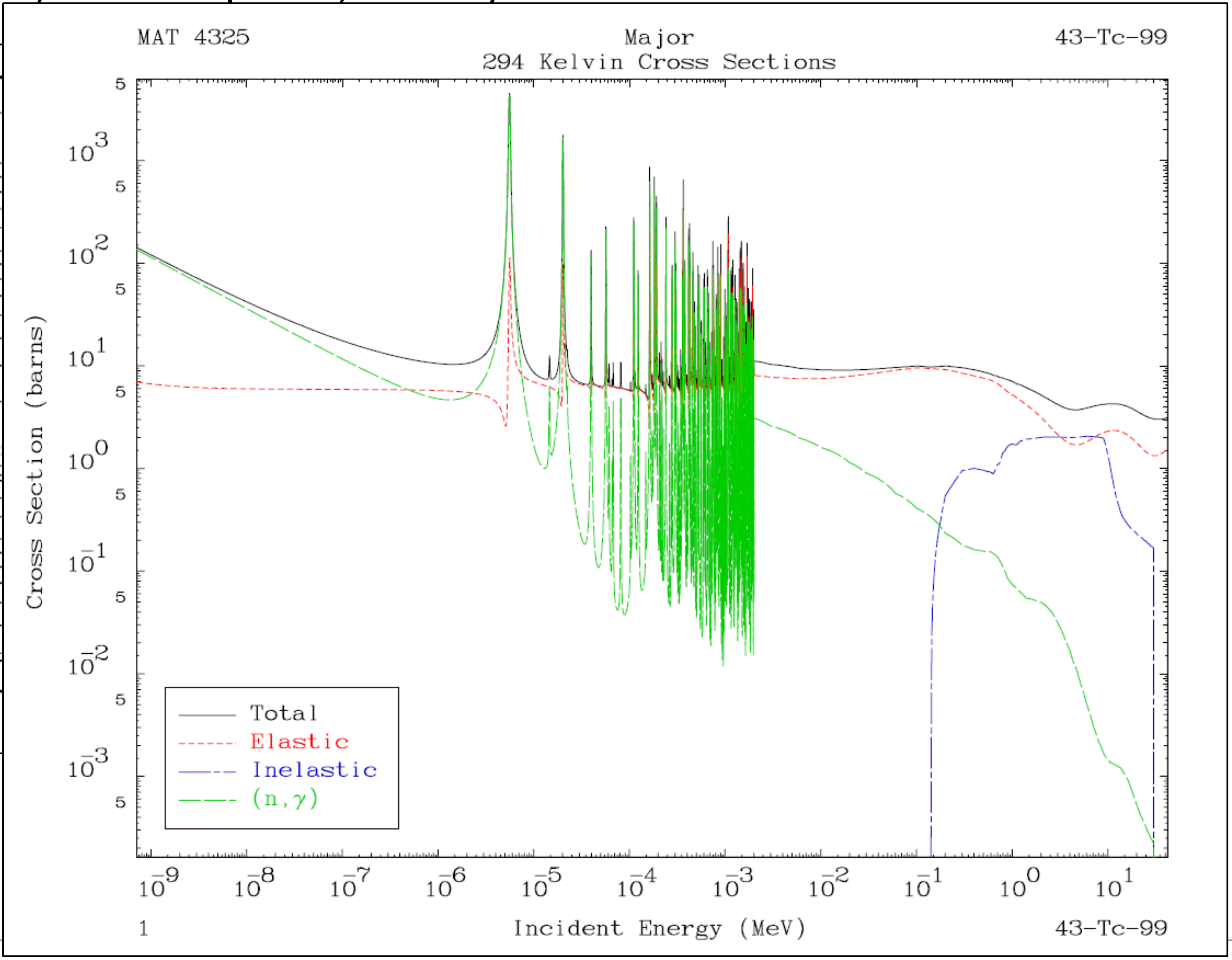




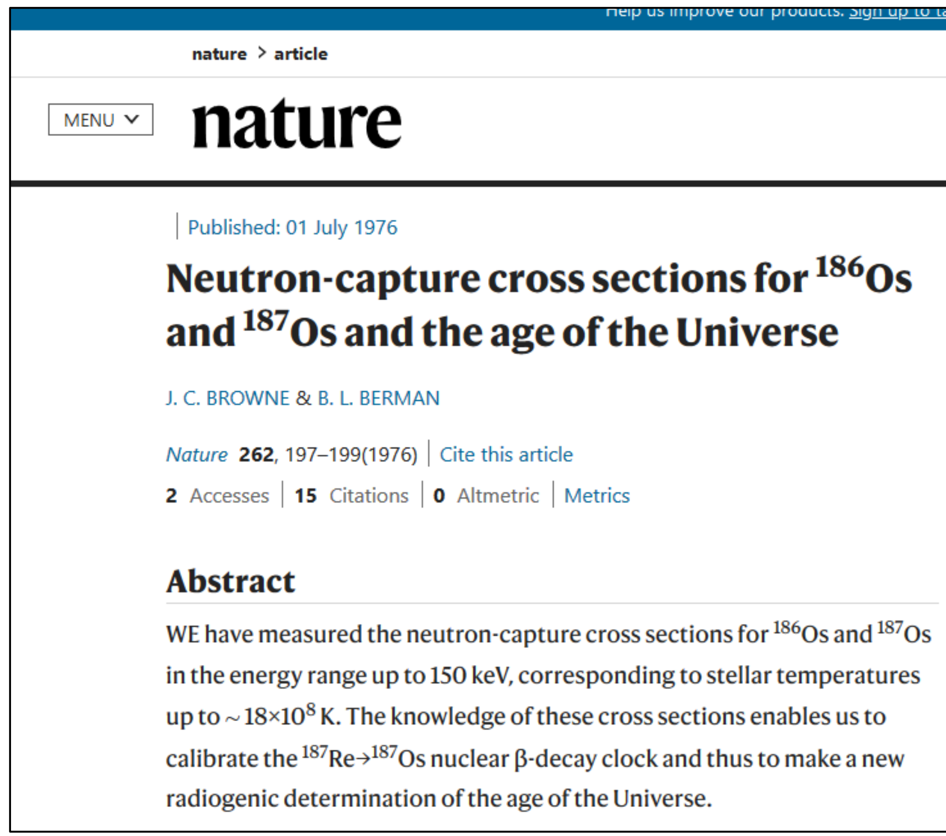
Who is using them ?

- All of us ?

- Neutron, gamma, charged-particles transport (Monte Carlo, deterministic)
- Reaction rates (energy, astrophysics, medical)
- Derived quantities based on neutron transport (decay heat, inventory, temperature)
- fuel/cladding performance,
- transient, safety parameters…

- Radiation shielding,

- Age of the Moon, …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Published: 01 July 1976

Neutron-capture cross sections for ^{186}Os and ^{187}Os and the age of the Universe

J. C. BROWNE & B. L. BERMAN

Nature **262**, 197–199(1976) | Cite this article

2 Accesses | **15** Citations | **0** Altmetric | Metrics

Abstract

WE have measured the neutron-capture cross sections for ^{186}Os and ^{187}Os in the energy range up to 150 keV, corresponding to stellar temperatures up to $\sim 18\times10^8$ K. The knowledge of these cross sections enables us to calibrate the ^{187}Re→^{187}Os nuclear β-decay clock and thus to make a new radiogenic determination of the age of the Universe.

Can we predict them ?

- No !!

Without experiments (*e.g.* n_TOF at CERN), no resonances can be predicted, no fission, capture, inelastic… can be accurately predicted.

- But, statistical rules exist (average quantities can be predicted), sometimes good enough (astrophysics), sometimes not (nuclear energy)

→ Phenomenological models are applied (good fitting models)

- Recently, with (very) large computer power, microscopic descriptions based on the n-n interaction produce some accuracy, and better predictions

→ Microscopic models can be applied (less parameters to fit)

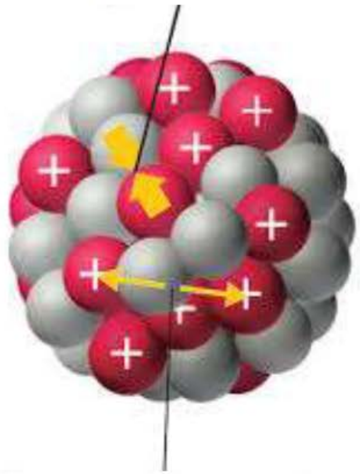
Can we predict them ?

A more « microscopic » description of the nucleus

e.g. Mean-Field

$$E_{MF} = \int \mathcal{E}_{nuc}(\mathbf{r})d^3\mathbf{r} + \int \mathcal{E}_{coul}(\mathbf{r})d^3\mathbf{r}$$

Strong nuclear force



Electrostatic repulsion

obtained on the basis of an Energy Density Functional generated by an effective n-n interaction !

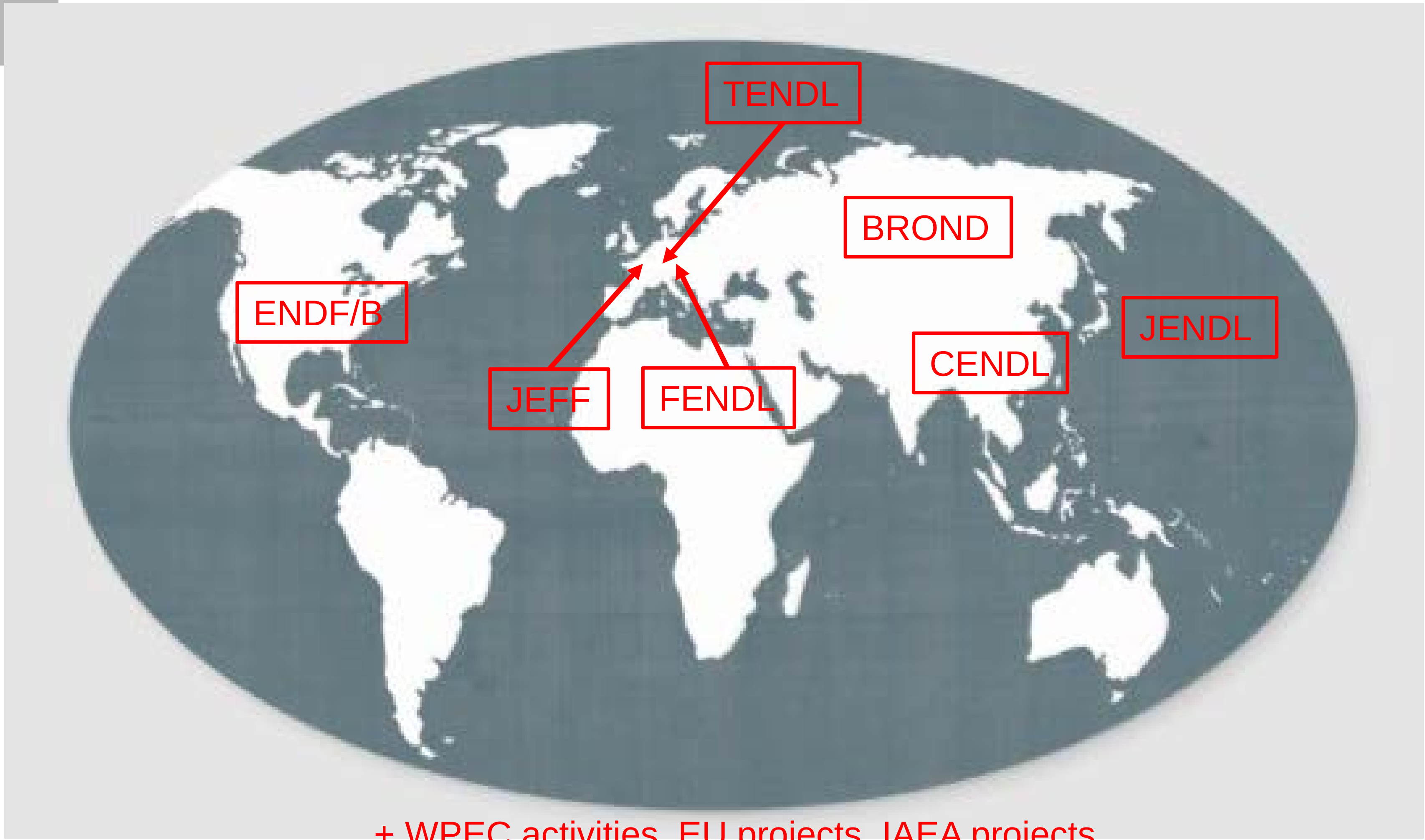
$$\begin{aligned}
\mathcal{E}_{\rm Sky} = & \sum_{q=n,p} \frac{\hbar^2}{2M_q}\tau_q + \frac{1}{2}t_0\left[\left(1+\frac{1}{2}x_0\right)\rho^2 - \left(\frac{1}{2}+x_0\right)\sum_{q=n,p}\rho_q^2\right] + \frac{1}{4}t_1\left\{\left(1+\frac{1}{2}x_1\right)\left[\rho\tau + \frac{3}{4}(\nabla\rho)^2\right]\right. \\
& \left. -\left(\frac{1}{2}+x_1\right)\sum_{q=n,p}\left[\rho_q\tau_q + \frac{3}{4}(\nabla\rho_q)^2\right]\right\} + \frac{1}{4}t_2\left\{\left(1+\frac{1}{2}x_2\right)\left[\rho\tau - \frac{1}{4}(\nabla\rho)^2\right] + \left(\frac{1}{2}+x_2\right)\right. \\
& \left. \times \sum_{q=n,p}\left[\rho_q\tau_q - \frac{1}{4}(\nabla\rho_q)^2\right]\right\} + \frac{1}{12}t_3\rho^\alpha\left[\left(1+\frac{1}{2}x_3\right)\rho^2 - \left(\frac{1}{2}+x_3\right)\sum_{q=n,p}\rho_q^2\right] \\
& + \frac{1}{4}t_4\left\{\left(1+\frac{1}{2}x_4\right)\left[\rho\tau + \frac{3}{4}(\nabla\rho)^2\right] - \left(\frac{1}{2}+x_4\right)\sum_{q=n,p}\left[\rho_q\tau_q + \frac{3}{4}(\nabla\rho_q)^2\right]\right\}\rho^\beta \\
& + \frac{\beta}{8}t_4\left[\left(1+\frac{1}{2}x_4\right)\rho(\nabla\rho)^2 - \left(\frac{1}{2}+x_4\right)\nabla\rho\cdot\sum_{q=n,p}\rho_q\nabla\rho_q\right]\rho^{\beta-1} + \frac{1}{4}t_5\left\{\left(1+\frac{1}{2}x_5\right)\left[\rho\tau - \frac{1}{4}(\nabla\rho)^2\right]\right. \\
& \left. + \left(\frac{1}{2}+x_5\right)\sum_{q=n,p}\left[\rho_q\tau_q - \frac{1}{4}(\nabla\rho_q)^2\right]\right\}\rho^\gamma - \frac{1}{16}(t_1x_1+t_2x_2)J^2 + \frac{1}{16}(t_1-t_2)\sum_{q=n,p}J_q^2 \\
& - \frac{1}{16}(t_4x_4\rho^\beta + t_5x_5\rho^\gamma)J^2 + \frac{1}{16}(t_4\rho^\beta - t_5\rho^\gamma)\sum_{q=n,p}J_q^2 + \frac{1}{2}W_0\left(J\cdot\nabla\rho + \sum_{q=n,p}J_q\cdot\nabla\rho_q\right).
\end{aligned}$$

Still *phenomenological*, but at the level of the effective n-n interaction
Obviously more complex, but models have now reached stability and **accuracy** !

Slide from S. Goriely, JEFF meeting, November 2019

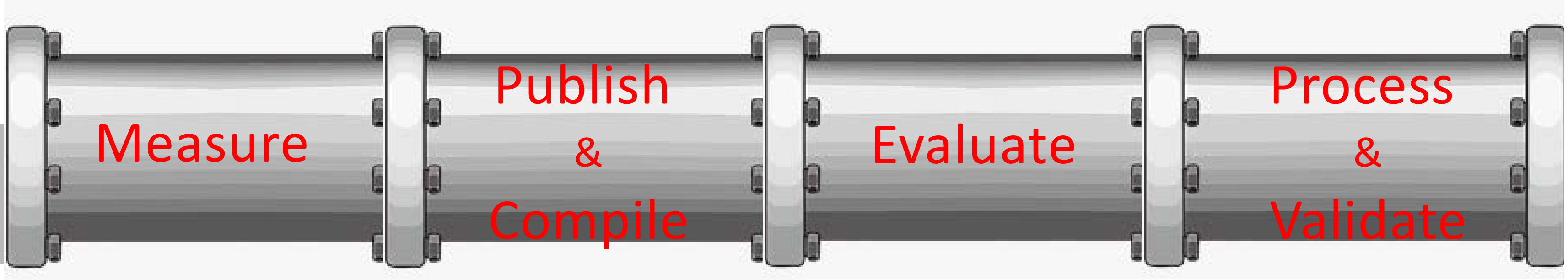
International efforts

- All evaluated nuclear data go into "nuclear data libraries": JEFF, ENDF/B, JENDL, CENDL, BROND, FENDL, TENDL… and many others

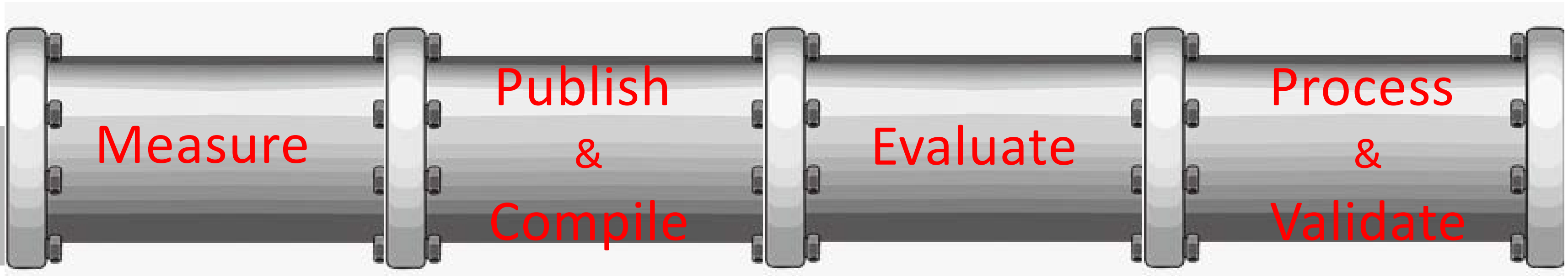


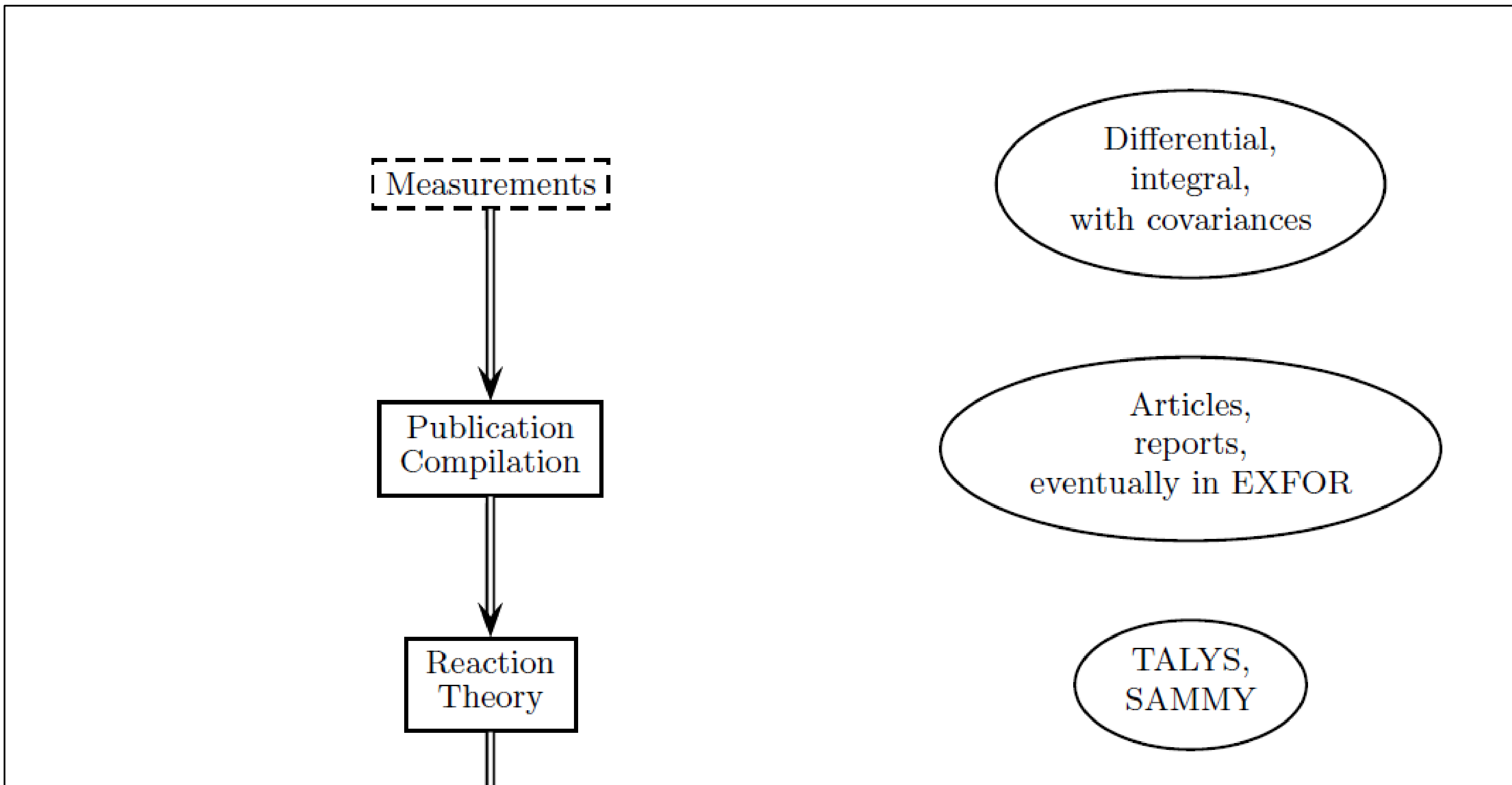
+ WPEC activities, EU projects, IAEA projects

Nuclear data lifecycle

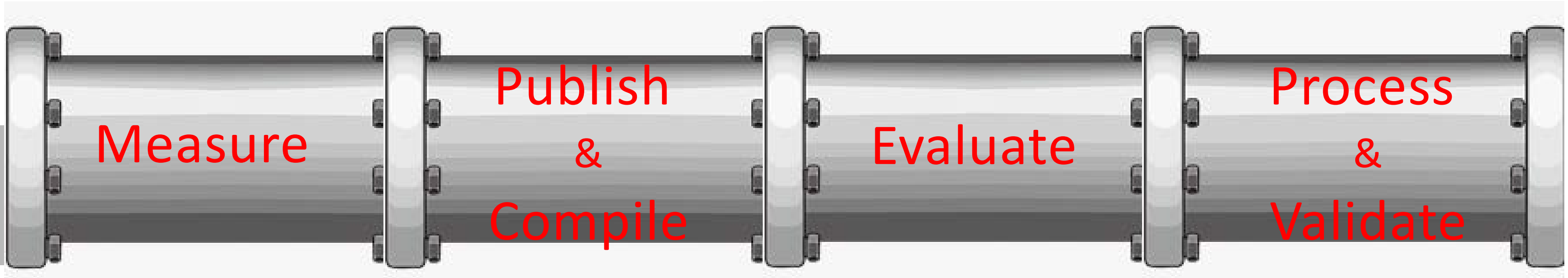


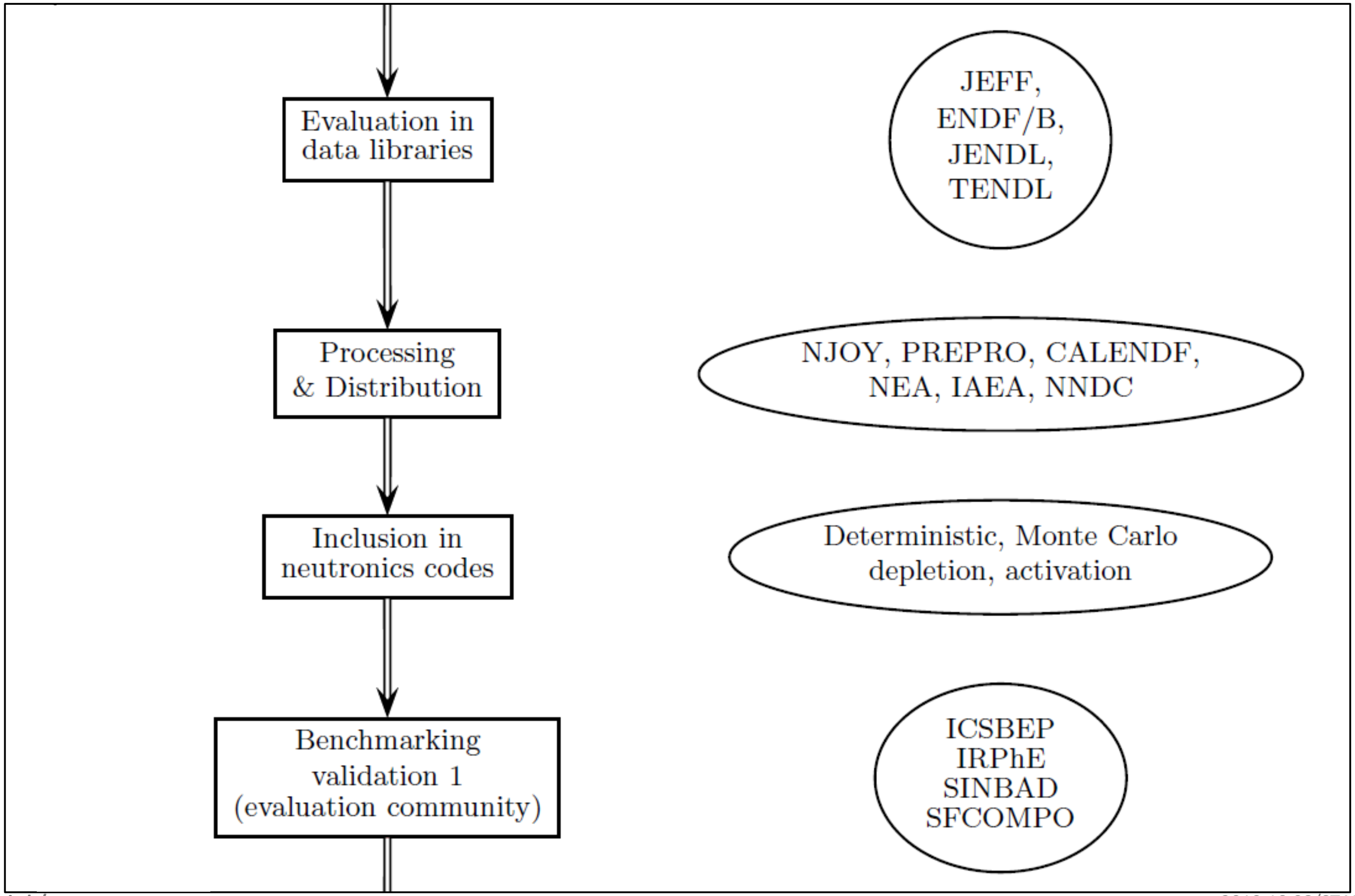
Nuclear data lifecycle



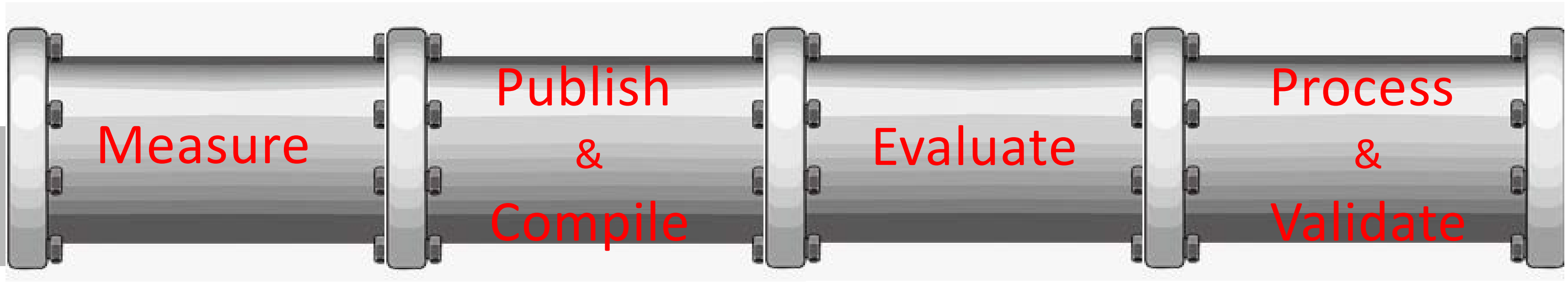


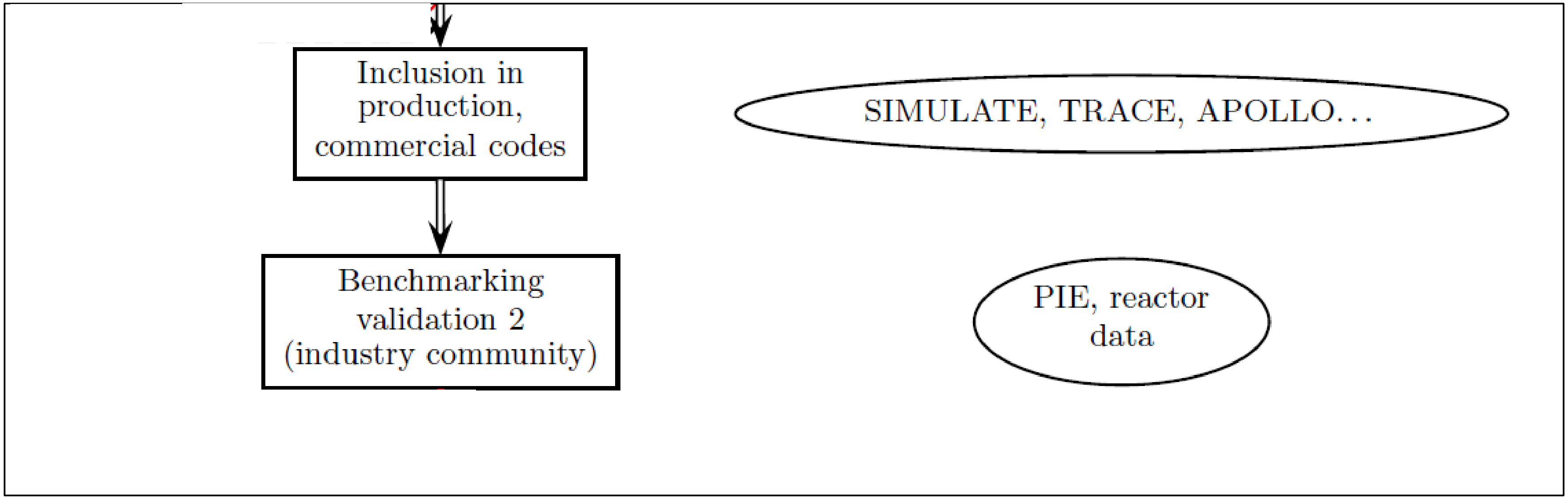
Nuclear data lifecycle





Nuclear data lifecycle





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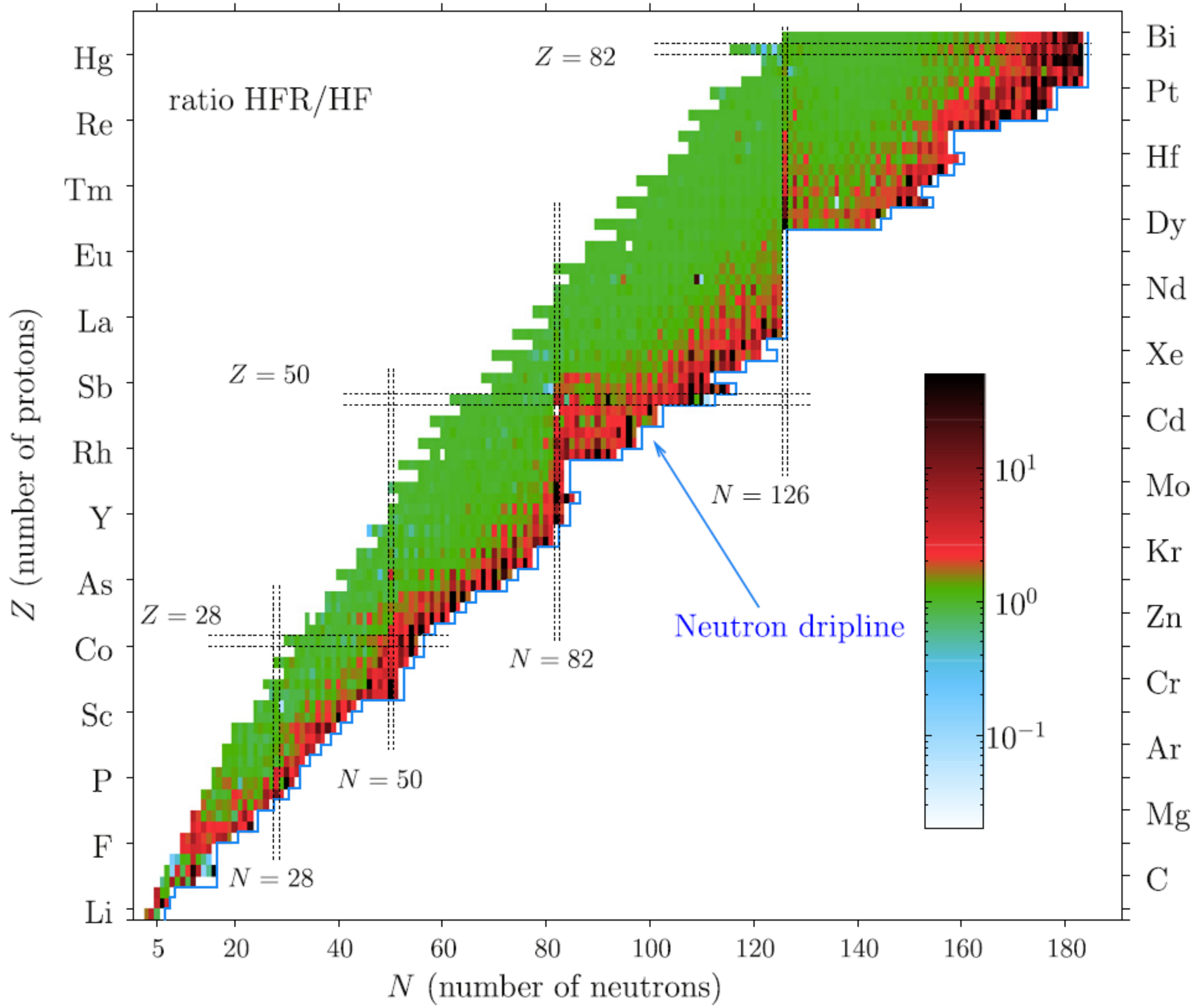
2. Application and examples

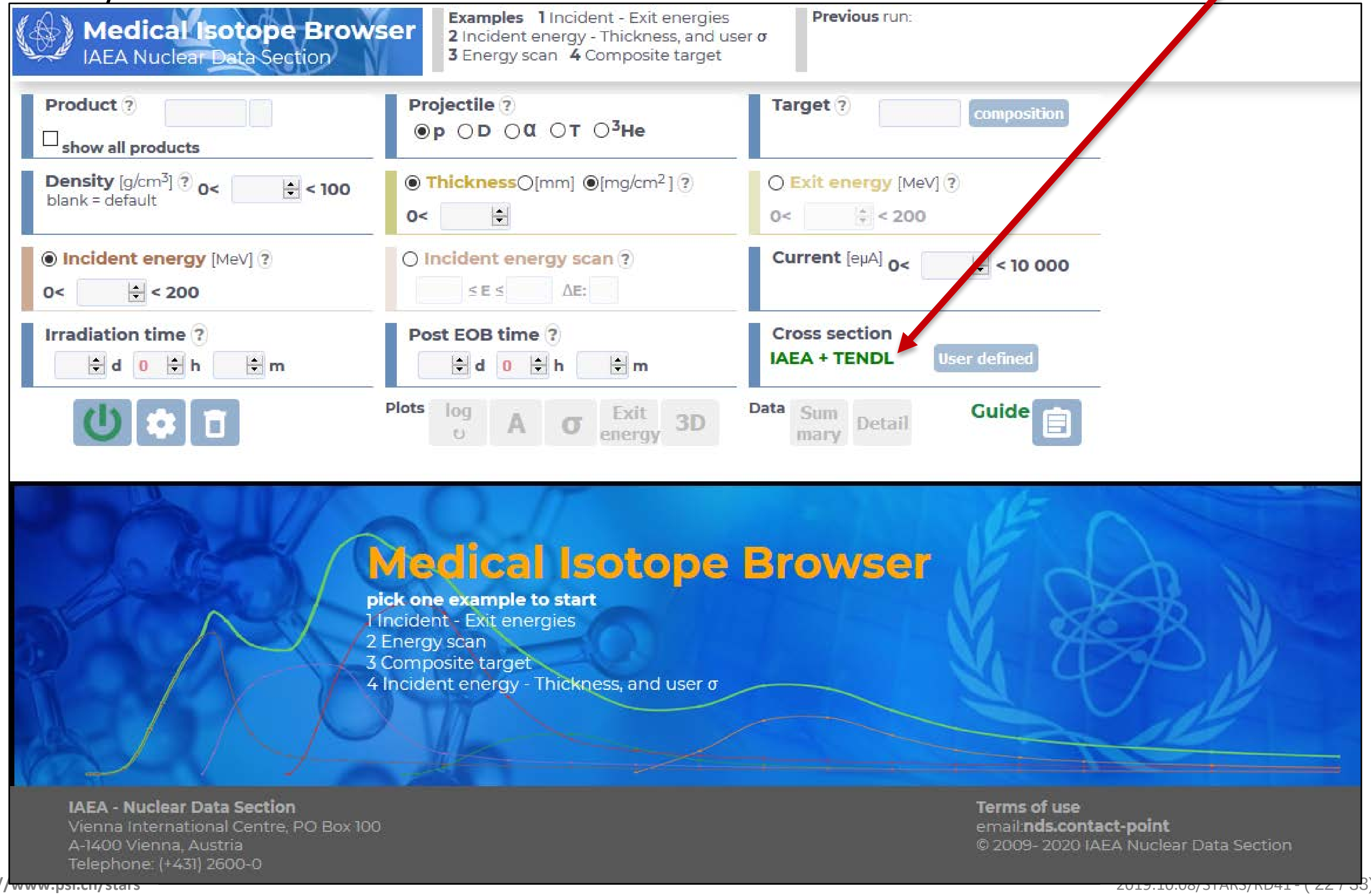
Application: in astrophysics

- Cross sections (or reaction rates MACS) are needed for 8000 isotopes (and isomers)
- Only ≈500 of them are (partially) experimentally known
- Impact of calculations (and models) is large, especially for extrapolation



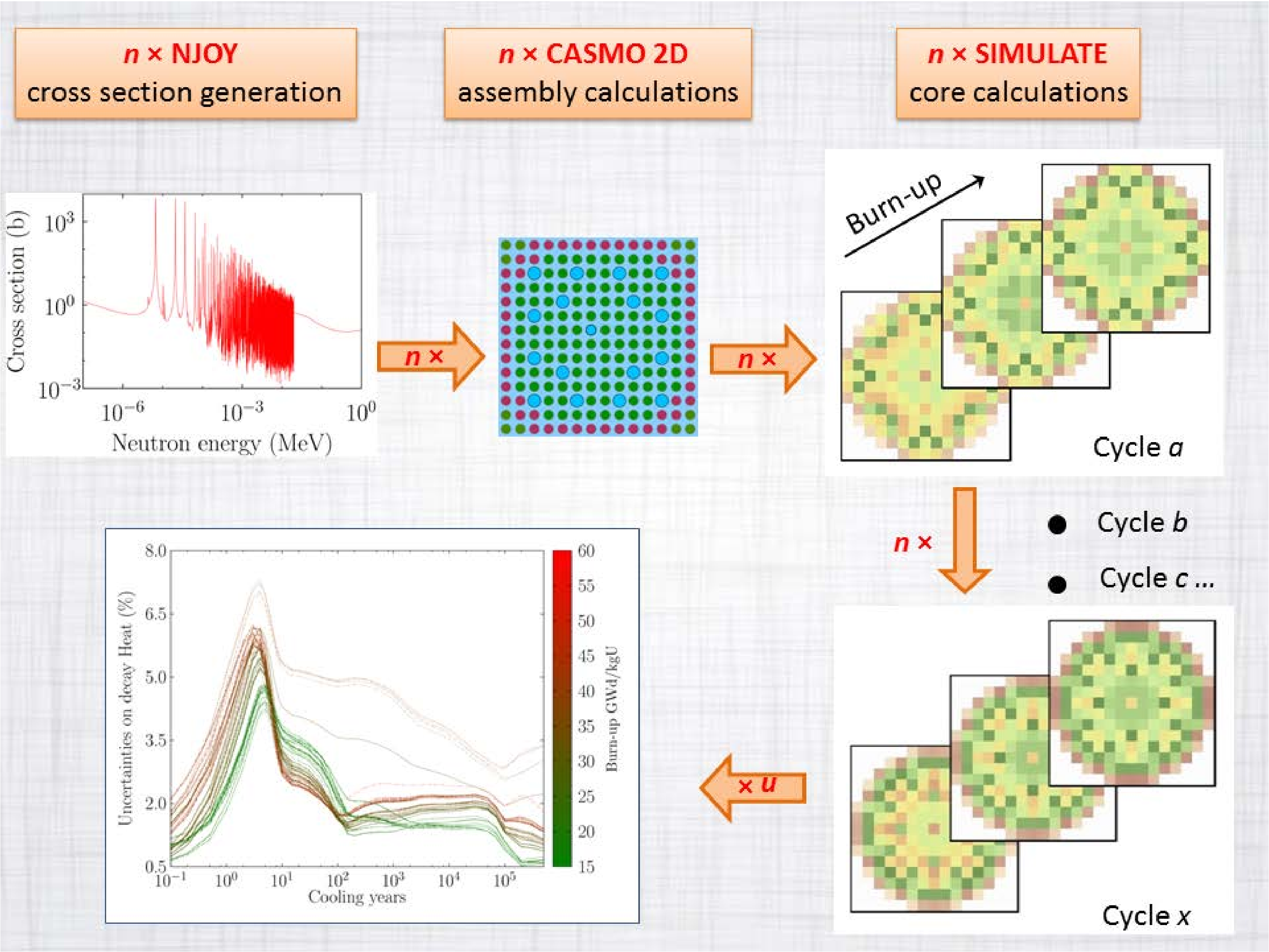
Medical application

- Many users in medical application rely on rate predictions based on nuclear data
- Last year, the IAEA has released a "Medical Isotope Brower" based on the TENDL library:



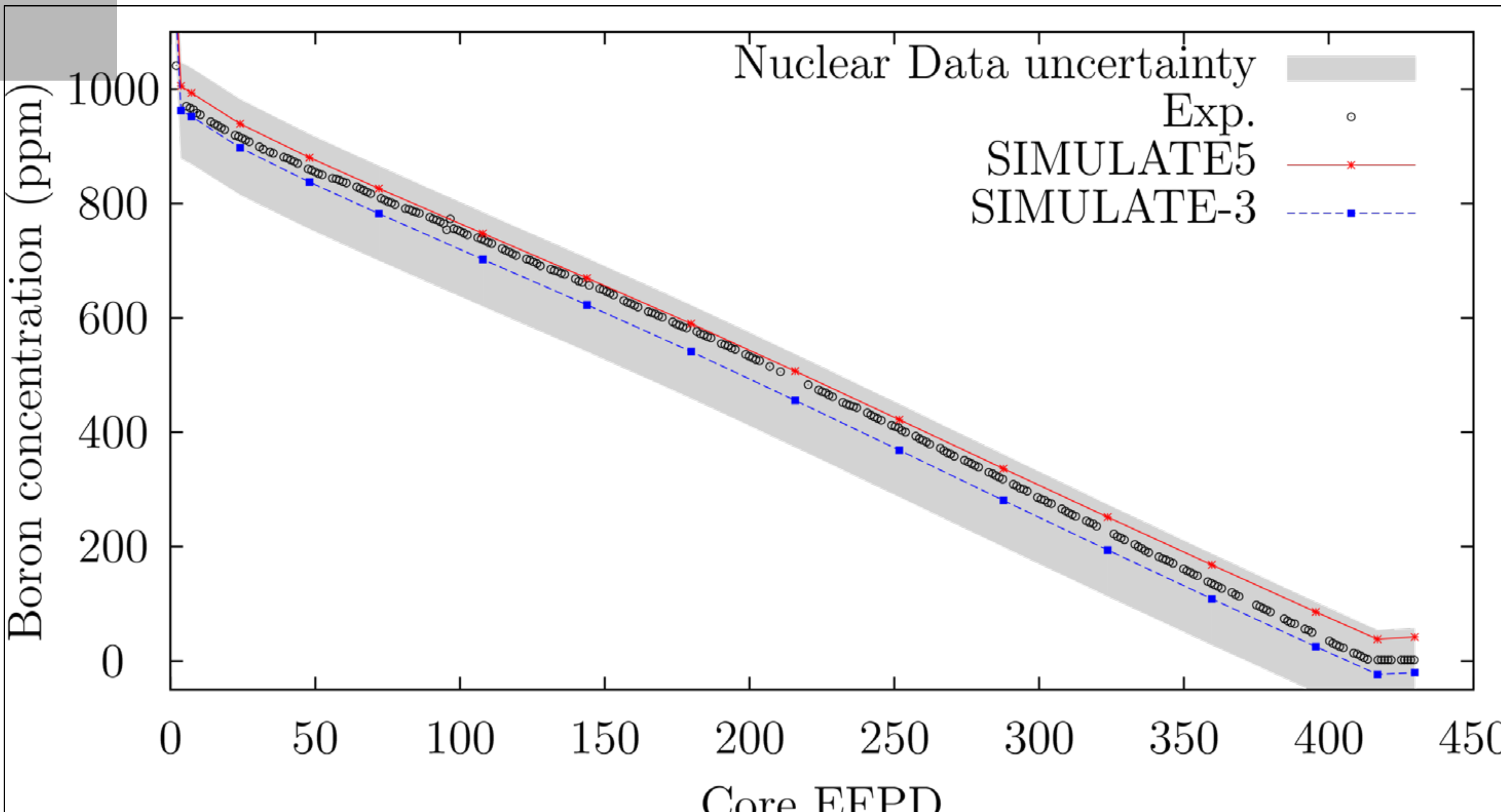
Application: Uncertainty in reactor/fuel

- Total Monte Carlo approach: random nuclear data for the full calculation chain.



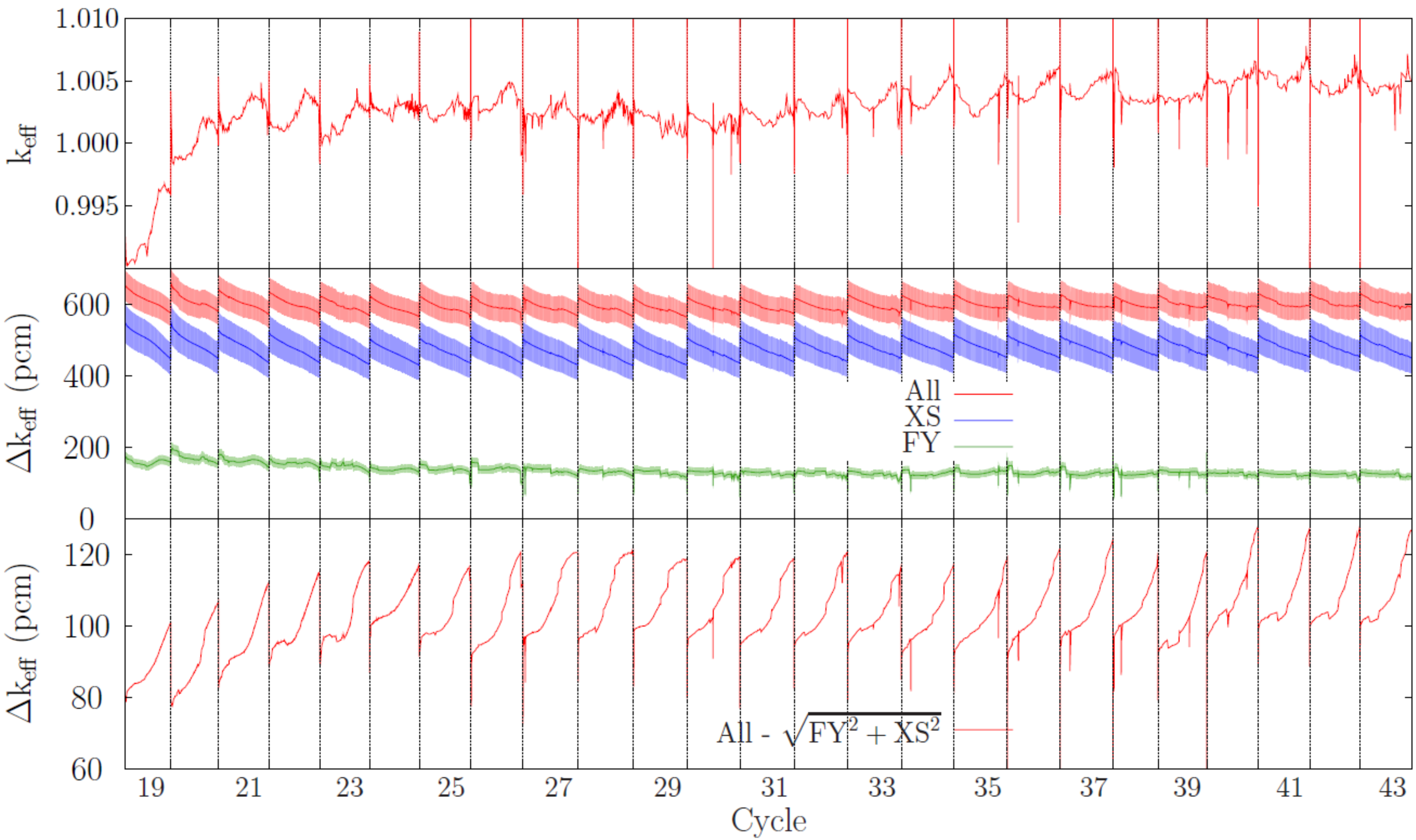
Application: in reactor physics and spent nuclear fuel (SNF)

- Example on a boron curve for real PWR cycles



Application: in reactor physics and SNF

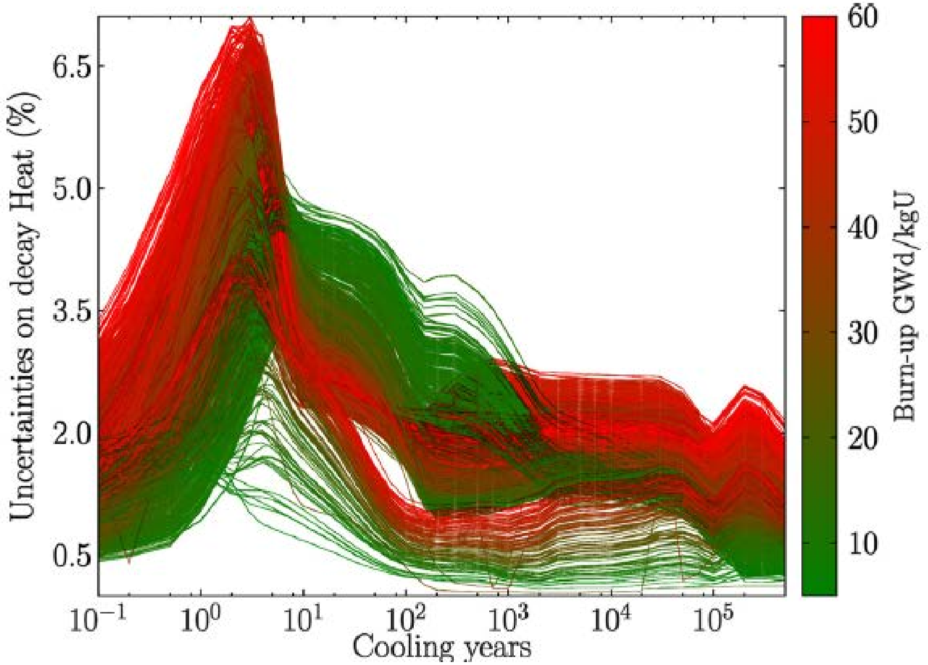
- Example on k_{eff} for real BWR cycles



Application: in reactor physics and SNF

- Example for the spent nuclear fuel (SNF) isotopic contents for BWR assemblies

	40 MWd/kgU	55 MWd/kgU
Isotope	uncertainty (%)	uncertainty (%)
^{235}U	1.9	4.0
^{238}U	0.02	0.04
^{239}Pu	2.2	2.7
^{242}Pu	3.4	3.8
^{243}Am	9.2	7.6
^{244}Cm	10	9
^{87}Rb	0.3	0.5
^{93}Zr	0.7	0.5
^{99}Tc	1.3	1.6
^{107}Pd	0.7	0.7
^{134}Cs	30	26
^{141}Ce	0.5	0.8
^{147}Sm	6.9	8.2



Other applications at PSI

- Nuclear data adjustments (S. Pelloni, LSM)
- Uncertainties (M. Hursin, EPFL)
- Measurements at CROCUS (V. Lamirand, EPFL)
- GEN-IV systems (LSM)
- Spallation & fusion (E. Alhassan LRT & GFA)
- DNBR (R. Mukin, LRT)
- Criticality-safety (A. Vasiliev, M. Frankl, LRT)
- Local effects in LWR core (M. Pecchia, LRT)
- Medical & Target preparations for n_TOF (D. Schumann, R. Dressler, J. Ulrich, LRC)

Applied Radiation and Isotopes 129 (2017) 96–102

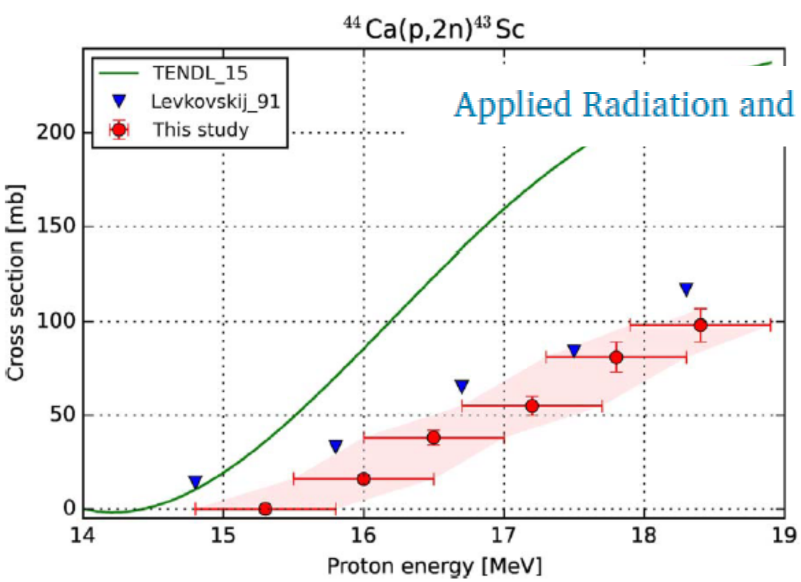


Fig. 6. Cross section of the reaction ^{44}Ca(p,2n) ^{43}Sc.

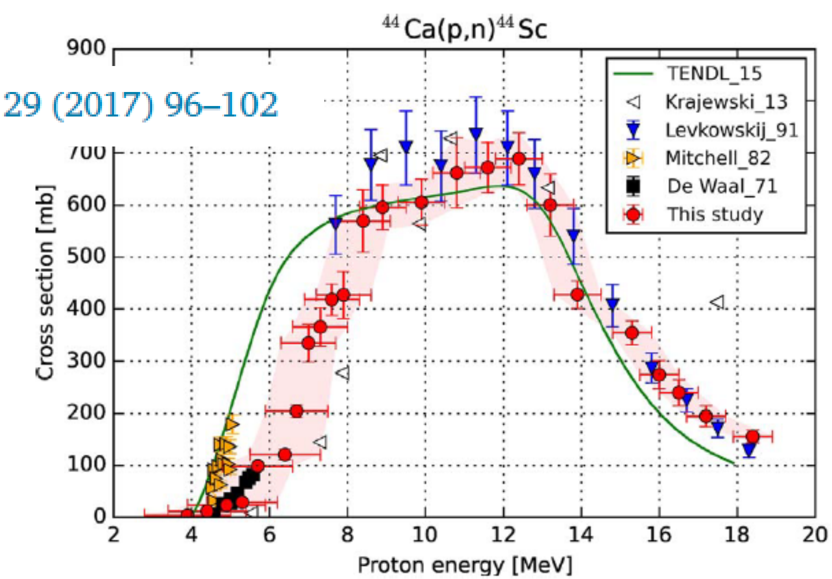


Fig. 8. Cross section of the reaction ^{44}Ca(p,n)^{44}Sc.

3. Current and Future developments

Better nuclear data

Example of k_{eff} as a function of decay time (actinide only)

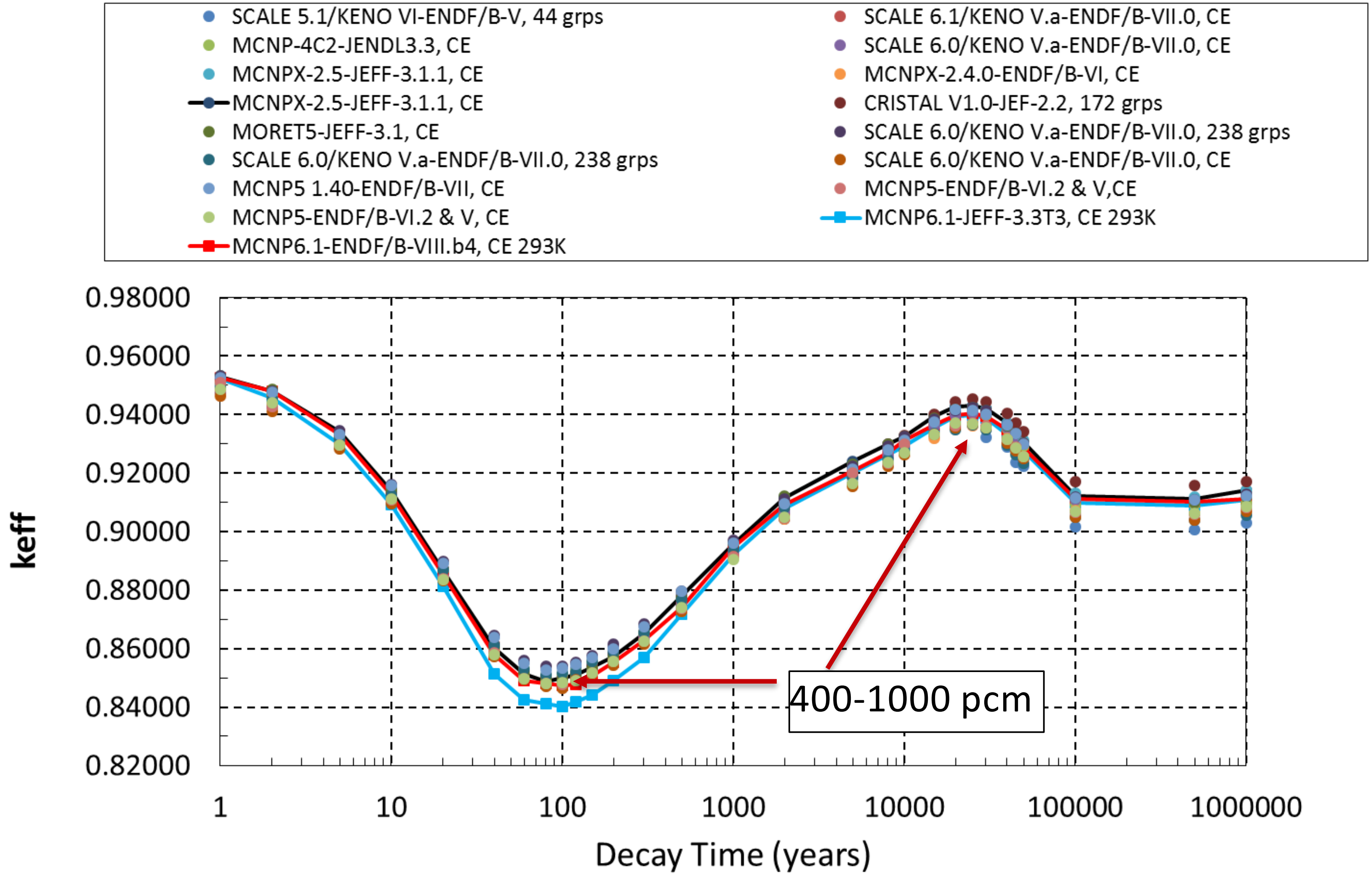


Figure from O. Cabellos, ICTP-IAEA, 2017

Better QA

Less “evaluators” worldwide

Loss of knowledge due to manual work, no reporting

+

Old nuclear data saying: “*manual work good, automation bad*”

=

Risk (and reality): repeating what was done before

Or worth: keeping everything unchanged to avoid breaking it

- Solution: Computer power/connection/ML is a push for
  - better structure, better automation
  - Machine readable databases, integrated calculation scheme

- The nuclear data community is slow to change, but
  - New horizon with automated approach (TENDL)
  - New OECD efforts within NSC/WPEC
  - Less evaluator worldwide: change is unavoidable

Better integration: from basic physics to spent fuel

- This is part of our efforts at the LRT: optimization of our resources
  - (1) link nuclear data to spent fuel (all core physics in one script): automation & integration of knowledge
  - (2) apply to all Swiss cores: systematization of work
  - (3) Calculate canister loading possibilities (k_{eff} and decay heat) with ML algorithms: optimization (currently performed by our Master student V. Solans)
  - (4) Calculate uncertainties and biases for canister loading: best estimate approach
- This opens possibilities for other applications (fuel accountability, inventory, additional reactors, saving on canisters, integration with LSM…)

Better measurements: back to the origin

- Because of our limited understanding of nuclear reaction mechanism, we still need measurements
- There is a need for better data (lower uncertainties, smaller biases)
- PSI has unique installations: isotope/target producer, intense neutron source
- Great combination for cross section measurements on small (nanograms) radioactive targets for fundamental and applied physics.
- SwissFel and its 3rd beam line is a unique opportunity !

Conclusions

- Everybody is using nuclear data, often without knowing it.

- The control from basic physics (nuclear data) to quantities of general public interest (nuclear waste) is a pledge for confidence and quality.

- A lot of work still needs to be done (if someone pays ?) to improve the prediction and the confidence interval

- Only four steps to summarize the whole concept: understanding, automation, systematization and optimization

- One challenging step: making the nuclear data ready for the computer area.

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Wir schaffen Wissen – heute für morgen