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Draft on experimental uncertainty estimation for PFNS-CRP document LA-UR-13-

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1 Uncertainty estimation for selected experimental PFNS data sets

1.1 Introduction

Here, we estimate uncertainties for experimental PFNS for selected data sets and isotopes which were identified as of high importance for nuclear applications [1]. In this paper, an "uncertainty" measures the dispersion of possible and often (to some extent) unknown errors pertinent to the measured values, see e.g. [2]. An "error" is the difference between the true value of an observable, here the PFNS, and the actual measured value. These errors can be of statistical and systematic nature. Corresponding statistical uncertainties quantify the possible dispersion of an uncorrelated random error for a specific data point which is independent of any other data point, while systematic uncertainties correspond to the dispersion of correlated errors which might affect part or all of one or even multiple data sets.

Here, we provide uncertainties and correlations between uncertainties of different data points in the form of covariances. A covariance matrix element $\mathrm{Cov}(x_i, x_j)$ for variables x_i and x_j is formerly defined as,

$$\mathrm{Cov}(x_i, x_j) = \langle (x_i - \langle x_i \rangle)(x_j - \langle x_j \rangle) \rangle , \tag{1}$$

where $\langle . \rangle$ signifies an expectation value. It is the second moment of the probability distribution function of the variables x_i and x_j, while the mean value $\langle x_i \rangle$ corresponds to the first moment. The diagonal of a covariance matrix,

$$\mathrm{Cov}(x_i, x_i) = \left\langle (x_i - \langle x_i \rangle)^2 \right\rangle = \mathrm{var}(x_i), \tag{2}$$

corresponds to the variance $\mathrm{var}(x_i)$ of the variable x_i. The correlation matrix element $\mathrm{Cor}(x_i, x_j)$ associated with $\mathrm{Cov}(x_i, x_j)$ is given by,

$$\mathrm{Cor}(x_i, x_j) = \frac{\mathrm{Cov}(x_i, x_j)}{\mathrm{var}(x_i)\mathrm{var}(x_j)}. \tag{3}$$

The diagonal elements of $\mathrm{Cor}(x_i, x_j)$ must always be 1, while the off-diagonal elements can assume values between -1 and 1, $-1 \leq \mathrm{Cor}(x_i, x_j) \leq 1$ for $i \neq j$. The off-diagonal elements measure the linear dependence of x_i and x_j; for independent variables, it is zero, 1 for exact positive linear dependence. However, $\mathrm{Cor}(x_i, x_j) = 0$ does not indicate that x_i and x_j are independent, because $\mathrm{Cor}(x_i, x_j)$ and $\mathrm{Cov}(x_i, x_j)$ only measure the "linear" dependence and no higher-order dependences.

In recent years, the development of new reactor types of the Generation IV triggered a demand for evaluated covariances to quantify their safety margins [3, 4]. Consequently, many modern evaluation techniques, see e.g. [5], provide evaluated covariances based on experimental and model covariances. Thus a sound estimate of experimental covariance matrices is important and an effort was made here to provide reasonable estimates for experimental covariances for selected PFNS data sets.

It should be pointed out that a covariance matrix is not a physical quantity. It is a measure of belief into the data set either assigned by the experimentalists of the respective data sets themselves or at a later point by evaluators, and are consequently subjective. In particular, systematic uncertainties are often obtained by expert judgment quantifying a possible error that cannot be completely determined, and are thus only an estimate, not a distinct bound on that possible error. In addition, to this "uncertainty on the known uncertainty", there might be unknown errors affecting a data set. If the data set is well-documented and the documentation is generally available and properly linked in a data base, such as EXFOR [6], these errors might be corrected at a later time or additional uncertainties might be added by an evaluator.

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In the present work, covariance matrices are estimated from the uncertainties tabulated in EXFOR [6] as well as from additional information found in the literature and in the EXFOR entry itself. Additional uncertainties may also be included to quantify sources of errors in the experiments that might have been not accounted for at the time of the measurement. Of course, this is only possible if detailed uncertainty information and a sufficient description of the experimental set-up are available. which is unfortunately not often the case. In some cases the experimental description is too sketchy to provide covariance matrix estimates. Hence, we stress again the long-standing demand [7, 8, 9] for detailed and easily accessible documentation of the experimental set-up as well as a thorough uncertainty analysis providing partial uncertainties. It is also helpful if this information is stored in EXFOR [6] or similar databases to use and access it more easily in the evaluation procedure. If detailed experimental and uncertainty information is not available, the evaluator might choose to reject an experimental data set for the evaluation as unknown associated uncertainties might lead to an improper weight of a specific data set and introduce a bias in the evaluated mean values as well as covariances.

1.2 Common uncertainty sources of PFNS experiments and their correlations

For the uncertainty analyses of the experimental data sets, uncertainties are partitioned into different contributions according to their respective sources, e.g., uncertainties due to the neutron detector efficiency determination, background subtraction, etc. This partition allows to estimate the correlations between uncertainties more easily as they are assigned to the partial uncertainty contributions. The correlations of some partial uncertainty contributions are well-known, for instance, statistical uncertainties have zero off-diagonal correlations by definition, while it can be difficult to estimate realistic correlations for total uncertainties including several sources. Certain uncertainty contributions can also be changed or replaced in a more transparent manner if they are considered to be unrealistic. In addition, it also simplifies the estimation of correlations between uncertainties of different experiments based on common uncertainty sources occurring in those experiments.

Below, partial uncertainty contributions typically encountered in PFNS experiments are listed based on information in [9, 10, 11, 12] including examples of underlying sources as well as a few general clues concerning the estimation of correlations.

- **Counting statistics** uncertainties with standard deviation Δs stemming from the finite number of prompt fission neutron counts are of statistical nature and their off-diagonal correlations Cor_s are zero.

- **Background** uncertainties with standard deviation Δb quantify possible ambiguities in the correction of background events. While the background events due to random coincidence, room return or scattering in air can be of random nature, the uncertainties associated to their correction might not be; for instance, in [27], the background counts were measured by placing a copper cone between the incident neutrons and the fissioning sample and assuming that the measured counts were only background events. However, the copper cone was not a perfect absorber and hence a systematic uncertainty applies to the background correction [38]. The uncertainties were estimated using MCNP simulations of the experimental set-up [38] and observables therein (e.g., copper cross sections), which are subject to correlated uncertainties that propagate to the non-zero off-diagonal correlations Cor_b associated to Δb.

- Uncertainties related to the **detector efficiency** determination with standard deviation Δd are of systematic nature and their often non-zero off-diagonal correlations Cor_d need to be estimated according to the underlying sources for Δd. Possible underlying uncertainty sources are the uncertain reference neutron production cross sections (see e.g. [31]), uncertain cross sections in a simulation program (see e.g. [29]), or extrapolations and interpolations (see e.g. [31, 27]), employed to obtain the detector efficiency curve.

- If the PFNS is measured in **ratio** to the PFNS of a **reference material**, e.g. ^{252}Cf, the final PFNS is obtained by multiplying the ratio with an accepted numerical representation of the reference PFNS. Normally, standard deviations Δr and correlations Cor_r are provided for these numerical representations, see e.g. [35, 36].

- The standard deviation Δt of the **time resolution** is often given in units of time and needs to be transformed into uncertainties relative to the PFNS. In double-time-of-flight experiments, where a fission detector provides the start signal (called "t_0") and the neutron detector the end signal, the time resolution can be partitioned into contributions with a standard deviation due to the finite channel width Δt_w and one due to an uncertainty in the determination of t_0, Δt_0. Usually Δt applies equally to the same measurement, however if several neutron or fission detectors are used in a measurement, the off-diagonal correlations Cor_t might be smaller than 1 for the different detectors.

- An uncertainty in the **time-of-flight length** with standard deviation Δl usually applies to the whole measurement. It is often given in units of length and needs to be transformed into uncertainties relative to the PFNS.

- Corrections for **multiple scattering** in the sample and in the collimator as well as for attenuation in the sample are undertaken in many PFNS experiments via computer simulations or analytical considerations. Due to simplifying assumptions in these procedures and usage of uncertain observables, uncertainties with standard deviation Δm and non-zero off-diagonal correlations Cor_m apply to the PFNS spectrum.

- The **detector response** function is not a delta-function in energy and thus the spectrum should be obtained through a de-convolution process. This is a complex process and simplifying assumptions (e.g. assuming a delta-function) lead to a bias of the spectrum at high outgoing energies with standard deviation Δc and non-zero off-diagonal correlation Cor_c.

- The geometry of some fission detectors can lead to an **angular distortion** of the spectrum. The correction might be subject to an uncertainty with standard deviation $\Delta\angle$ and correlations $\mathrm{Cor}_\angle$ that depend on the specific method used to estimate the corrections.

- Sometimes, experimental **uncertainties relative to the outgoing neutron energy** with standard deviations ΔE are provided by the experimentalists. These are for instance stemming from the uncertainty in energy calibration, time resolution or time-of-flight length and the correlations Cor_E can be estimated according to their underlying sources. The uncertainties relative to energy need to be transformed into uncertainties relative to the PFNS.

While it is of course desirable to have such detailed uncertainty information, it is often missing. In particular, correlation information is often not available and has to be estimated by expert judgment. The underlying sources of the uncertainties can provide helpful guidelines for estimating this correlation. Hence, detailed uncertainty and experimental set-up information is needed.

When it comes to correlations between uncertainties of different experiments, it is even more difficult to estimate them in a reasonable manner. We recommend to compare the uncertainty sources of two experiments as well as their underlying sources and infer their correlations. It should be pointed out that even if the same neutron detector type was used in two experiments, it might not necessarily lead to correlations of the detector efficiency uncertainties; for instance, if the detector efficiency curve was determined in one case with a computer program and in another case relative to neutron production cross sections, the underlying sources of uncertainties are different and thus the correlations between those two are zero. However, if the same neutron detectors with the same detector efficiency curve were used in two experiments, the associated uncertainties are correlated. This case occurs for instance in measurement series of several isotopes in the same experimental set-up. A typical uncertainty source leading to correlations between uncertainties of two experiments is the uncertainty related to a common reference isotope if for both measurements the same numerical representation was chosen for the reference PFNS.

1.3 Estimation of covariances for total uncertainties based on partial uncertainty information

We describe the procedure to estimate covariances of total uncertainties from partial uncertainty information. The partial uncertainties quantify the limited knowledge pertinent to certain "components" of the measurement, e.g., background reduction, detector efficiency determination, etc. Typical partial uncertainty contributions of PFNS experiments are listed in Section 1.2.

Different uncertainty contributions can apply to an experimental data set depending on how the efficiency of the outgoing neutron detector was determined. In general, three different cases are encountered [13, 14, 15]:

Case 1 The neutron detector efficiency is determined without one specific reference material, for instance relative to several neutron production cross sections or via a computer program [31, 29].

Case 2 The PFNS measurement is undertaken in ratio to the PFNS of one well-known reference material/isotope, e.g. ^{252}Cf. The isotope in question as well as its reference are measured under the same conditions; i.e., using the same experimental set-up and a reference as well as a target sample with the same geometry.

Case 3 The neutron detector is calibrated via a PFNS measurement of a reference isotope. Contrary to Case 2, the neutron detector need not be calibrated in the same set-up as the isotope in question is measured.

Case 1 If the detector efficiency is determined directly without relying on one specific reference isotope, the covariance matrix elements $\mathrm{Cov}\left(N_i, N_j\right)$ of the total uncertainties associated with the PFNS N_i and N_j are obtained by the sum of all partial uncertainty contributions:

$$\begin{aligned}\mathrm{Cov}\left(N_i, N_j\right) = {} & \Delta s_i \Delta s_j \delta_{ij} + \Delta b_i \Delta b_j \mathrm{Cor}_b\left(N_i, N_j\right) + \Delta d_i \Delta d_j \mathrm{Cor}_d\left(N_i, N_j\right) + \mathrm{Cov}_t\left(N_i, N_j\right) + \mathrm{Cov}_l\left(N_i, N_j\right) \\ & + \Delta m_i \Delta m_j \mathrm{Cor}_m\left(N_i, N_j\right) + \Delta c_i \Delta c_j \mathrm{Cor}_c\left(N_i, N_j\right) + \Delta\angle_i \Delta\angle_j \mathrm{Cor}_\angle\left(N_i, N_j\right) + \mathrm{Cov}_E\left(N_i, N_j\right). \end{aligned} \quad (4)$$

with the Kronecker delta $\delta_{ij} = 1$ for $i = j$ and $\delta_{ij} = 0$ for $i \neq j$. The indexes i and j of a variable x indicate that x is given for the outgoing neutron energies E_i and E_j. It is implicitly assumed that the correlations between different uncertainty contributions are zero. It is straight forward to add other uncertainty contributions not explicitly named here. No PFNS of a reference isotope is employed to obtain the PFNS in Case 1, hence no associated uncertainty Δr needs to be considered.

The outgoing neutron energy covariances $\mathrm{Cov}_E\left(N_i, N_j\right)$ are already transformed into covariances for the PFNS N_i and N_j. To obtain those from ΔE_i and Cor_E in outgoing neutron energy space, we use linear error propagation,

$$\mathrm{Cov}_E\left(N_i, N_j\right) = \frac{\partial N}{\partial E}\,|_{E_i}\; \Delta E_i \mathrm{Cor}_E(E_i, E_j) \Delta E_j \frac{\partial N}{\partial E}\,|_{E_j}\;. \quad (5)$$

We use a Maxwellian distribution with a normalization constant C and temperature T,

$$N_i = C\sqrt{E_i}\exp\left(-E_i/T\right), \tag{6}$$

to estimate the partial derivative $\partial N/\partial E\mid_{E_i}$ with

$$\frac{\partial N}{\partial E}\mid_{E_i} = \frac{N_i}{E_i}\left(\frac{1}{2}-\frac{E_i}{T}\right). \tag{7}$$

For N_i the actual experimental data can be used. Of course, this approach suffers from (a) the shortcomings of a Maxwellian to describe actual experimental data and (b) neglecting higher-order terms of uncertainties.

The covariances $\mathrm{Cov}_t(N_i, N_j)$ in terms of the PFNS associated with the finite time resolution Δt are obtained in a two-step process. First, time resolution uncertainties Δt given relative to the time-of-flight are transformed into uncertainties relative to the outgoing neutron energy by using linear error propagation and a non-relativistic approximation of the energy-time relationship,

$$\mathrm{Cov}_t(E_i, E_j) = 4\frac{\Delta t^2}{t_i t_j}E_i E_j. \tag{8}$$

We implicitly assume that the time resolution uncertainty Δt is constant for all times-of-flight t_i and is thus a fully correlated uncertainty component. In a second step, $\mathrm{Cov}_t(E_i, E_j)$ are transformed into covariances associated to the PFNS using again linear error propagation (see Eq. (5)):

$$\mathrm{Cov}_t\left(N_i, N_j\right) = 4\frac{\sqrt{E_i E_j}}{ml^2}N_i N_j\left(\frac{1}{2}-\frac{E_i}{T}\right)\left(\frac{1}{2}-\frac{E_j}{T}\right)(\Delta t)^2. \tag{9}$$

Analogously, a time-of-flight length uncertainty Δl is transformed into covariances associated to the outgoing neutron energy by:

$$\mathrm{Cov}_l(E_i, E_j) = 4\frac{(\Delta l)^2}{l^2}E_i E_j, \tag{10}$$

and then in covariances relative to the PFNS by:

$$\mathrm{Cov}_l\left(N_i, N_j\right) = \frac{4}{l^2}N_i N_j\left(\frac{1}{2}-\frac{E_i}{T}\right)\left(\frac{1}{2}-\frac{E_j}{T}\right)(\Delta l)^2. \tag{11}$$

Of course, Eq. (4) can be generalized to estimate covariances between two different experiments that need not be for the same isotope. Statistical uncertainties will not appear in covariances between two experiments k and h, hence the covariances between experiments read:

$$\begin{aligned}\mathrm{Cov}\left(N_i^k, N_j^h\right) &= \Delta b_i^k \Delta b_j^h \mathrm{Cor}_b\left(N_i^k, N_j^h\right) + \Delta d_i^k \Delta d_j^h \mathrm{Cor}_d\left(N_i^k, N_j^h\right) + \mathrm{Cov}_t\left(N_i^k, N_j^h\right) + \mathrm{Cov}_l\left(N_i^k, N_j^h\right) \\ &+ \Delta m_i^k \Delta m_j^h \mathrm{Cor}_m\left(N_i^k, N_j^h\right) + \Delta c_i^k \Delta c_j^h \mathrm{Cor}_c\left(N_i^k, N_j^h\right) + \Delta\angle_i^k \Delta\angle_j^h \mathrm{Cor}_\angle\left(N_i^k, N_j^h\right) + \mathrm{Cov}_E\left(N_i^k, N_j^h\right).\end{aligned} \tag{12}$$

While it is straight forward to combine the partial uncertainty contributions to give total uncertainties and associated covariances, it is often difficult to assess correlations, especially between different experiments.

Case 2 In a PFNS measurement of an isotope A in ratio to a reference PFNS B,

$$R_i = N_i^A/N_i^B, \tag{13}$$

using the same neutron detectors, the neutron detector efficiency drops out and thus the associated uncertainties Δd. In addition, some part of the background counts cancel each other, for instance the random coincidences, part of the room-return background, etc. However, not all of the background contributions drop out and thus a non-zero background uncertainty $\Delta b'$ might apply; for instance, if ^{235}U is measured relative to ^{252}Cf, often ^{252}Cf is measured with incident neutron beam off while it is on for ^{235}U. Hence, a background due to neutrons from the incident neutron beam applies to the ^{235}U counts but not to the ^{252}Cf counts. If the two samples as well as the fission chamber have the same geometry, the uncertainty due to an angular distortion $\Delta\angle$ should cancel out to a large extent. However, statistical uncertainties Δs, multiple scattering and deconvolution uncertainties, Δm and Δc respectively, apply to the ratio R_i. Outgoing neutron energy uncertainties ΔE, uncertainties due to the finite time resolution Δt and time-of-flight length Δl uncertainties reduce due to the ratio measurement. This reduction can be observed, when representing the PFNS N_A and N_B by Maxwellian distributions for the ratio R,

$$R_i = \frac{C_A}{C_B}\exp\left\{E_i\left(\frac{1}{T_B}-\frac{1}{T_A}\right)\right\} = C_R\exp\left\{E_i\left(\frac{1}{T_B}-\frac{1}{T_A}\right)\right\}, \tag{14}$$

with Maxwellian temperatures T_A and T_B. The first partial derivative of R respective to E thus reads:

$$\frac{\partial R}{\partial E}\ |_{E_i}= C_R \exp\left\{E_i\left(\frac{1}{T_B}-\frac{1}{T_A}\right)\right\}\left(\frac{1}{T_B}-\frac{1}{T_A}\right) = R_i\left(\frac{1}{T_B}-\frac{1}{T_A}\right), \tag{15}$$

where one can use again the actually measured value R_i. Consequently, $\mathrm{Cov}_E(R_i,R_j)$, $\mathrm{Cov}_t(R_i,R_j)$ and $\mathrm{Cov}_l(R_i,R_j)$ read:

$$\mathrm{Cov}_E(R_i,R_j) = R_iR_j\left(\frac{1}{T_B}-\frac{1}{T_A}\right)^2 \Delta E_i\Delta E_j \mathrm{Cor}_E(E_i,E_j), \tag{16}$$

$$\mathrm{Cov}_t(R_i,R_j) = 4R_iR_j\left(\frac{1}{T_B}-\frac{1}{T_A}\right)^2 \frac{(\Delta t)^2}{ml^2}(E_iE_j)^{3/2}, \tag{17}$$

$$\mathrm{Cov}_l(R_i,R_j) = 4R_iR_j\left(\frac{1}{T_B}-\frac{1}{T_A}\right)^2 E_iE_j\frac{(\Delta l)^2}{l^2}. \tag{18}$$

The covariance matrix elements for the total uncertainties of the ratio data are thus given by

$$\begin{aligned}\mathrm{Cov}\left(R_i,R_j\right) = {} & \Delta s_i\Delta s_j\delta_{ij} + \Delta b'_i\Delta b'_j\mathrm{Cor}_{b'}\left(R_i,R_j\right) + \mathrm{Cov}_{t'}\left(R_i,R_j\right) + \mathrm{Cov}_{l'}\left(R_i,R_j\right) \\ & + \Delta m_i\Delta m_j\mathrm{Cor}_m\left(N_i,N_j\right) + \Delta c_i\Delta c_j\mathrm{Cor}_c\left(N_i,N_j\right) + \mathrm{Cov}_{E'}\left(N_i,N_j\right),\end{aligned}$$

where the prime, "$'$", indicates that the uncertainties partly cancel out in the ratio measurement compared to Case 1.

The experimental PFNS are obtained by multiplication of the measured ratio data R_i with a numerical representation $N^{(B,n)}$ of the reference isotope B,

$$N_i^A = R_iN_i^{(B,n)}. \tag{19}$$

The covariances $\mathrm{Cov}\left(N_i^A,N_j^A\right)$ associated to the values N_i^A are derived in linear error propagation,

$$\mathrm{Cov}\left(N_i^A,N_j^A\right) = N_i^{(B,n)}N_j^{(B,n)}\mathrm{Cov}\left(R_i,R_j\right) + R_iR_j\Delta r_i\Delta r_j\mathrm{Cor}_r(N_i^{(B,n)},N_j^{(B,n)}), \tag{20}$$

using the standard deviations Δr and correlation matrix Cor_r associated with the numerical representation $N^{(B,n)}$ of the reference PFNS.

Case 3 Here, we treat the case in which the detector efficiency ε is calibrated by the PFNS measurement with values C^B of a reference isotope B and using a known numerical representation $N^{(B,n)}$ of the reference PFNS, i.e.

$$\varepsilon_i = \frac{C_i^B}{N_i^{(B,n)}}. \tag{21}$$

The experimental PFNS of the isotope in question A can then be derived by

$$N_i^A = \frac{C_i^A}{C_i^B}N_i^{(B,n)}. \tag{22}$$

The covariances of the total uncertainties,

$$\mathrm{Cov}(N_i,N_j) = N_i^AN_j^A\left[\frac{\mathrm{Cov}\left(C_i^A,C_j^A\right)}{C_i^AC_j^A} + \frac{\mathrm{Cov}\left(C_i^B,C_j^B\right)}{C_i^BC_j^B} + \frac{\Delta r_i\Delta r_j\mathrm{Cor}_r(N_i^{(B,n)},N_j^{(B,n)})}{N_i^{(B,n)},N_j^{(B,n)}}\right], \tag{23}$$

comprise covariances Cov_A, Cov_B and $\Delta r_i\Delta r_j\mathrm{Cor}_r(N_i^{(B,n)},N_j^{(B,n)})$ of the actual measured values C_i^A, C_i^B and the numerical representation $N_i^{(B,n)}$ of the reference PFNS.

In Cov_A as well as Cov_B, statistical uncertainties Δs, background uncertainties Δb, the finite time resolution Δt, the uncertainty in the time-of-flight length Δl, multiple scattering Δm, angular distortion correction uncertainties $\Delta\angle$ and energy-dependent ones ΔE enter for each measurement separately, while detector efficiency and deconvolution uncertainties drop out. This entails a detailed uncertainty analysis of measurements A and B. Another way to represent this is to consider the uncertainties Cov_B and $\Delta r_i\Delta r_j\mathrm{Cor}_r(N_i^{(B,n)},N_j^{(B,n)})$ in the covariances associated with the detector efficiency determination $\Delta d_i\Delta d_j\mathrm{Cor}(N_i^A,N_j^A)$ and to use Eq. (4) instead. This is formerly correct, however it might be less transparent than a detailed analysis of measurement B.

1.4 Uncertainty estimation for selected ^{239}Pu experimental PFNS

In this section, we illustrate the concepts and procedures discussed above in the case of ^{239}Pu measurements. No experimental covariance matrices were estimated for the data sets of [16, 19, 21, 17] as insufficient uncertainty and/ or experimental information was available for these data sets to provide reasonable estimates. The data set reported in [20] was not considered since the measured spectrum comprises prompt and delayed neutron counts.

Experimental covariances were estimated [22] for the data sets of [23, 24, 25, 26, 27, 29, 31] based on information provided in EXFOR [6], the respective literature as well as by preliminary MCNP studies [37, 38] indicating missing corrections to data of [27, 29, 31]. The data sets of [23, 29, 31] correspond to a measurement of the type of Case 1 of Section 1.3, i.e., the detector efficiency was computed or measured relative to several neutron production cross sections. The measurements of [24, 25, 26] were undertaken in ratio to ^{252}Cf(sf) PFNS (Case 2 of Section 1.3), while the neutron detector efficiency of the measurement of [27] was calibrated with the ^{252}Cf(sf) PFNS (Case 3 of Section 1.3). Table 1 indicates where the partial uncertainty sources for [23, 24, 25, 26, 27, 29, 32] were extracted from.

Table 1: The uncertainty sources (unc. source) explicitly provided in the EXFOR [6] entry, described in the literature or added by the authors are listed here for experimental data sets where an experimental covariance matrix was estimated as part of this collaboration. Further literature apart from the main reference is also provided

Experiment	Literature used	Unc. source in EXFOR	Unc. source in literature	Corrected unc. source
Nefedov, 1983	[23, 26]	Δs, Δb, Δd	Δt, Δl	Δc
Boytsov, 1983	[24, 26]	Δs, Δb, Δt all from [26]	Δs, Δt, Δb, Δr	Δb, Δr, Δl
Starostov, 1983	[25, 26]	Δs, Δb	Δl, Δt both of of [26]	Δr
Starostov, 1985	[26]	Δs, Δb, Δd	Δs, Δb, Δt, Δl	Δb, Δr, Δl, Δc
Lajtai, 1985	[27, 28]	Δs, Δb, Δd	Δd, Δc, $\Delta\angle$, Δt	Δb, Δr
Staples, 1995	[29, 30]	Δs, Δb, Δd, ΔE	Δs, Δb, Δd, Δm, ΔE	Δd, Δm, Δc
Knitter, 1975	[31, 32, 33, 34]	Δs, Δb, ΔE	Δs, Δb, Δd, Δt	Δd, Δm, Δc

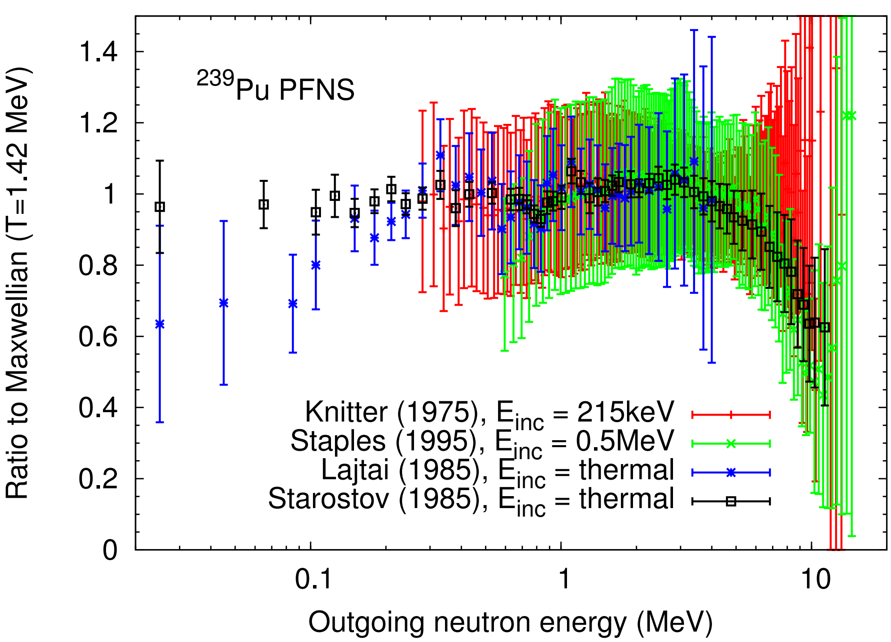

Figure 1: The experimental ^{239}Pu PFNS of [31, 29, 25, 26, 23, 24, 27] are shown (Preliminary).

Most of the data points of [23, 24, 25] were incorporated in [26] and form a major part of it. In the uncertainty analysis, we used the original data sets [23, 24, 25] an the new part of [26] as more uncertainty information was available for [23, 25] compared to [26]. As [24, 25] and part of [26] were measured in ratio to the ^{252}Cf(sf) PFNS, all data were rebinned to the lattice of the ^{252}Cf(sf) PFNS standard [35, 36]. The ratio data and associated covariances were transformed into PFNS space according to Eqs. (19) and (20) employing reference uncertainties with standard deviation Δr and correlations Cor_r provided by [35, 36]. In Figs. 1 and 2, the combined and rebinned data sets as well as their relative uncertainties are shown and are called for simplicity 'Starostov, 1985'. For [24] and part of [26], no time-of-flight length uncertainties Δl and background uncertainties Δb were provided; they were added in the present uncertainty analysis based on the corresponding information for the data sets of [23, 25]. Not all uncertainty

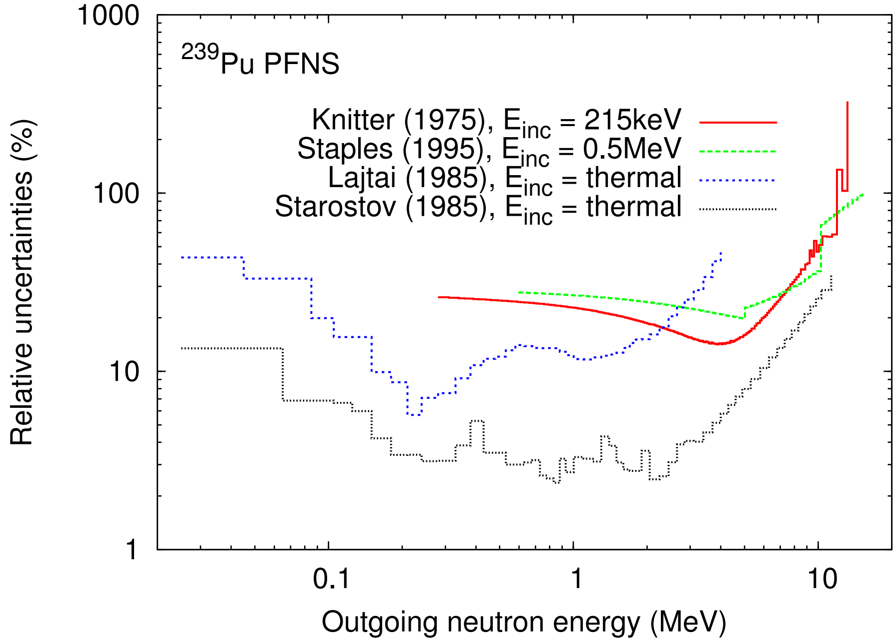

Figure 2: The total relative uncertainties estimated for ^{239}Pu PFNS of [31, 29, 25, 26, 23, 24, 27] are shown (Preliminary).

sources could be verified for this combined data set due to missing experimental information; especially, background and multiple scattering corrections cannot be tested, although their contribution should be small in this specific experimental set-up.

The data set of [27] was measured using the ^{252}Cf(sf) PFNS for the neutron detector calibration. We chose the data set of [35, 36] for the numerical representation of the ^{252}Cf(sf) PFNS and considered Δr and Cor_r provided by [35, 36] in the experimental covariance matrix for [27]. In addition, a background correction uncertainty Δb was added as preliminary MCNP studies [38] showed that they were over-corrected in [27, 28].

For the data of [29], MCNP calculations [37] uncovered that the multiple scattering and attenuation in the sample were much larger (10-15%) than the 1% of the PFNS estimated in [29]. Hence, an additional multiple scattering and attenuation uncertainty Δm was added to the experimental covariance matrix. Also, the detector efficiency uncertainties were increased at higher outgoing neutron energies to account for the uncertain cross sections used in the computations. A one-to-one correspondence was assumed between time-of-flight and outgoing neutron energy in [29] which can lead to distortions of the PFNS [37] at higher outgoing neutron energies. Hence, and uncertainty Δc was added to account for this possible bias.

MCNP studies for multiple scattering in the sample and the collimator [37] were undertaken as well for the data set of [31]. They showed that their contribution to the low outgoing neutron energy part to the spectrum are substantial contrary to the "small effect" mentioned in [31, 32]. Due to missing experimental set-up information, this effect cannot be corrected and an uncertainty Δm was assigned. No deconvolution correction is mentioned in the literature for this data set and hence an uncertainty Δc was considered in the experimental covariances.

The data set including data of [23, 24, 25, 26] has the smallest uncertainties in the present analysis, see Fig. 2. Concerns were raised if the uncertainties of [23, 24, 25, 26] might be under-estimated as we cannot test the validity of their multiple scattering and background corrections. Given the experimental set-up and our present understanding of the measurement, those contributions should be small however.

Ongoing experiments [11] should be able to probe the low outgoing neutron energy range (<0.5 MeV) to a sufficient accuracy to identify possible problems with the data of [23, 24, 25, 26]. This new experiment is especially important, as the PFNS mean values of [24] and [27] are discrepant up to 200 keV, see Fig. 1 ; however their one-sigma error bars overlap and more accurate measurements are needed to test this region. At outgoing neutron energies above 8 MeV, Fig. 1, discrepancies between the mean values of [29, 23, 24, 25, 26] and [31] can also be observed. However, the large one-sigma error bars have a substantial overlap as well.

The uncertainties of [27] and [23, 24, 25, 26] are correlated as in both their covariances the uncertainties associated with the ^{252}Cf(sf) PFNS of [35, 36] were considered. A non-zero correlation between the multiple scattering and deconvolution uncertainty contributions of [31, 29] arises as those were estimated based on similar or the same simulations. [Covariance figures will be supplied later].

Table 2: Experimental PFNS for different isotopes. Their incident neutron energy E_{inc}, outgoing neutron energy E_{out}, first author, uncertainty quantification quality as well as main reference are given.

Isotope	E_{inc} (MeV)	Source/EXFOR#	First Author	Year	E_{out} (MeV)	UQ quality	Main Reference
^{239}Pu	0.01 - 0.058	20997.001	Abramson	1977	0.55-14.253	Insufficient	[16]
^{239}Pu	0.04	20575.001	Conde	1965	0.3 - 7.5	Insufficient	[19]
^{239}Pu	thermal	20616.001	Werle	1972	0.104 - 9.495	ok, but thermal and delayed spectrum	[20]
^{239}Pu	thermal	41502.001	Batenkov	2004	0.12 - 19	Insufficient	[21]
^{239}Pu	thermal	40137.001	Belov	1969	0.3 - 7	Insufficient	[17, 18]
^{239}Pu	thermal	40871.001	Nefedov	1983	0.139 - 7.15	Detailed	[23]
^{239}Pu	thermal	40873.001	Boytsov	1983	0.021 - 4.5	Scarce	[24]
^{239}Pu	thermal	40872.001	Starostov	1983	3.007 - 11.2	Detailed	[25]
^{239}Pu	thermal	40930.001	Starostov	1985	0.020658 - 11.287	Limited	[26]
^{239}Pu	thermal	30704.001	Lajtai	1985	0.03 - 3.855	Detailed for ^{252}Cf(sf)	[27]
^{239}Pu	0.5 1.5 2.5 3.5	13982.001	Staples	1995	0.596 - 15.952 1.696 - 15.192 2.808 - 14.485 4.088 - 13.828	Ok but incomplete	[29]
^{239}Pu	0.215	20576.001	Knitter	1975	0.28 - 13.87	Ok but incomplete	[31]
^{239}Pu	1-8	14290.003	Noda	2010	1.5-9.5	Ok but incomplete (n detector)	[39]
^{239}Pu	1.5	private communication	Lestone	2013	1.5-10.5	Ongoing	[40, 41]
^{239}Pu	1-200	private communication	Chatillon	2013	0.2-8 [verification missing]	Ongoing	in preparation

2 Table summarizing experimental PFNS data sets

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