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Covariance analysis of $^{235}U(n_{th},f)$ independent and cumulative fission yields : propositions for JEFF4

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Abstract. The study of fission yields has a major impact on the characterization and understanding of the fission process and its applications. Fission yield evaluations represent the synthesis of experimental and theoretical knowledge in order to perform the best estimation of independent fission yields. Today, the lack of correlations between the different fission observables induces several inconsistencies in the evaluations. Different works proposed to estimate the correlations of the independent fission yields satisfying the consistency to the chain yield evaluations. Nevertheless, none of them introduces a prior correlations of the independent and chain evaluations in the evaluation process. Covariance matrix of fission yields depends on the evaluation method used according to the kinds of existing measurements. The consistency is deeply entangled to the statistical agreement between each dataset considering the covariance of measurements. Moreover, covariance of model parameters does not represent the only contribution to the evaluation covariance matrix. Thus, a new evaluation process is crucial to provide a complete and coherent evaluation file. The LEPh Laboratory of CEA Cadarache is developing this program for the future version of the JEFF-library.

1 Status on correlation matrix of fission yields

1.1 Previous analysis

1.1.1 Context and notations

Fission yields correspond to several observables describing the particles produced during the fission process. Starting from the scission point up to the decay products, we can cite:

- Fission fragment mass yields (before neutron emission) $Y(A^*)$
- Fission fragment isobaric distributions $P(Z|A^*)$ (before neutron emission)
- Neutron multiplicity $\upsilon(A^*)$
- Fission product mass yields $Y(A)$ (after neutron emission)
- Fission product isobaric distribution $P(Z|A)$ (after neutron emission)
- Fission product independent yields $Y(A,Z,m)$
- Isomeric ratio $IR(A,Z,m)$
- Kinetic energy distributions $P(E_k|A,Z)$
- Fission product cumulated yields $C(A,Z,m)$
- Chain yields $C(A)$

Historically, JEFF evaluations are based on experimental data associated with phenomenological models (Brosa [1], Zp model [2], Madland-England [3]) to extend the range of fission products up to the isotopes of interest for the applications. Thus, we find the same classification in the sorting of the experimental data. Some data are also model-dependent as, for instance, several chain yields where Zp model correction are applied. Up to now, the evaluations of the independent yields (after neutron emission) and the cumulative yields respect the conservation laws. Nevertheless the uncertainty analysis of each observable is based on the experimental data where they exist and model dependent for the extrapolated yields.

1.1.2 Correlation matrix of mass yields:

During the last decades, to fill the lack of a covariance matrix respecting the difference between the large independent yield uncertainties and the precise chain yields, several methodologies were proposed.
C. Devillers [4] proposed in 1977 a sum method of covariance in order to respect the precise chain yield uncertainties. This method is applied by K. Tsubakihara et al. [5] for the JENDL library (**Fig. 1**), L. Fiorito et al. [6, 7] for the JEFF one and T. Kawano et al. [8] followed by M.T. Pigni et al. [9] for the ENDF/B-VII.1 library. This method consists to determine the covariance

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elements between Y(A,Z,m) to obtain the variance of C(A,Z,m). This method assumes a null correlation between cumulative yields. Then, this only allows the filling of the lacks between independent and cumulative yields in order to respect the compatibility of the uncertainties of the two evaluations. Nevertheless, the hierarchy between the observables and the propagation of conservation laws is not properly defined.

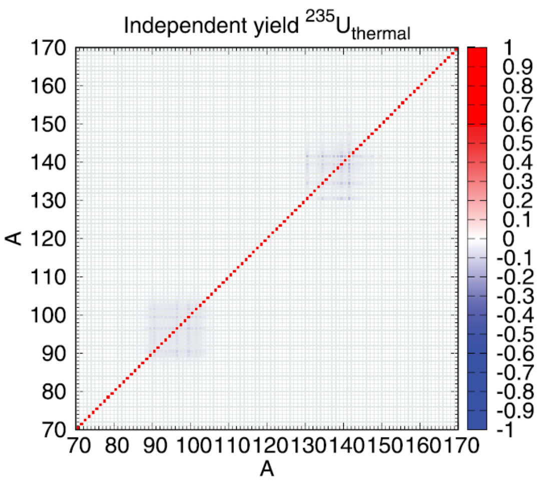

Fig. 1. Mass yield correlation matrix from K. Tsubakihara et al. (2021) [5].

In 2015, N. Terranova et al. [10,11] proposed a new approach considering the physical fission process to describe the covariance of the independent fission yields. Based on Brosa's model [1] and the Wahl parametrization of the Zp model [2] to describe the fission fragment distributions, the neutron emission yields $\upsilon(A^*)$ are adjusted in order to describe the JEFF-3.1.1 independent fission product yields [12]. The covariance matrix is obtained using the generalized perturbation theory. The cumulative yields are deduced from the previous calculations. Nevertheless, the calculated uncertainties of the chain yields are overestimated in comparison to those of JEFF-3.1.1. **Figs. 2 & 3** present the mass yield correlation matrix from the model parameter considering or not the prompt neutron yield uncertainties in order to observe the impacts of different model components.

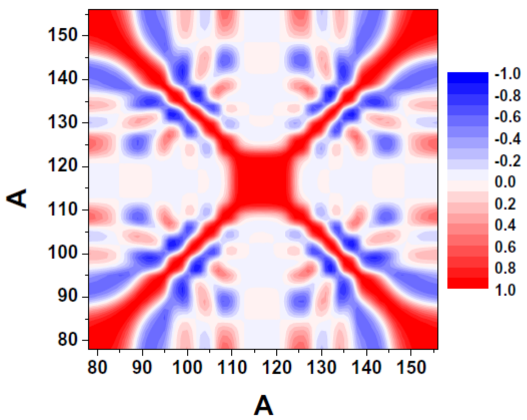

Fig. 2. Mass yield correlation matrix from CEA (2015) [10, 11] considering only the uncertainties of Brosa's fission mode parameters

The LOHENGRIN (ILL) collaboration on fission studies developed an experimental methodology to obtain absolute measurement of mass yields with associated experimental correlations (**Fig. 4**). There is not model input in this analysis and the correlation matrix come from the competition of the positive component of systematic uncertainties and the negative component due to the self-normalization [13].

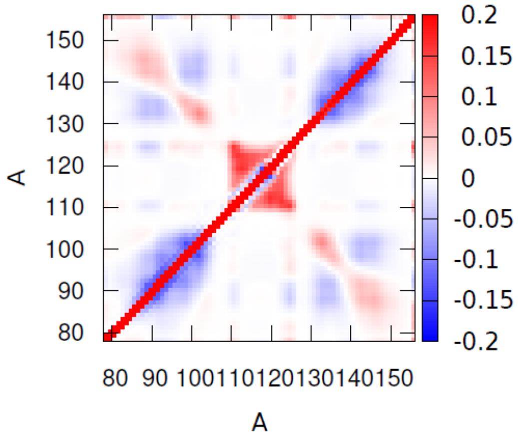

Fig. 3. Mass yield correlation matrix from CEA (2015) [10, 11] considering the uncertainties of Brosa's fission mode parameters and the prompt neutron yield uncertainties (saw tooth).

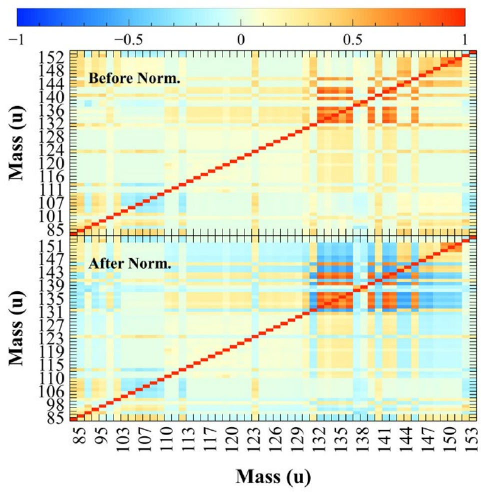

Fig. 4. (top) Experimental mass rate correlation of $^{233}U(n_{th},f)$ and (bottom) absolute yield correlation after self-normalization [13].

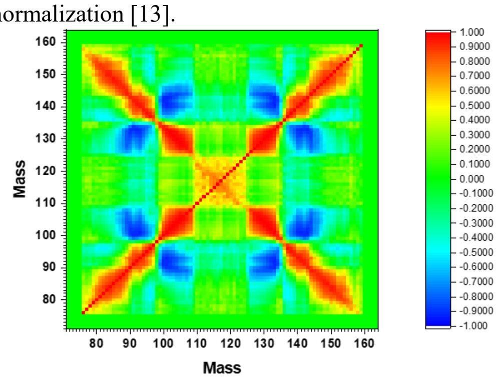

Fig. 5. K.H. Schmidt et al. GEF-2021/1.1 [14]

Another approach proposed by the NEA is the GEF modelling [14] based on a Brosa-like fission fragment description and many phenomenological parameters to describe a very large range of fissioning systems. A Monte Carlo code allows the generation of covariance of fission yields for one fissioning system (see **Fig. 5**) and between fissioning systems.

1.1.3 Correlation matrix of independent and cumulative yields:

A focus on the correlation matrices of independent fission yields according to the GEF model **(Fig.6)** and the Terranova's method (**Fig.7**) shows the same cross pattern but with important differences on the intensity of correlations. The analysis of L. Fiorito et al. [6, 7] is based on the sum rule to be consistent with the chain yield evaluation proposed by C. Devillers which structures the correlations matrix to go from about 5-10% independent yield uncertainty to 1-5% cumulative yield uncertainties (**Fig. 8 & 9**).

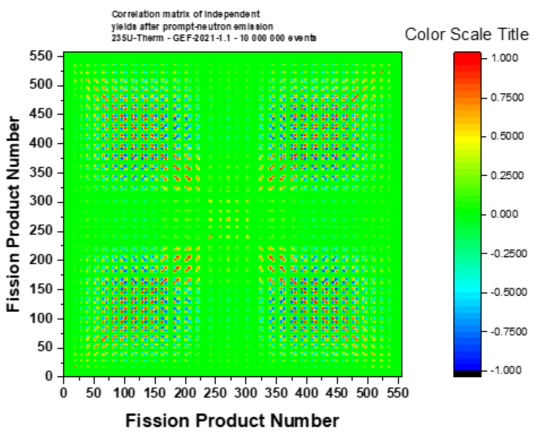

Fig. 6. Correlations of independent fission yields from the GEF-2021/1.1 code [14]

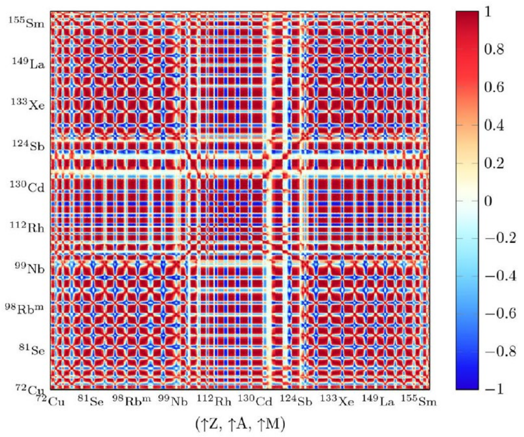

Fig. 7. Independent yield correlation matrix from CEA (2015) [10, 11]

1.1.4 discussion:

Thus, we observe that the structures are mainly due to the analysis methodologies. For the mass yields (post-neutron), a cross pattern comes from the mass modal model and the mass conservation law of the pre-neutron mass yields (see **Figs. 2 & 5**). When this stage of pre-neutron mass modeling is not introduce, the pattern of the correlation matrix presents only one diagonal as shown in **Figs. 1 & 4.**

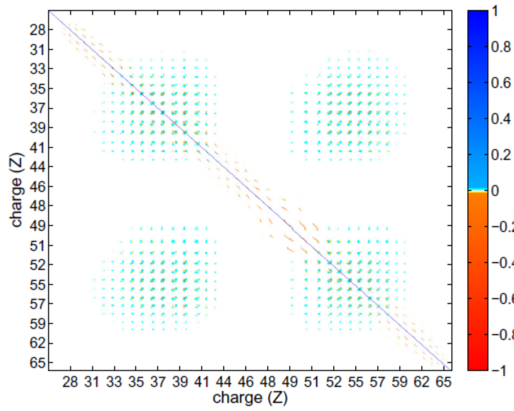

Fig. 8. Correlation matrix of independent yields proposed by L. Fiorito et al. [6, 7]

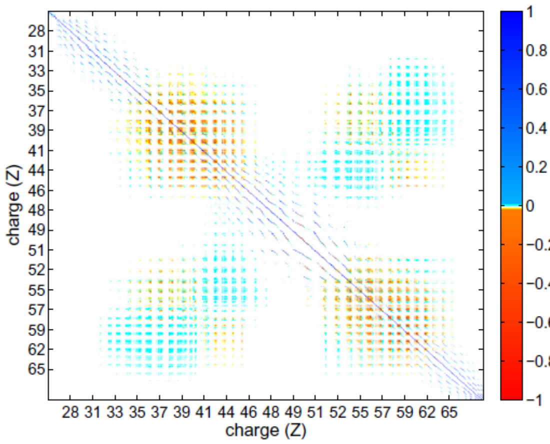

Fig. 9. Correlation matrix of cumulated yields proposed by L. Fiorito et al. [6, 7]

Adding the charge analysis, we observe a second cross pattern coming from the nuclear charge conservation law. This isotopic component of correlations can induce extreme correlations (see **Fig. 6 & 7**) when the component from Brosa's model (pre-neutron mass) and the components from conservation laws go in the same direction.

All of these works are based on partial evaluations of:

- mass, isobaric, isomeric (independent) yields,
- chain yields
- modeling of pre-neutron yields using Brosa's mode
- Wahl's systematics of charge polarization.

In all these studies, the weight of the model assumption is not necessarily clear.

1.2 New complete evaluation of ^{235}U(n_{th},f) proposed to JEFF-4

In the recent work [15], we proposed a new consistent evaluation of independent and cumulative yields with minimal impact from models. This analysis is based on the following decomposition of independent fission yields:

$$Y(A,Z,m) = Y(A) \cdot P(Z|A) \cdot IR(A,Z,m) \quad (1)$$

Where $Y(A)$ is the mass yield (post-neutron), $P(Z|A)$ the charge distribution and $IR(A,Z,m)$ the probability populating the metastable of the ${}^{A}_{Z}X$ nucleus.

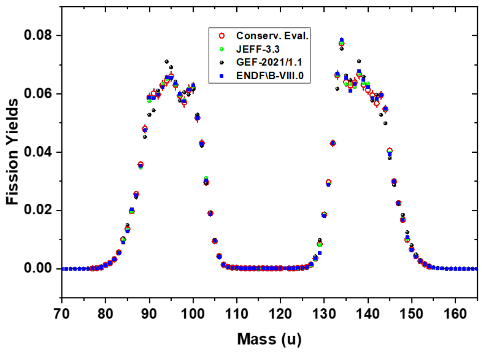

Fig. 10. Comparison of the conservative evaluation (see §2) of ^{235}U(n_{th},f) [15] with the GEF model [14] and the JEFF-3.3 [12] and ENDF\B-VIII.0 evaluation [19].

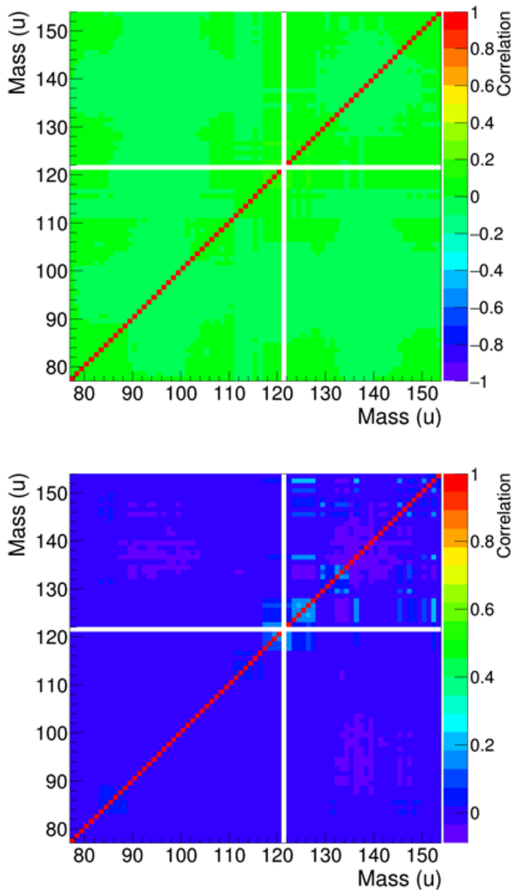

Fig. 11. (top) Mass yield correlations from of ^{235}U(n_{th},f) [15] (bottom) zoom on the structures, principally on the heavy mass region.

For ^{235}U(n_{th},f), the EXFOR database [16] is sufficiently complete to perform a mass yield evaluation without any model input. We obtained a new evaluation of mass yields which is in agreement to the JEFF-3.3 evaluation [12] (**Fig. 10**). The rigorous statistical analysis [15, 18] allows us to propose a synthesis of experimental data converging to quasi-null correlations of mass yields (see **Fig. 11 & §2**). Consistently to previous discussion (see §1.1.4), we observe a single diagonal pattern due to absence of mass number conservation law (at post-neutron stage) but a negative component due to the self-normalization step (**Fig. 11, bottom**).

For the isotopic yields, at this moment, we based our analysis on the JEFF-3.3 library considering this work as a synthesis of experimental isotopic rates completed by the GEF model in the tails of distributions. We apply the conservation laws and the self-normalization process in order to provide the correlation matrix of isotopic and isomeric distributions. As expected, the charge conservation law induces a cross pattern (no proton emission in thermal neutron induced fission). The self-normalization regulates the correlations in order to respect the sum rule:

$$Var[Y(A)] = \sum_{Z,m} Var\, Y(A,Z,m) + 2 \sum_{\substack{Z<Z' \\ m<m'}} Cov\big(Y(A,Z,m), Y(A,Z',m')\big) \quad (2)$$

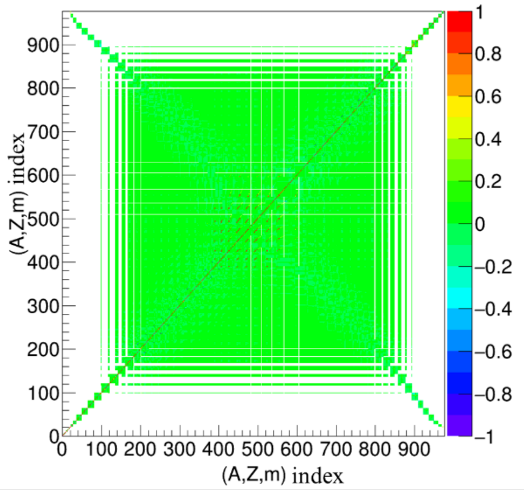

Fig. 12. Independent fission yield correlation matrix of ^{235}U(n_{th},f) [15].

To take into account the chain yields in the mass yield analysis, we have to calculate the $C(A,Z,m)$ cumulative yields from the independent yields. These observables are calculated using the decay data defining the Q matrix:

$$C(A,Z,m) = \boldsymbol{Q}\, Y(A,Z,m) \quad (3)$$

With:

$$Q = (\boldsymbol{I} - \boldsymbol{B})^{-1} \quad (4)$$

Where $\boldsymbol{I}$ is the identity matrix and $\boldsymbol{B}$ is the matrix of decay corresponding to the list of (A,Z,m) isotopes. The $\boldsymbol{B}$ matrix is deduced from the JEFF-3.3 decay data file [12]. Thus, at the end of the process, we generate the evaluations of both independent and cumulative yields in a global process with complete and consistent correlation matrices (**Figs. 12 & 13**).

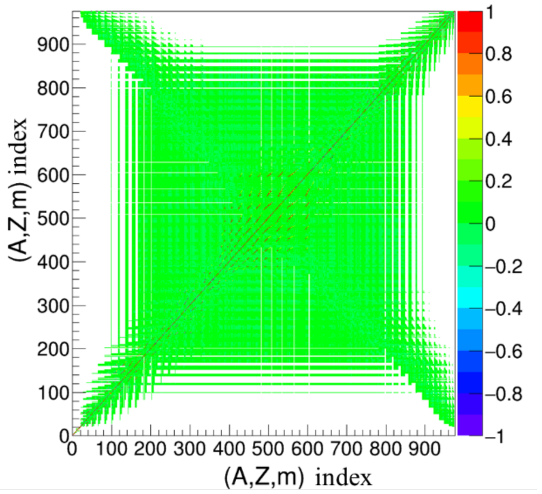

Fig. 13. Correlation matrix of cumulative yields of $^{235}U(n_{th},f)$ [15].

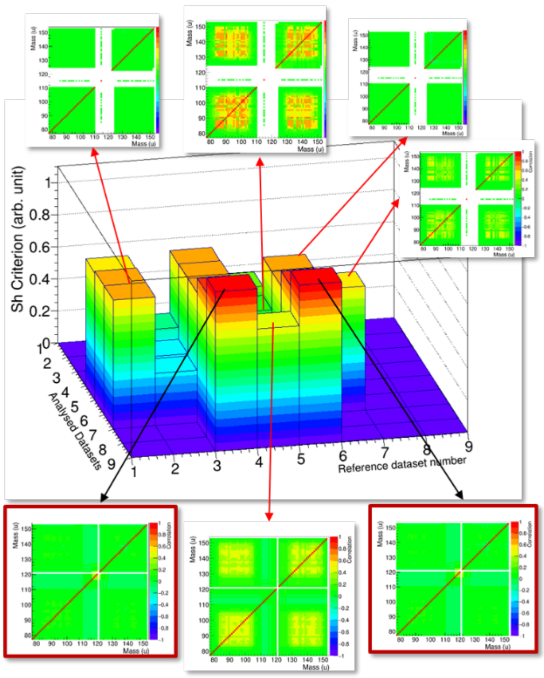

Fig. 14. Histogram of Shannon entropy criterion as a function of the reference dataset and the number of datasets used. The extrema are illustrated with different correlation matrices of evaluated mass yields (White bands correspond to the unevaluated mass yields).

2 Multiplicity of mass yield evaluations

The $^{235}U(n_{th},f)$ mass rate measurements are sufficiently complete to cover the fission product mass range (> 99.5%) to ensure the self-normalization of the fission yields. Nevertheless, the dataset provided by experimentalists are not in agreement and a cross-normalization of fission rates per mass is required [17]. We have shown that the use of one reference measurement to cross-normalized all the datasets (before absolute normalization) introduces different results according to the reference dataset. **Fig. 14** illustrates the dependence in the choice of reference dataset that maximizes or minimizes the information provided. The quantification of information is deduced from the Shannon entropy [18]

This result corresponds to non-ergodic analysis paths. The second limit of this process is that the tested mass rates are reduced to the overlapping mass rates (white cross). Thus, in order to solve this non-ergodic analysis, we have developed a new methodology.

2.1 Several analysis approaches

The test and the sorting of datasets are the key point to obtained mean values per mass representing the data selected. Considering that the experimental information is reduced to mean values and the standard deviations for the majority of datasets, we assume a Gaussian distribution for each mass yield (per experiment). Then, the maximum of likelihood method is equivalent to the minimum of the generalized least square χ_g^2.
The analysis is performed in two steps. First, we compute the mean value for each mass by using all the measurements. Then we test if the evaluation based on these mean values is compatible at 3σ with all the datasets used. If the test fails, a regularization is necessary. In this work, we have developed 4 different regularization. The two global ones are:

- Strict solutions: the sorting of mass measurements follows the main contributors χ_g^2 up to obtain an acceptable χ_g^2, i.e. with a $P-value > 1-CL$ [20]. Different sorting of data are possible and we kept the two possible solutions, called "strict-1" and "strict-2";
- Conservative solutions: we apply the same statistical test after adding an uncertainty of 2.5% for all masses to obtain an acceptable $P-value$ (See [15]). It turns out that the measurements of the mass 153 have huge discrepancies. Therefore we decided to have two different evaluations with two values of the mass 153 with an additional uncertainty of 2.5%. The two solutions are called: "conservative 1" and "conservative 2";

The two local regularizations are:

- Conservative Student solution: for mass where measurements disagree, we consider additional uncertainty per mass based on Student law standard deviation.
  - Conservative Best Estimated (B.E.) solution: we add adjusted uncertainties per mass to obtain acceptable χ_g^2 test.

By definition, only the two last solutions preserve all the initial datasets. In the large majority of masses, the mean values are very insensitive to the regularization methods except for incoherent datasets (A= 153, see effect §3). Conversely, standard deviations of mean values are very sensitive to the different approaches. **Fig. 15** presents the relative uncertainties according to the regularization methods. In comparison to the results presented in [15], the two last regularizations are new additional calculations. We observe that the uncertainty of the Best Estimated (B.E.) method or the Student approach are very similar and included between the conservative and strict methods. These two local regularizations produce the most realistic solution considering all the data adjusted on the database.

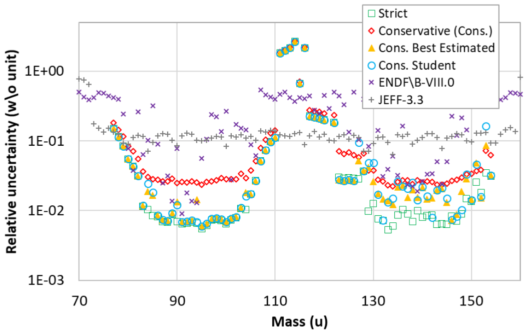

Fig. 15. Relative uncertainties of the different solutions as a function of mass in comparison to current evaluations.

2.2 Quantification of information provided by correlations

The output quantities of the evaluations are the mean values, the standard deviations and the correlations. We have seen that the mean values of different solutions are stable. Nevertheless, the standard deviations are dependent on the regularizations of datasets. In this part, we discuss the sensitivity of the correlations to the different evaluations.

Table 1. Shannon entropies associated to the different evaluations.

Regularizations	Shannon Entropy
Strict Eval.	6,222
Strict Eval. + Syst. 1%	6,228
Strict Eval. + Syst. 2%	6,233
Strict Eval. + Syst. 3%	6,236
Conservative Eval.	6,240
Cons. Eval. + Syst. 1%	6,238
Cons. Eval. + Syst. 2%	6,236
Cons. Eval. + Syst. 3%	6,233
Best estimated Eval.	6,222
Student Eval.	6,209

In our analysis tools, we are able to integrate the available experimental correlation matrices and to deduce the dependence of the evaluated correlation matrix to the lacks of experimental correlations, testing the effects of additional systematic uncertainties. We conclude that the evaluated uncertainties increase of 1% for 3% systematic uncertainty added. This effect is due to the cross-correlation between datasets in the renormalization process.
To compare the correlation matrices of all analyses, we have calculated the Shannon entropy associated to the different correlation matrices [18] by considering the previous kinds of regularizations. Table 1 presents the Shannon entropy calculations for the ten following cases. The information quantity distributed is very stable in contrary to the previous method using a reference dataset (**Table 1**).

2.3 Probability density functions (PDF) of fission rates

The evaluation of fission yields involves mixing of fission rates from different datasets assuming a Gaussian distribution for each measurement. The generalized least squares method (χ_g^2) is equivalent to the maximum likelihood method for Gaussian distributions associated to measurements. If there is no bias, this method leads to a minimal variance (Cramer Rao theorem) and the covariance matrix corresponds to the Fischer matrix associated to data [20].

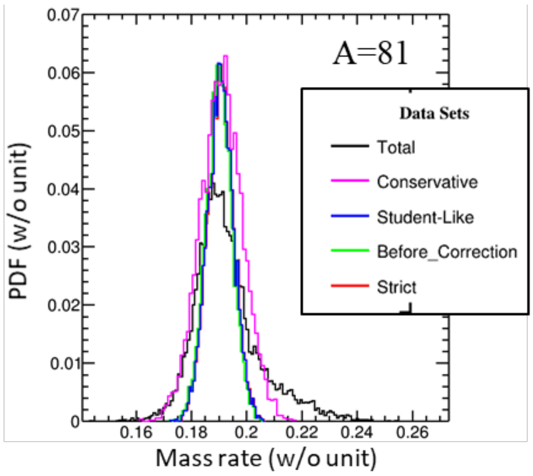

Fig. 16. PDFs of evaluated mass rates before absolute normalization for an acceptable $\chi_g^2(A)$ test.

Then, the mean value of data (maximum likelihood) is also distributed according to a Gaussian distribution. To test this property, we developed a Bayesian Monte Carlo (BMC) considering the different approaches and the empirical mean calculation to illustrate the PDFs of the evaluated values. **Fig. 16** presents the PDF for a regular dataset (A=81 u) and **Fig. 17** for an un-regular dataset (A=84 u). In the first case, the “Strict”, “Student-like” and “without regularization” are similar. The empirical mean value is not the minimal variance estimator and the conservative assessment take into account a penalty to obtain an acceptable χ_g^2 test for all datasets. In the second case (**Fig. 17**), when the $\chi_g^2(A)$ test is not acceptable, we see clearly the bias in the mean values according to the regularization methods applied. In particular, we see that the mean value without any regularization underestimates the uncertainty for the dataset selected and presents a bias in comparison to the strict method. The Strict method applies a sorting on available data which generates two solutions for this mass A=84. The conservative method corresponds to a Gaussian reduction method for an incompatible initial dataset, hence the name used.

These BMC calculations illustrate that the most accurate assessment naturally corresponds to the selection of compatible data and leads to a Gaussian PDF associated to the mean rate.

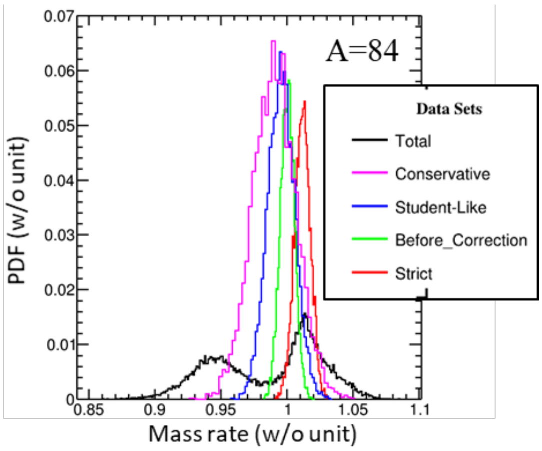

Fig. 17. PDF of evaluated mass rate before absolute normalization for a rejected $\chi_g^2(A)$ test.

2.4 Exclusion diagram

The comparison of the mean values of the different proposed evaluations is illustrated in **Figs. 18 & 19**. We see a general good agreement for the principal masses. Nevertheless, few mass yields of JEFF-3.3 and ENDF\B-VIII.0 evaluations are excluded for a confidence level of 99.7%. The limit of this plot is not to consider the correlation matrix. The P-values take into account the correlation matrices and have been presented in ref. [15].

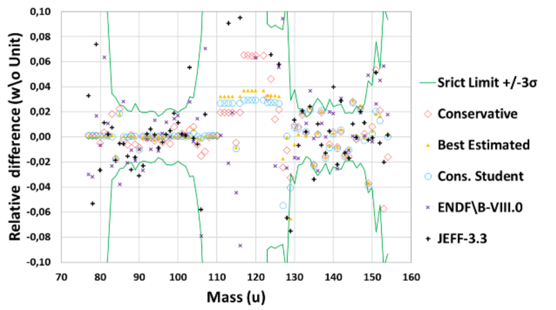

Fig. 18. Relative difference of available evaluations in comparison to the strict one. The green lines correspond to the limits for 99.7% confidence level.

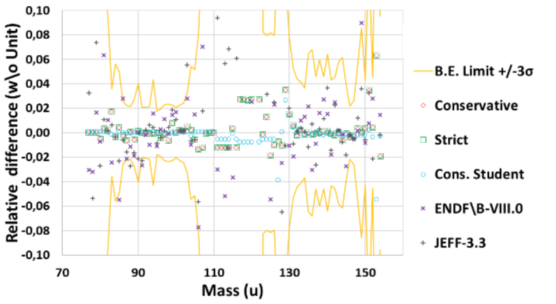

Fig. 19. Same plot as the previous one but in reference to the B.E. evaluation. The yellow lines correspond to the limits for 99.7% confidence level.

3 Cumulative yields

In this analysis, we determined the mass yields considering the experimental data on fission products and chain yields in the same analysis process (see § 1.2 and [15]). Thus, the correlation matrix of chain yields is coherent with those of independent yields.
For the conservative analysis, we obtain a limit of precision of 2.5 % in the best cases compared to about 1% accuracy in JEFF-3.3 (**Fig. 20**). Note that the independent yields and the chain yields of JEFF-3.3 are two different evaluations whereas in our analysis both observables are extracted from a single analysis. **Fig. 21** presents the relative uncertainties of the chain yields for the strict evaluation. We find the same precision level as JEFF-3.3. The process used in this work allows us to claim that the strict evaluation is the best estimation of the mass yields assuming no bias in the chain yields. **Figs. 22 & 23** present selected chain yields for current evaluations. The mass A=153 illustrates the impact of the sorting of data to obtain an acceptable statistical compatibility test of datasets. For this mass, two groups of two measurements exist. The recursive process of renormalization, test, sorting of data, renormalization, test (etc.) according to the major contributor of χ_g^2 generates several compatible data groups providing the different evaluations.

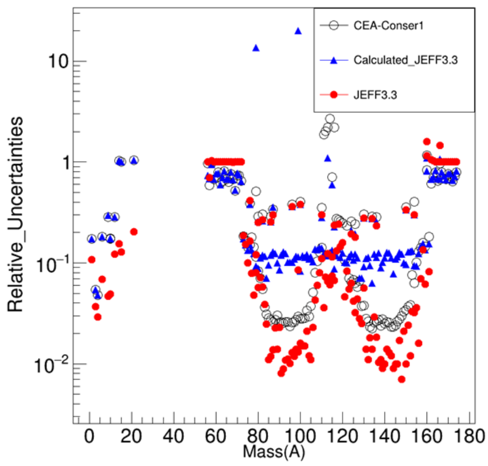

Fig. 20. Relative uncertainties of chain yields as a function of mass for the "conservative 1" evaluation.

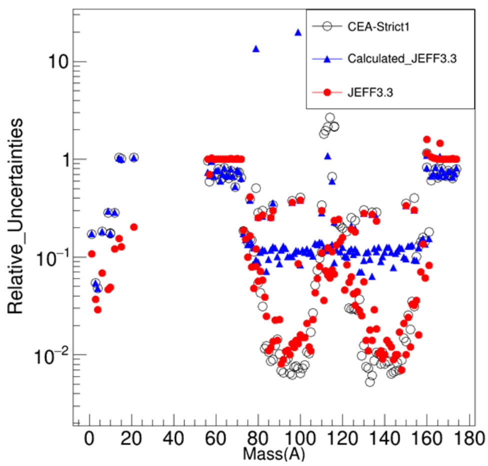

Fig. 21. Relative uncertainties of chain yields as a function of mass for the "Strict 1" evaluation.

4 Impact on reactivity calculations

A first calculation of the impact of the new fission yield evaluations concerns the reactivity effect on pin fuel calculations. Calculations are achieved with the Apollo2 [21, 22] code from CEA. Table 2 synthesises the reactivity calculations for two different burn up values. **Figs. 24 & 25** show the main contributors to the variation of reactivity in reference to the calculation with JEFF-3.3. These slight variations are due to the compatibility of our evaluations with the JEFF-3.3 one.

Table 2. Variation of reactivity in reference to JEFF-3.3 calculations.

	Δρ [pcm]	
Burn up (GWd/t)	1.5	50
CEA_Strict-1 /JEFF-3.3 -1	-58	24
CEA_Conservative-1 / JEFF-3.3 -1	-8	9

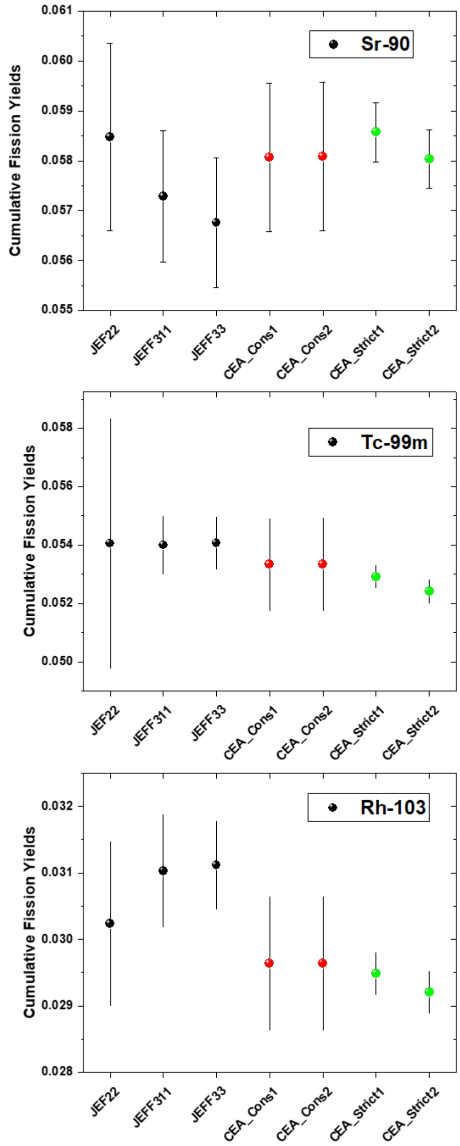

Fig. 22. Comparison of current evaluations for three important light chain yields.

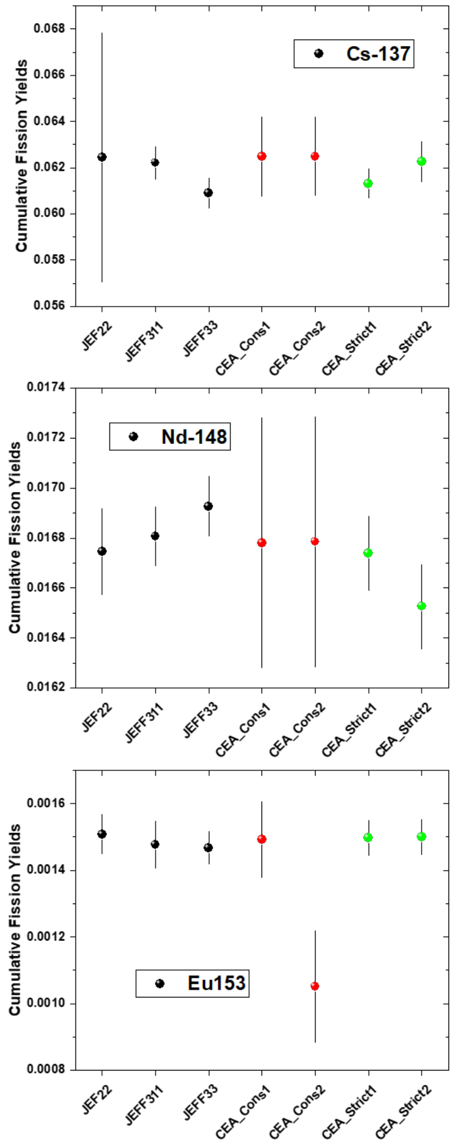

Fig. 23. Comparison of current evaluations for three important heavy chain yields.

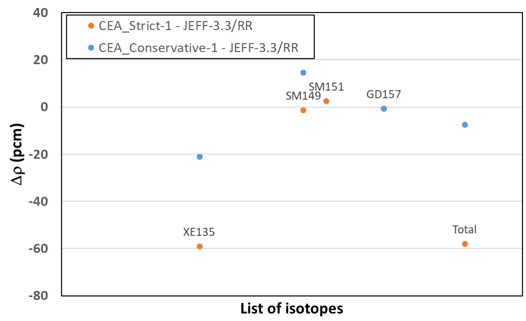

Fig. 24. (Preliminary results) Variation of reactivity for the "Strict 1" and "Conservative 1" FY evaluations in reference of the JEFF-3.3 for a burn up of 1.5 GWd/t.

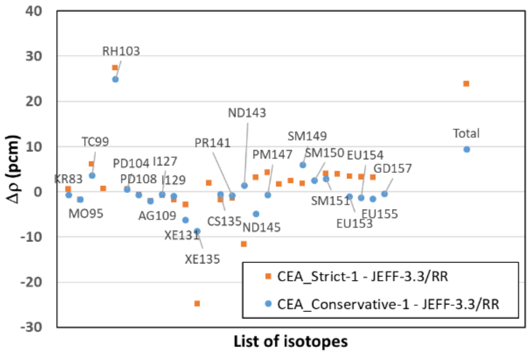

Fig. 25. (Preliminary results) Variation of reactivity for the "Strict 1" and "Conservative 1" FY evaluations in reference of the JEFF-3.3 for a burn up of 50 GWd/t.

5 Conclusion and Perspectives

The proposed analysis method of several fission observables allows us to determine a consistent evaluation of the independent and cumulative yields.
In this global approach, we conclude that the sorting of the data is an essential milestone to reach the best estimation of fission yields, limiting the bias and minimizing the uncertainties. If there is no bias in the very accurate measurements of chain yields, then the consistency of our work with the JEFF-3.3 library allows us to conclude that the strict evaluation corresponds to the best assessment of the $^{235}U(n_{th},f)$ mass yields. Nevertheless, this force us to exclude a part of the available measurements. On the other hand, to limit the potential bias of evaluation, the conservative method proposed a synthesis of the actual experimental knowledge taking into account the inconsistency of the datasets used. These two evaluations represent the upper and lower limits on the precision on these observables.

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