DE LA RECHERCHE À L'INDUSTRIE

CyRUS : A code dedicated to the calculation and the analysis of the uncertainties of the decay heat

WONDER 2012 | Jean-Christophe BENOIT

SEPTEMBER 24 – 28, 2012

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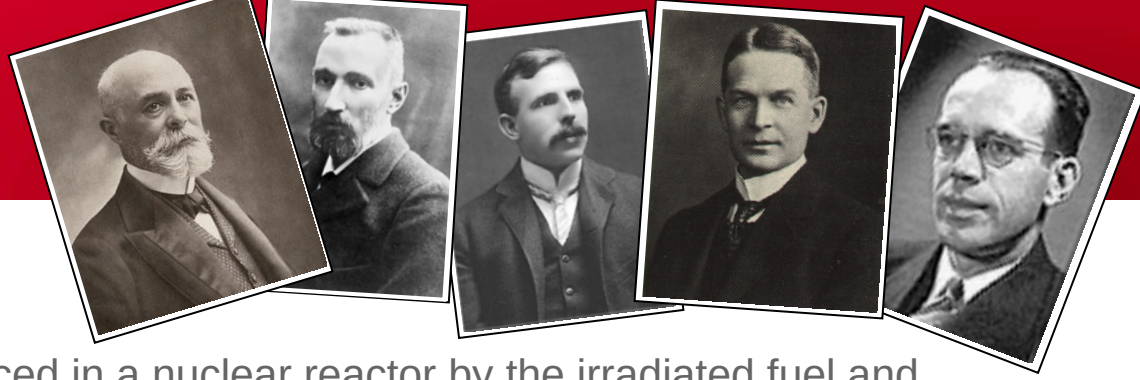
INTRODUCTION

INTRODUCTION

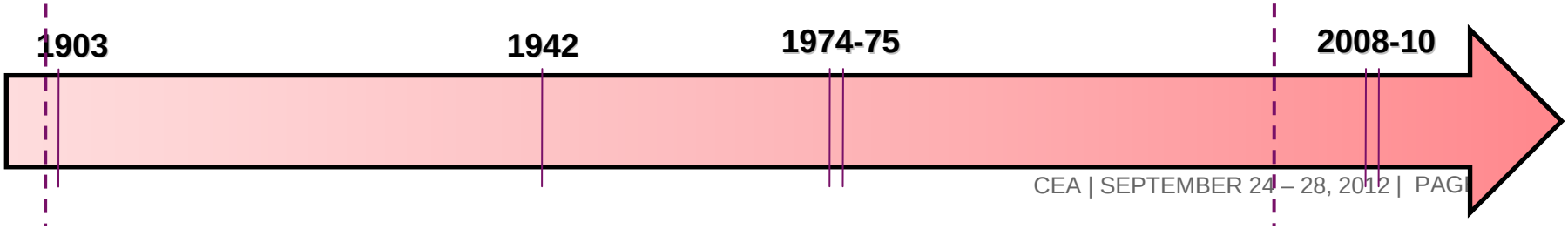


Definition

- Decay Heat (DH) : Heat produced in a nuclear reactor by the irradiated fuel and structures when the reactor is stopped. It is linked to the α, β, γ radioactivity.

An issue for a long time

- 1900 : First discovered by P. CURIE, A. LABORDE (1903) in radium salts during the early years of radioactivity, Theoretical explanation by E. RUTHERFORD and F. SODDY (1904),
- 1940 : Characterization (Plutonium Project) in order to safely build a reactor to produce plutonium (BORST, BRADY, DAY & CANNON),
- 1974 : ANS Standard on decay heat,
- 1975 : First codes in order to propagate the uncertainties of nuclear data on the decay heat (SCHENTER, SCHIMTTROTH, SPINRAD...)
- 2008 : MERCI experiment (UOx pin PWR)
- 2010 : PUIREX during PHENIX Final Tests (whole core of the 350 MWth SFR)

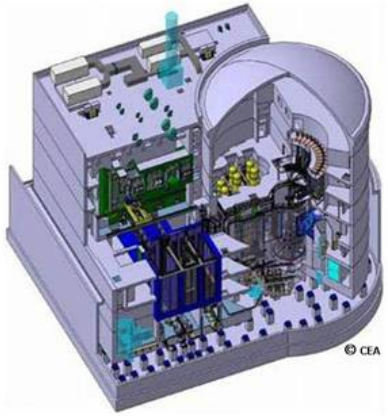


INTRODUCTION

Reasons for a more precise calculation

- More Safety and more Savings

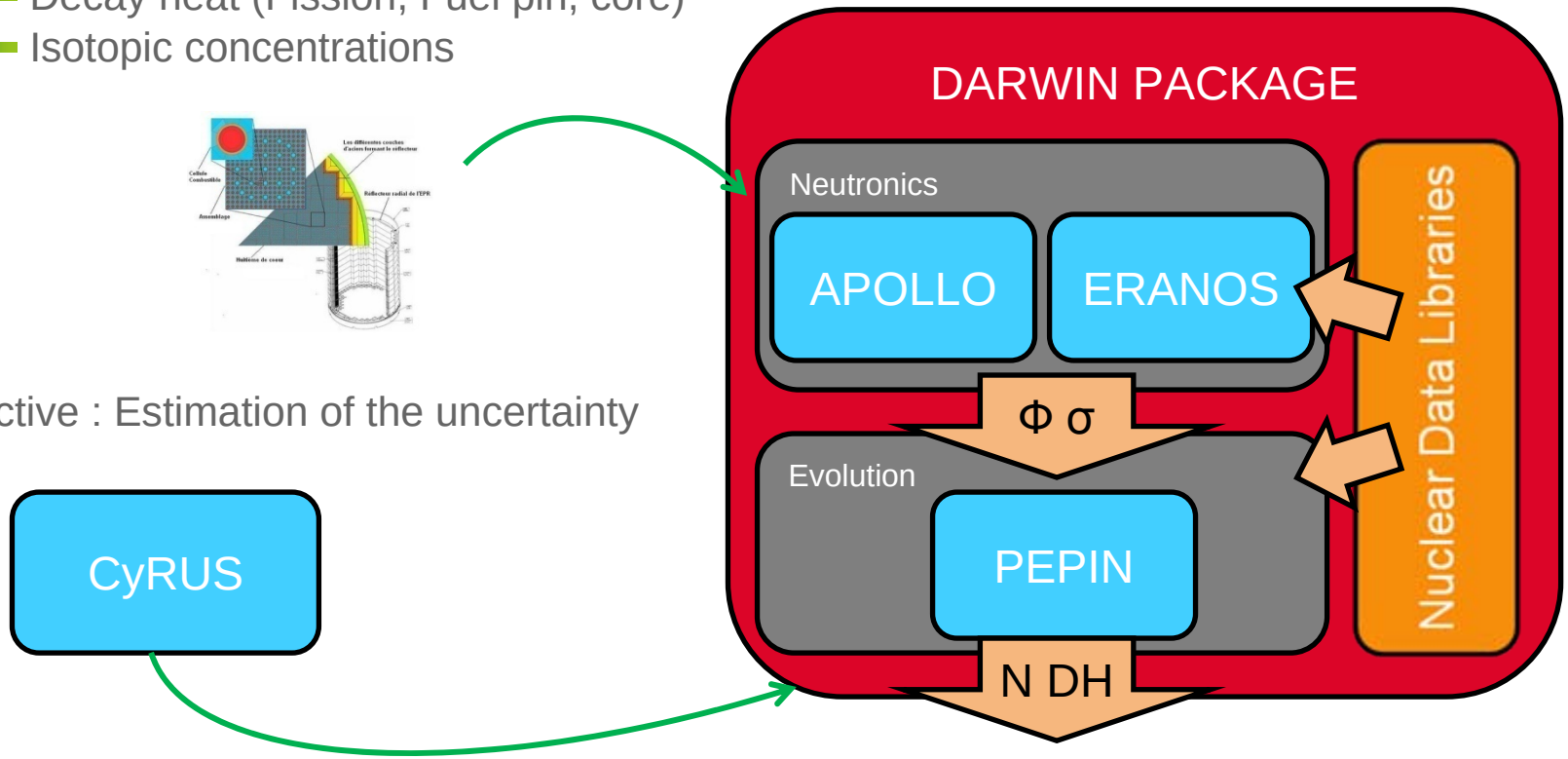
Nuclear stage impacted	Time of cooling
Safety Systems of cooling	0.1 second to 8 days
Unloading of sub-assemblies from the core	5 to 25 days
Road transport	1 to 10 years
Reprocessing, Vitrification, Storage	4 to 3000 years
Storage	50 to 300 000 years and more



Develop predictive and validated codes

- Validation : Comparison Calculation / Measurements
  - Decay heat (Fission, Fuel pin, core)
  - Isotopic concentrations

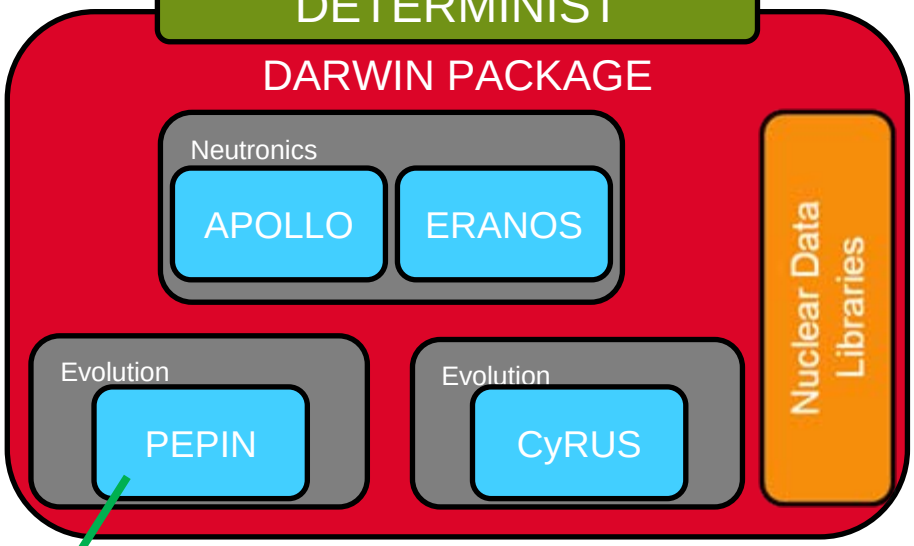
- Predictive : Estimation of the uncertainty



METHODOLOGY

METHODOLOGY

DETERMINIST



$p^0 + \delta p$

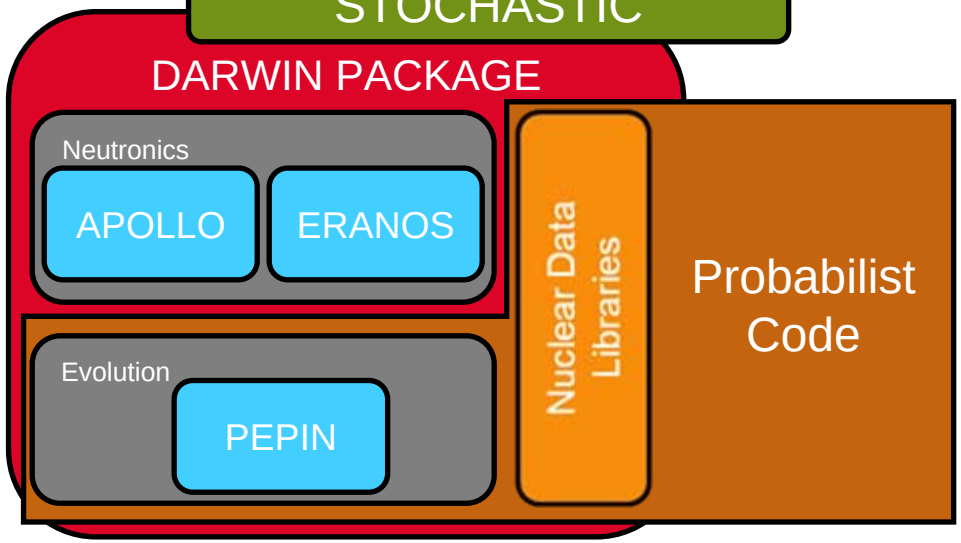
n parameters
n+1 calculations

$$S(DH/p_i) = \frac{p_i^0}{DH|_{p^0}} \frac{\partial DH}{\partial p_i}\bigg|_{p^0}$$

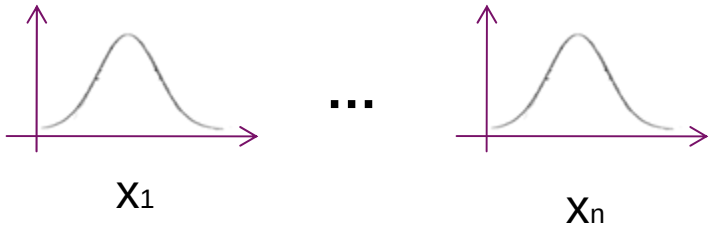
$$= \frac{p_i^0}{DH|_{p^0}} \frac{DH(p_i^0 + \delta p_i) - DH(p_i^0)}{\delta p_i}$$

$$\mathrm{var}(DH) = {}^tS\, VAR(p)\, S$$

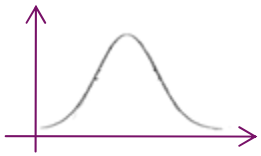
STOCHASTIC



- m samples of the n parameters



- m evolution calculations



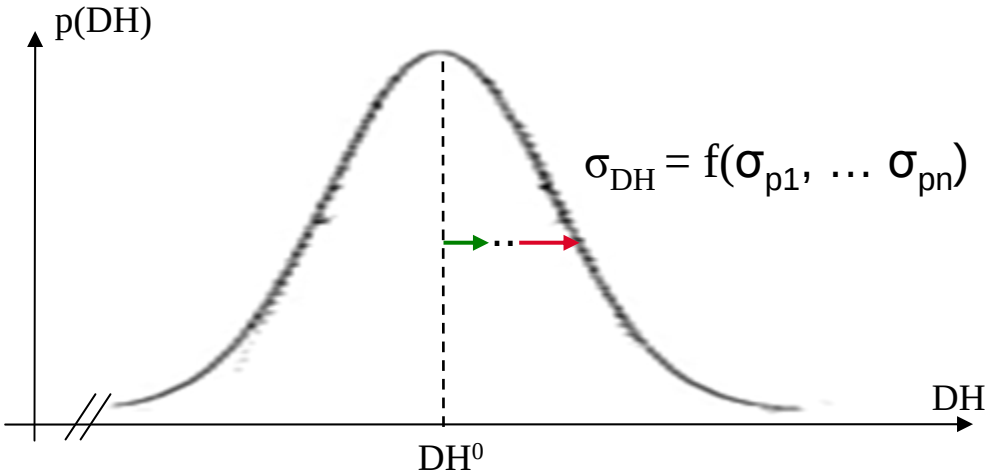
Detailed Determinist Propagation Method

- 1st order error propagation formula

$$\mathrm{var}(DH) = \underbrace{\begin{pmatrix} S_{DH/p_1} & \cdots & S_{DH/p_n} \end{pmatrix}}_{\text{Calculation}} \underbrace{\begin{pmatrix} \mathrm{var}(p_1) & & \\ & \ddots & \\ \mathrm{cov}(p_1, p_n) & & \mathrm{var}(p_n) \end{pmatrix}}_{\text{Libraries}} \begin{pmatrix} S_{DH/p_1} \\ \vdots \\ S_{DH/p_n} \end{pmatrix}$$

?

- Two assumptions :
  - 1st order formula
  - DH is normally distributed



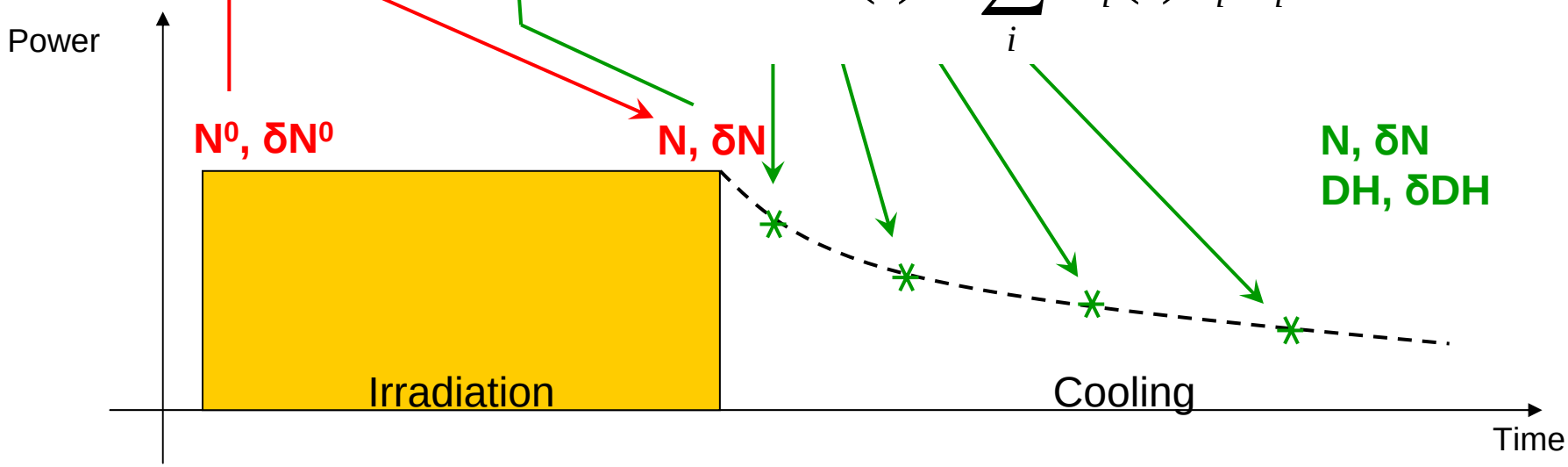
Validated during my PhD

METHODOLOGY

$$\frac{\partial N_i(t)}{\partial t} = N_f \sigma_f \phi Y_{fi} + \sum_{j=1}^{i-1} \left(\lambda_{ij} + \sigma_{ij}\phi\right) N_j(t) - \left(\lambda_i + \sigma_i \phi\right) N_i(t)$$

$$\frac{\partial N_i(t)}{\partial t} = \sum_{j=1}^{i-1} \lambda_{ij}\, N_j(t) - \lambda_i\, N_i(t)$$

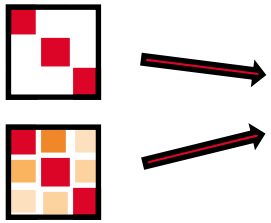
$$DH(t) = \sum_i N_i(t) \lambda_i \overline{E_i}$$



METHODOLOGY

Many results

- The uncertainty of the decay heat, δDH
- The contribution of any nuclide to the uncertainty of the decay heat + The reason of this contribution (sensibility or variance), δNi, S(DH/Ni) → δDH
- The contribution of any parameter to the uncertainty of the nuclei + The reason of this contribution (sensibility or variance), δpj, S(Ni/pj) → δNi
- The number of nuclei to which a parameter contribute significantly to the uncertainty δpj → δN1, δN2, δN3
- The possibility to modify the covariance matrix of the parameters and to see the change on the uncertainty of the decay heat quickly (in less than 1 minute).

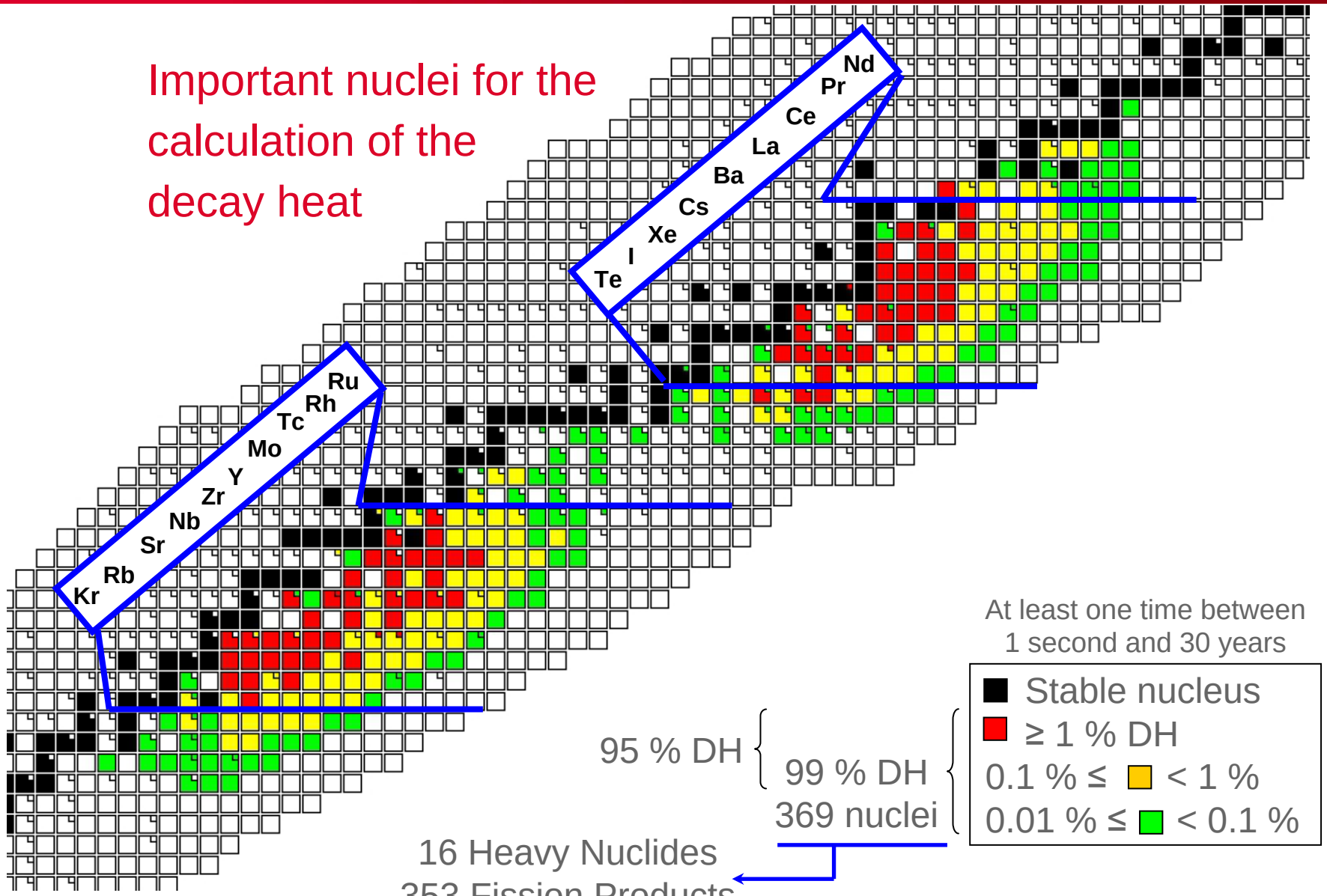


δDH

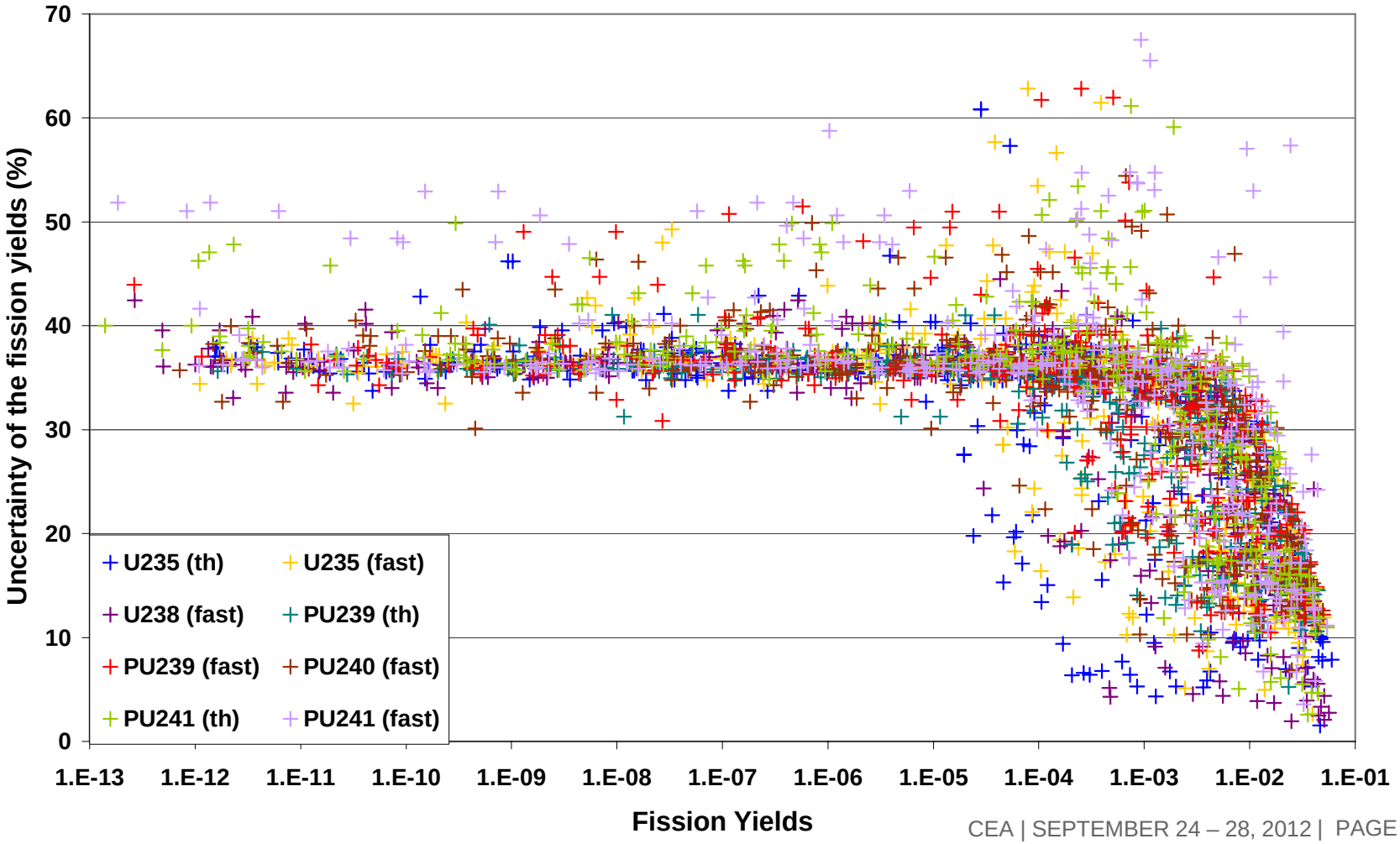
NUCLEAR DATA (JEFF3.1.1)

NUCLEAR DATA

Important nuclei for the calculation of the decay heat

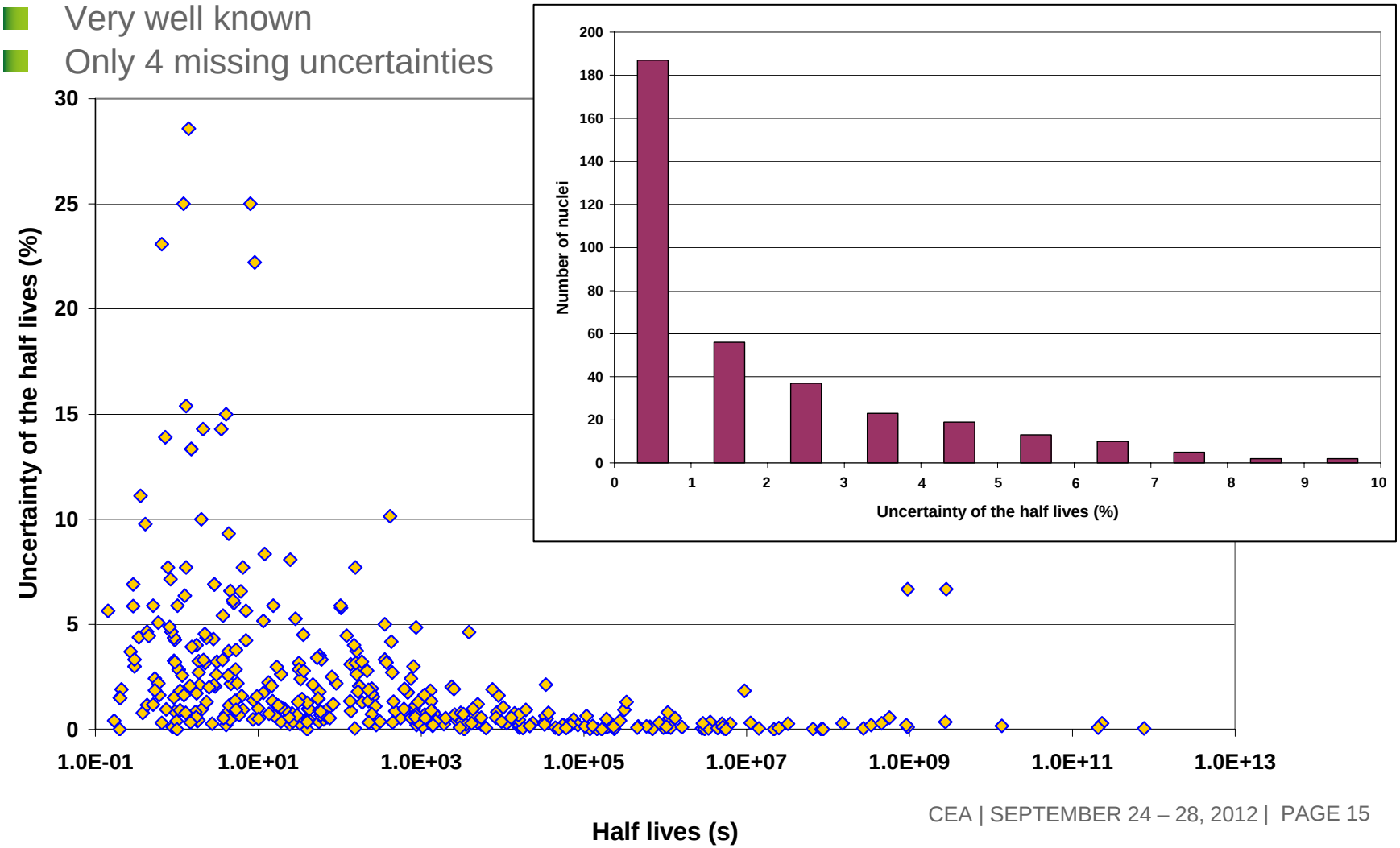


Independent Fission Yields (JEFF3.1.1, 353 FP)



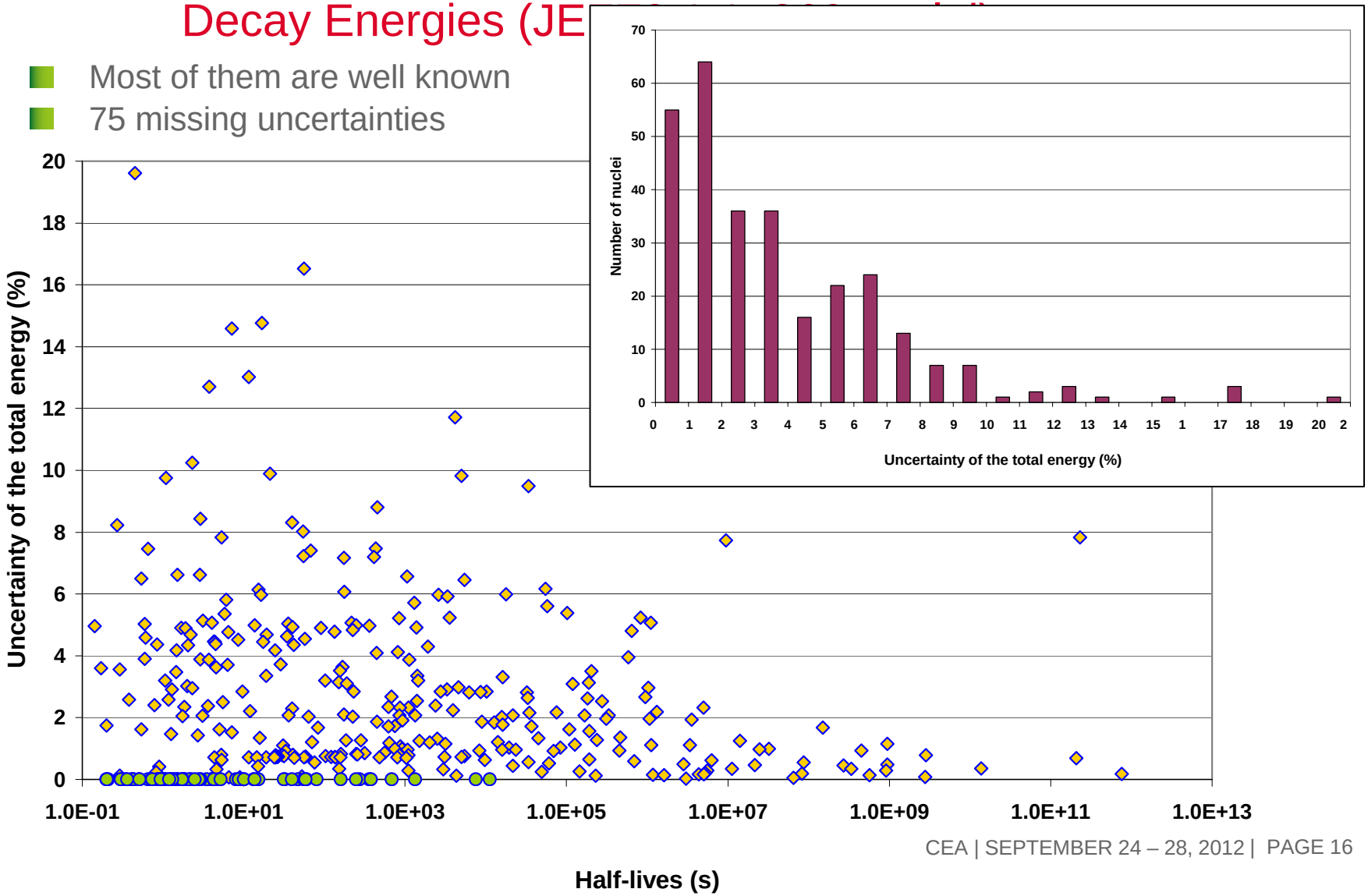
Half lives (JEFF3.1.1, 369 nuclei)

- Very well known
- Only 4 missing uncertainties



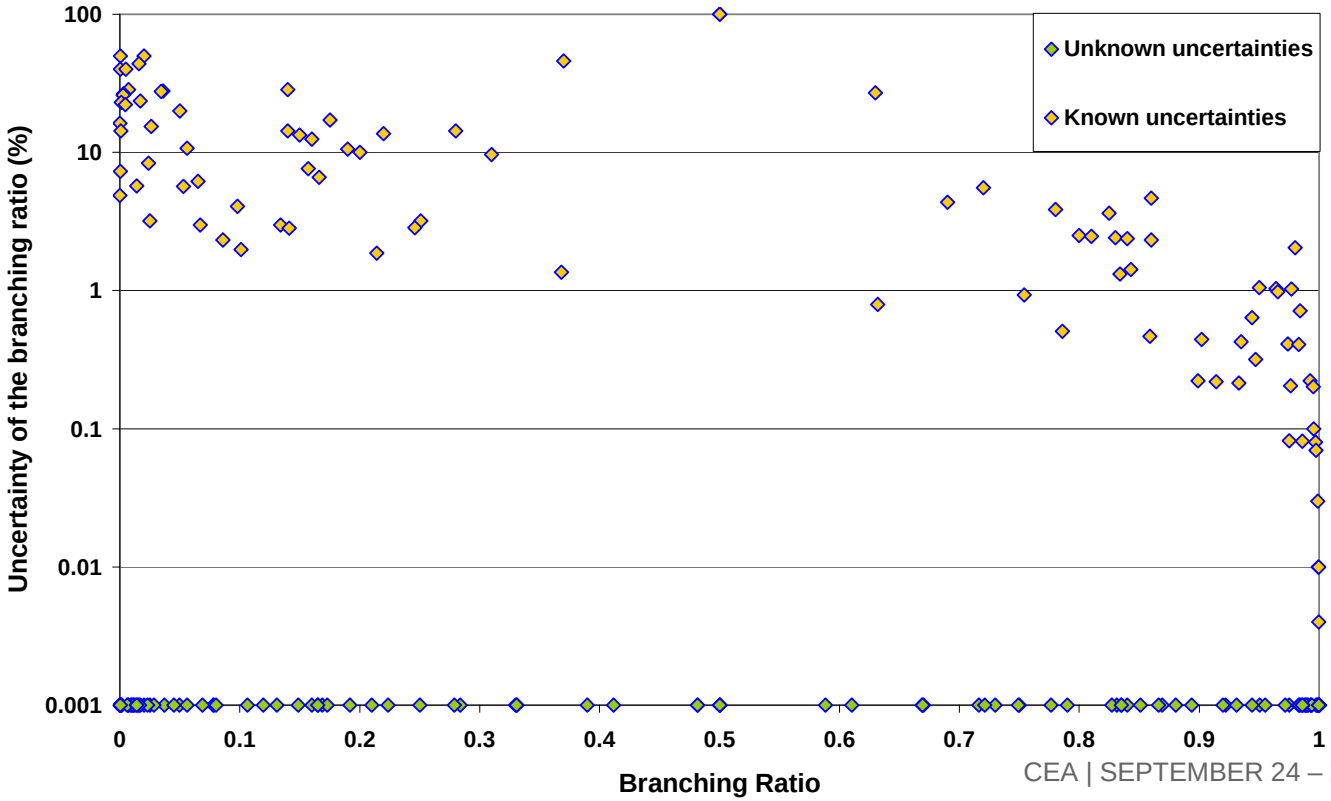
Decay Energies (JE

- Most of them are well known
- 75 missing uncertainties



Branching Ratios (JEFF3.1.1)

- Lots of data are missing (94 known uncertainties and 128 missing)
- Low impact on the uncertainty of decay heat
  - Low values of branching ratios ↔ High uncertainties
  - High values of branching ratios ↔ low uncertainties



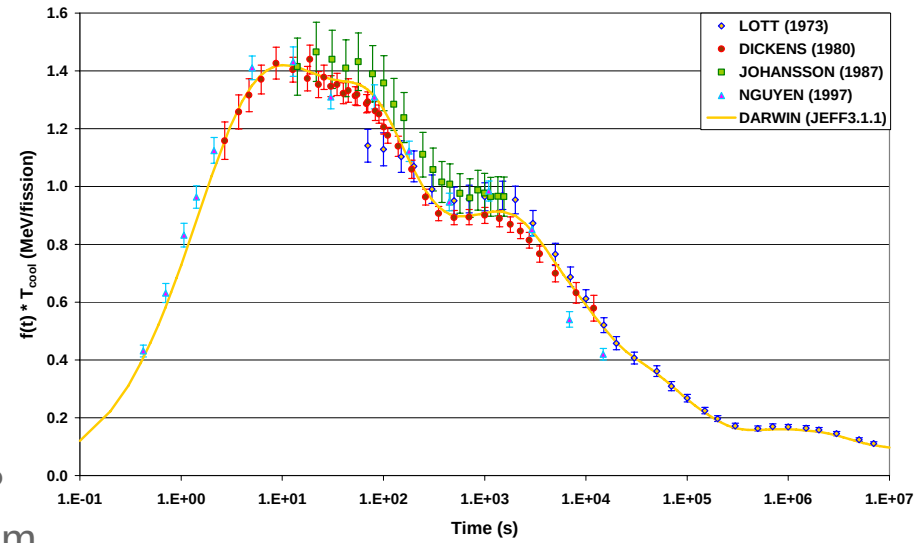
RESULTS

Burst fission curve of ^{235}U (th)

- Definition : Heat produced by the fission of one nucleus of a fissionable nuclide.

- What is it used for ?
  - Validation of nuclear data libraries (no impact of neutronics),
  - Fast calculations of decay heat with fits of several exponentials (ANS Standard),
  - Past : More precise than summation calculations because of missing nuclear data,

- Why ^{235}U (th) :
  - Widely studied in order to perform an ANS Standard for decay heat



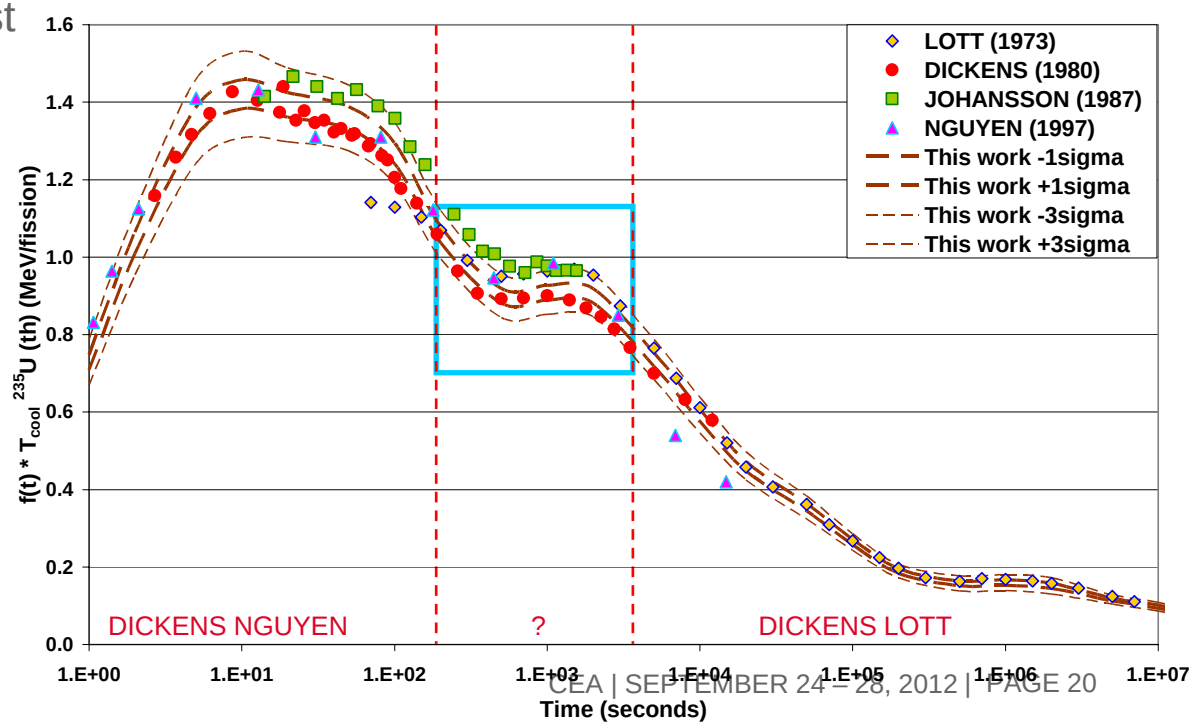
- Questions :
  - Consistency of the library (value + uncertainty) with the measurements ?
  - In case of a use of BFC derived from DARWIN+JEFF3.1.1, what should be the value of the uncertainty of the calculation
  - ¨Parameters of importance for the calculation of the uncertainty of the decay heat ?

Comparison between the calculation and the experiments

- Good consistency of the decay data of JEFF3.1.1,
- Issue at 1 000 seconds
  - Scientific community seems to rely on DICKENS measurements,
  - LOTT, NGUYEN and JOHANSSON agree perfectly
  - NGUYEN and JOHANSSON (end of the studied range of time), LOTT (beginning of the studied range of time)
- In the case of a use of burst fission curves fitted from DARWIN/JEFF3.1.1 values and uncertainty from CyRUS, the overall uncertainty must be ± 3 σ :

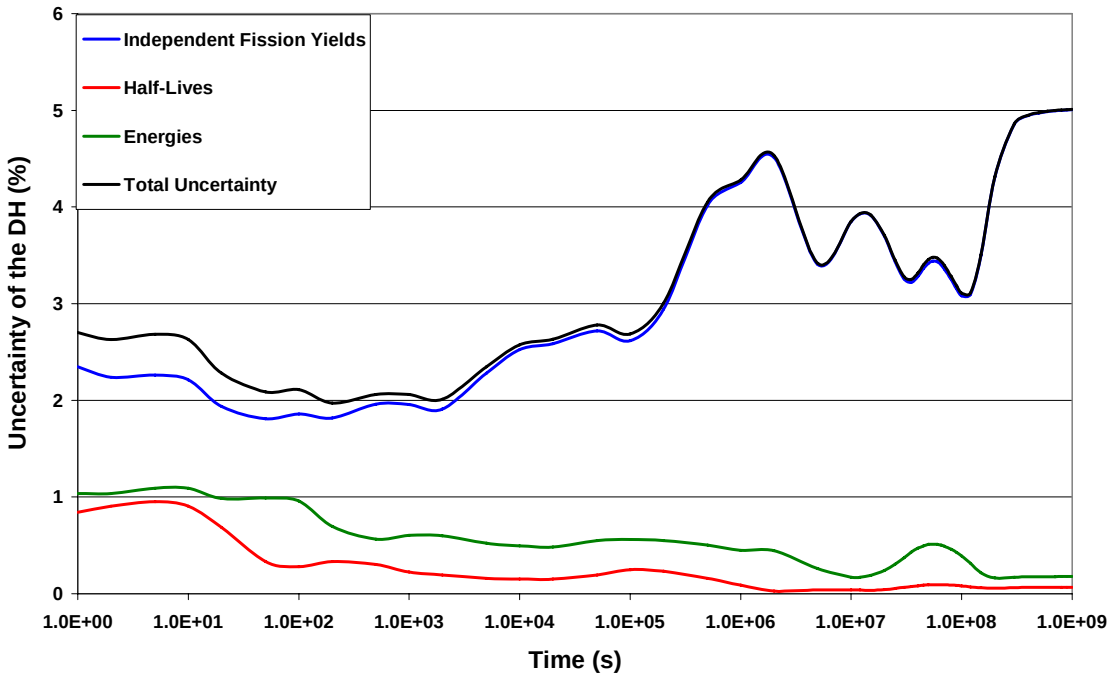
$$9\,\%,\ t \in \left[1;2.10^{5}\right] s$$

$$15\,\%,\ t \in \left[2.10^{5};1.10^{7}\right] s$$



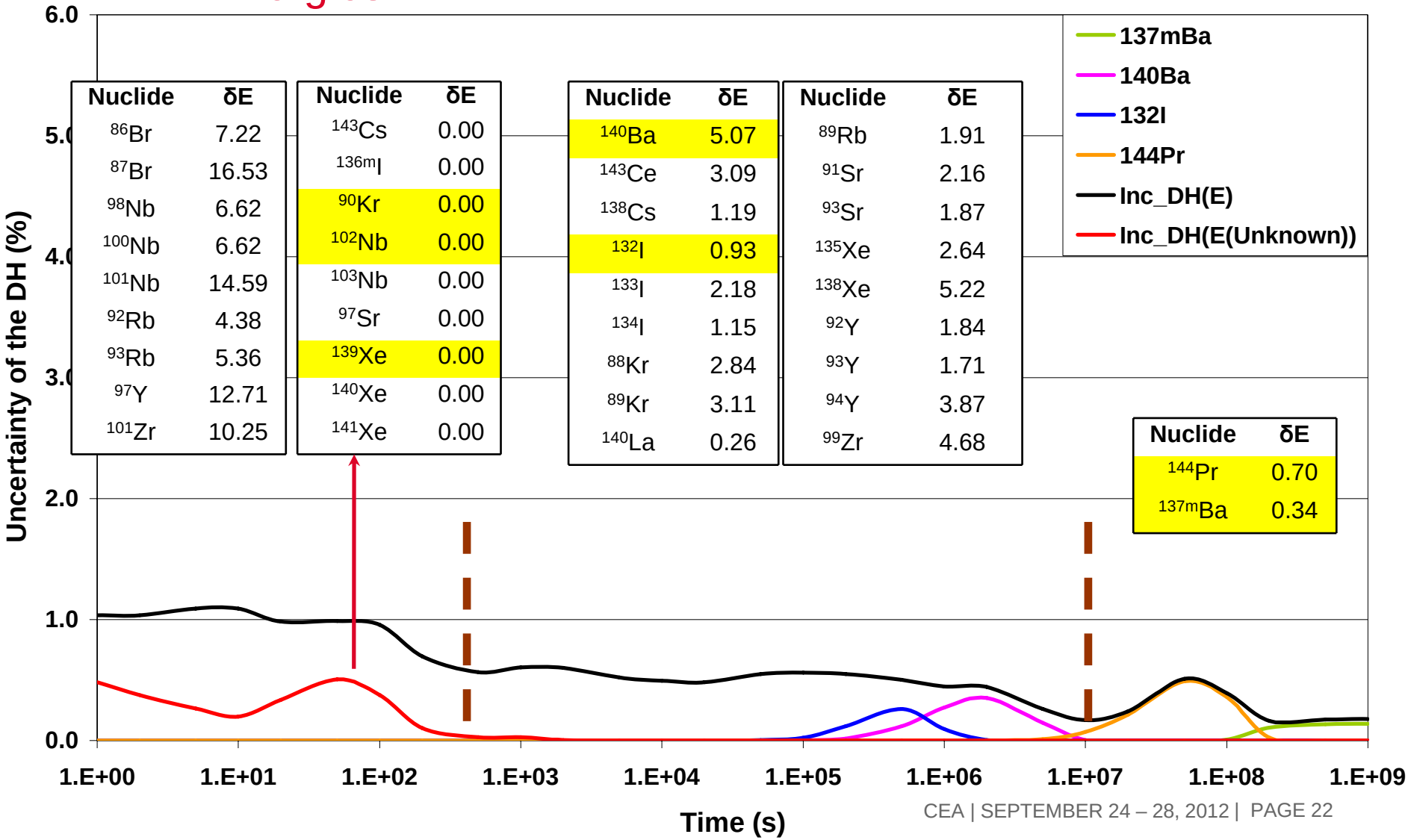
Details about this uncertainty

- Range of the uncertainty of the decay heat [2 % ; 5 %]
- Increase of the uncertainty of the decay heat ↔ Decreasing number of important nuclei + “no correlation” assumption
- Specific structures appear,
- Isotopic concentrations are predominant ↔ independent fission yields



RESULTS

Energies



Nuclide	δE
^{86}Br	7.22
^{87}Br	16.53
^{98}Nb	6.62
^{100}Nb	6.62
^{101}Nb	14.59
^{92}Rb	4.38
^{93}Rb	5.36
^{97}Y	12.71
^{101}Zr	10.25

Nuclide	δE
^{143}Cs	0.00
^{136m}I	0.00
^{90}Kr	0.00
^{102}Nb	0.00
^{103}Nb	0.00
^{97}Sr	0.00
^{139}Xe	0.00
^{140}Xe	0.00
^{141}Xe	0.00

Nuclide	δE
^{140}Ba	5.07
^{143}Ce	3.09
^{138}Cs	1.19
^{132}I	0.93
^{133}I	2.18
^{134}I	1.15
^{88}Kr	2.84
^{89}Kr	3.11
^{140}La	0.26

Nuclide	δE
^{89}Rb	1.91
^{91}Sr	2.16
^{93}Sr	1.87
^{135}Xe	2.64
^{138}Xe	5.22
^{92}Y	1.84
^{93}Y	1.71
^{94}Y	3.87
^{99}Zr	4.68

Nuclide	δE
^{144}Pr	0.70
^{137m}Ba	0.34

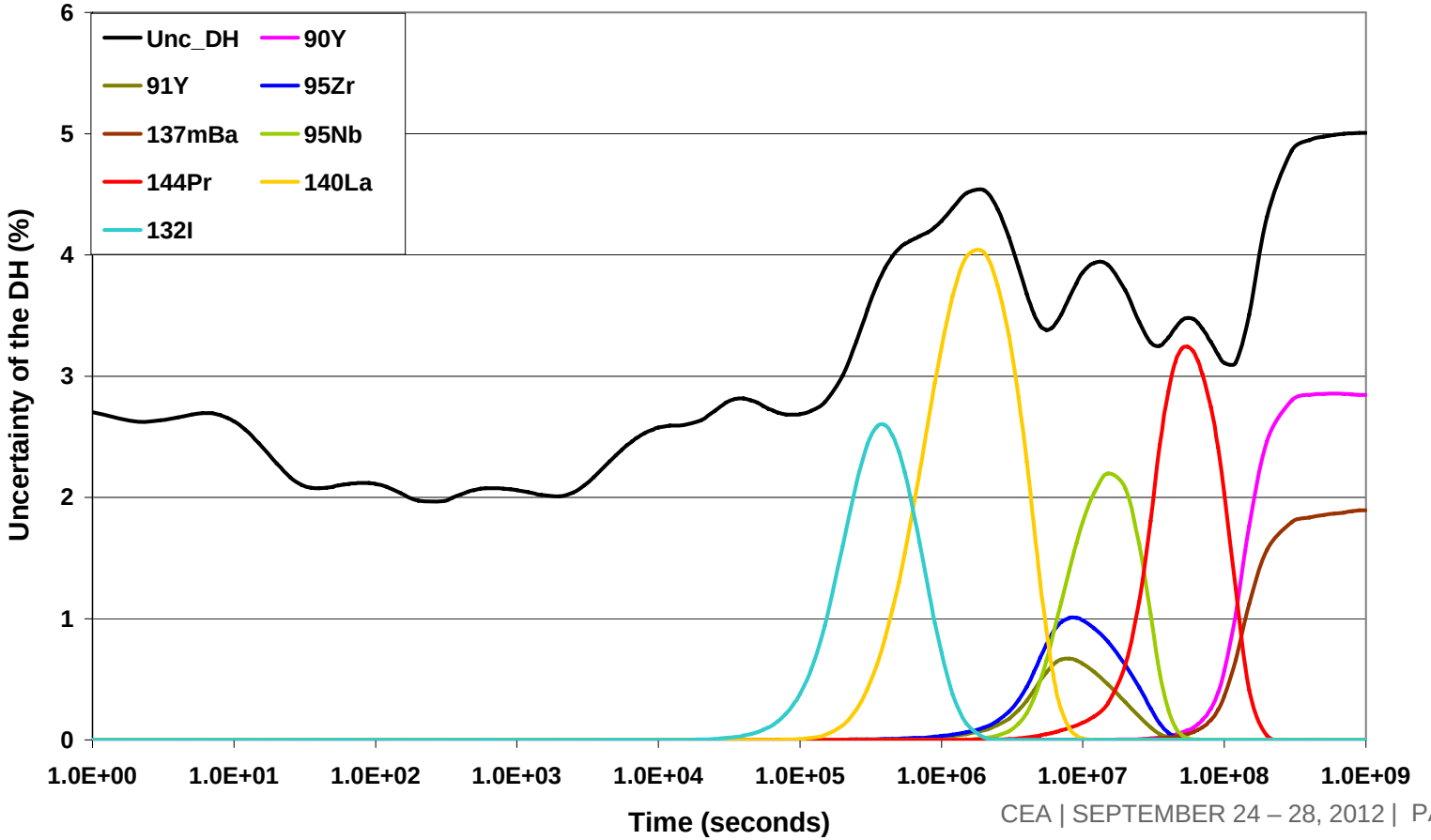
Half-lives

- DH could benefit from an improvement of those nuclei : ^{90}Rb, ^{97m,98m}Y, ^{99}Zr, ^{97m,100,101,102,102m,103}Nb, ^{104,105,107}Mo, ^{102}Tc (JEF/DOC–1413)
- Short Half-lives

NUCLIDES	Half - life	
	Value (s)	Unc. (%)
^{90}Rb	158	3.17
^{97m}Y	1.17	2.56
^{98m}Y	2	10
^{99}Zr	2.2	4.55
^{97m}Nb	52.7	3.42
^{100}Nb	1.5	13.33
^{101}Nb	7.1	4.23
^{102}Nb	1.3	15.39
^{102m}Nb	4.3	9.30
^{103}Nb	1.5	13.33
^{102}Tc	5.28	2.84
^{104}Mo	60	3.33
^{105}Mo	35.6	4.49
^{107}Mo	3.5	14.29

Concentration : Major contributors

- Work is done for cooling times greater than 10^5 seconds → fewer nuclides contribute to the value and the uncertainty of the decay heat.



RESULTS

Cooling time (s)	Ratio of the impact of parameters to the uncertainty of the concentration of the nuclides (%)			
$5{,}0.10^5$	^{132}I			
	$Y^i(^{132}Te)$	58.69		
	$Y^i(^{132}Sb)$	21.27		
	$Y^i(^{132m}Sb)$	11.56		
	$Y^i(^{132}Sn)$	8.2		
$2{,}0.10^6$	^{140}La			
	$Y^i(^{140}Cs)$	82.3		
	$Y^i(^{140}Xe)$	14.26		
	$Y^i(^{140}Ba)$	3.44		
$1{,}0.10^7$	^{91}Y		^{95}Zr	
	$Y^i(^{91}Kr)$	50.55	$Y^i(^{95}Sr)$	57.18
	$Y^i(^{91}Rb)$	47.96	$Y^i(^{95}Y)$	34.17
	$Y^i(^{91}Sr)$	1.12	$Y^i(^{95}Rb)$	8.61
$1{,}5.10^7$	^{95}Nb			
	$Y^i(^{95}Sr)$	57.18		
	$Y^i(^{95}Y)$	34.17		
	$Y^i(^{95}Rb)$	8.61		
$5{,}5.10^7$	^{144}Pr			
	$Y^i(^{144}La)$	92.8		
	$Y^i(^{144}Ba)$	6.9		
$3{,}0.10^8$	^{90}Y		^{137m}Ba	
	$Y^i(^{90}Kr)$	64.72	$Y^i(^{137}Xe)$	82.61
	$Y^i(^{90m}Rb)$	28.24	$Y^i(^{137}I)$	17.03
	$Y^i(^{90}Br)$	6.46		

Nuclides	Ind. Fiss. Yield	δY^i (%)
^{90}Kr	4.50E-02	8.13
^{90m}Rb	7.17E-03	33.72
^{91}Kr	3.28E-02	15.34
^{91}Rb	2.23E-02	21.97
^{95}Sr	4.67E-02	10.03
^{95}Y	1.18E-02	30.55
^{132m}Sb	9.02E-03	18.00
^{132}Sb	1.22E-02	18.00
^{132}Te	1.61E-02	22.70
^{140}Cs	2.11E-02	23.68
^{144}La	8.09E-03	32.14
^{137}Xe	2.73E-02	19.21

CONCLUSION

Conclusion

- Lots of results (sensitivity, correlation, uncertainty),
- It is possible to see the propagation of the uncertainties,
- Validity of a determinist code / Stochastic code,
- Major contributors to the decay heat uncertainty are listed.

Prospects

- Options to be added (Use cumulative fission yields (Y^c) during irradiation when it is possible),
- Check the impact of neutronics for the propagation during irradiation.

Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Cadarache | 13108 St Paul-lès-Durance Cedex
T. +33 (0)4 42 25 31 30 | F. +33 (0)4 42 25 48 49

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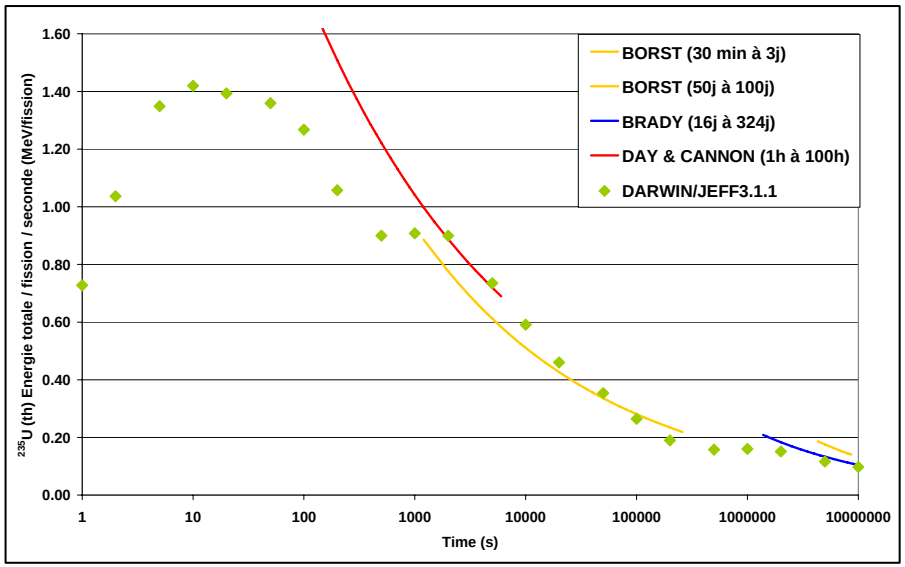
History

- Decay Heat (DH) : Heat produced by radioactivity (after irradiation)
- Discovery
  - First discovered by P. CURIE, A. LABORDE (1903) in radium salts,
  - Theoretical explanation by E. RUTHERFORD and F. SODDY (1904),
  - Related by G.E.M JAUNCEY (1946) Am. J. Phys. *The Early Years of Radioactivity*
- Characterization (Plutonium Project)
  - Build a reactor to produce plutonium → safety
  - Burst Fission Curves
  - BORST, BRADY, DAY & CANNON (1942 – 1943)



Characterization of a parameter

- Nuclear data are measured or calculated from measurements
  → Measurement fluctuations are often normally distributed
  → Nuclear data are normally distributed

- From the normal distribution, it is easy to link the confidence interval to the standard deviation.

- Libraries : parameter = mean value + standard deviation

$$p = p^0 + \delta p$$

Propagation of the variance of nuclear data

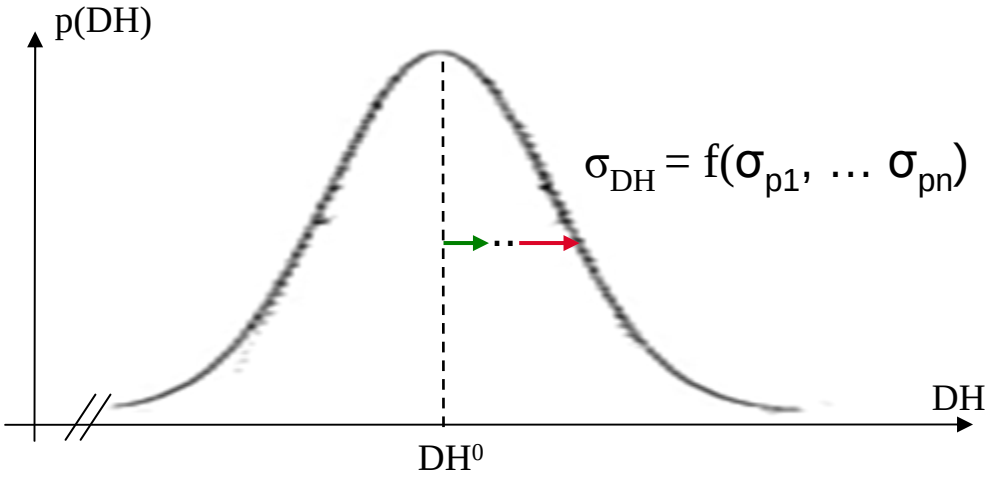
- Link between the variance of the DH and the variance of the parameters : the error propagation formula

$$\mathrm{var}(PR) = \sum_{i=1}^{n} \left(S_{DH/p_i}\right)^2 \mathrm{var}(p_i) + \sum_{i=1}^{n} \sum_{k=1,k\neq i}^{n} S_{DH/p_i} S_{DH/p_k} \sqrt{\mathrm{var}(p_i)}\sqrt{\mathrm{var}(p_k)}\, corr(p_i, p_k)$$

Calculation **Libraries**

- Two hypotheses :
  - 1st order formula
  - DH is normally distributed

Ok (PhD thesis)



RESULTS

Calculation of the uncertainty of a burst fission curve

No irradiation

- Route

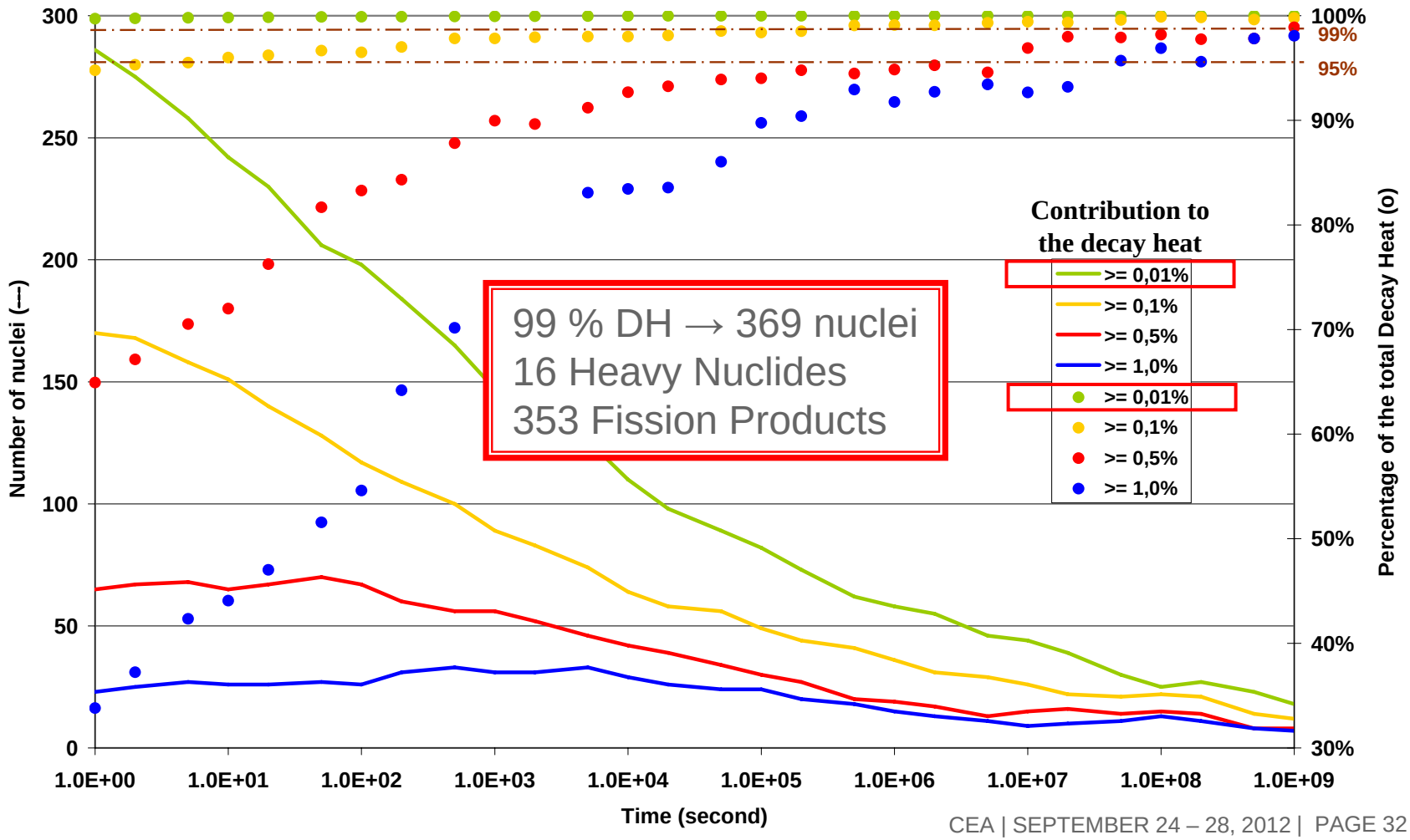
N^0 $\qquad\qquad\qquad\qquad$ N

$$\begin{pmatrix} Y^i_{^{235}U \to N_1}(th) \\ \vdots \\ Y^i_{^{235}U \to N_n}(th) \end{pmatrix} \xrightarrow[\text{Data : } Y^i,\ \lambda,\ br]{\textbf{Equation of evolution during cooling}} \begin{pmatrix} N_1(t) \\ \vdots \\ N_n(t) \end{pmatrix} \xrightarrow[\text{Data : } N,\ \lambda,\ E]{\textbf{Formula of the decay heat}} DH(t)$$

- Moreover
  - Nuclear data : JEFF3.1.1
  - No correlation
    - Ok for λ and E
    - ? For Y^i

Reactor : Sodium Fast Reactor
Irradiation : 1 year
3.0 10^{15} n/cm²/s
fast spectrum
Composition : Fissile assembly

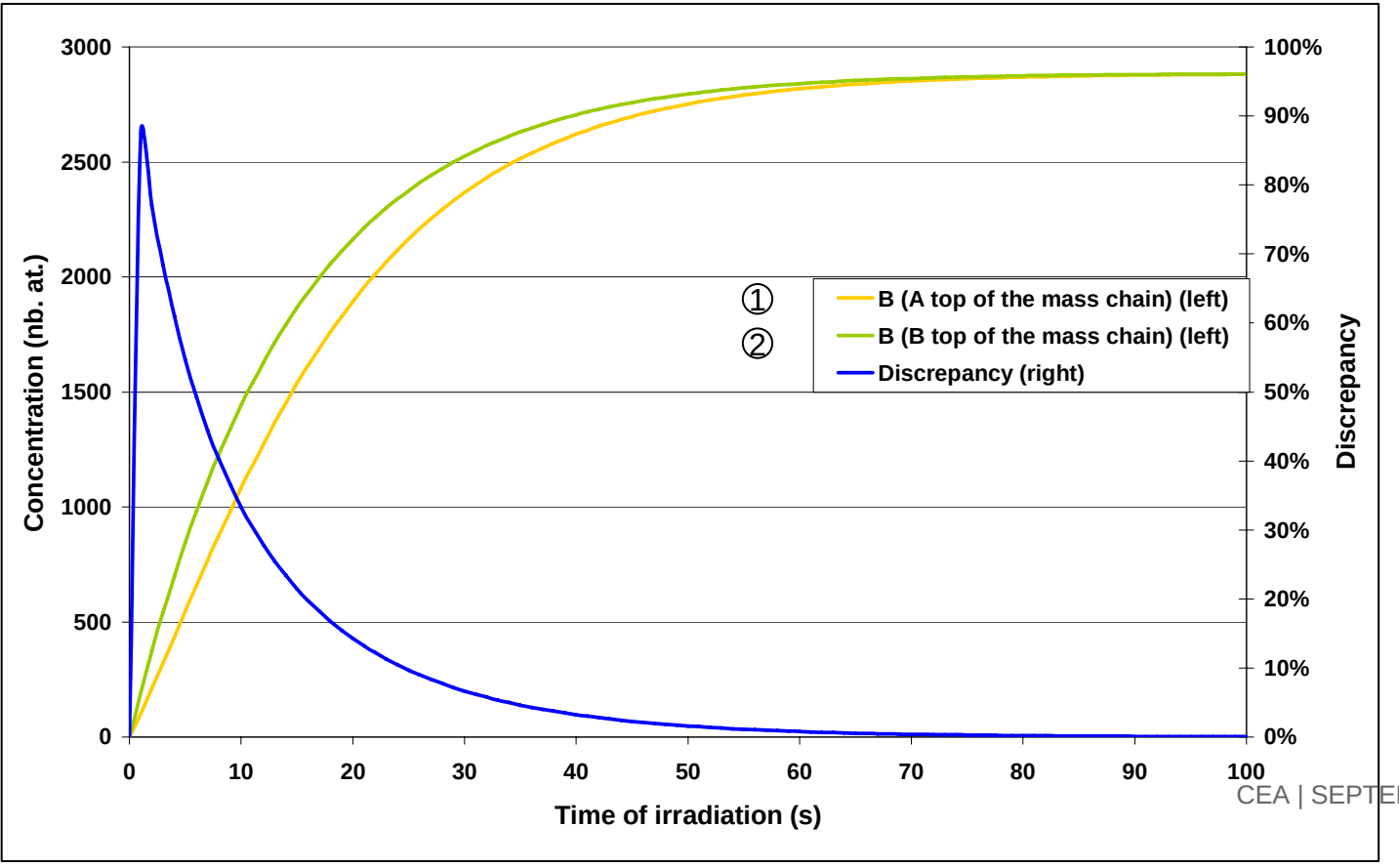
Important nuclei

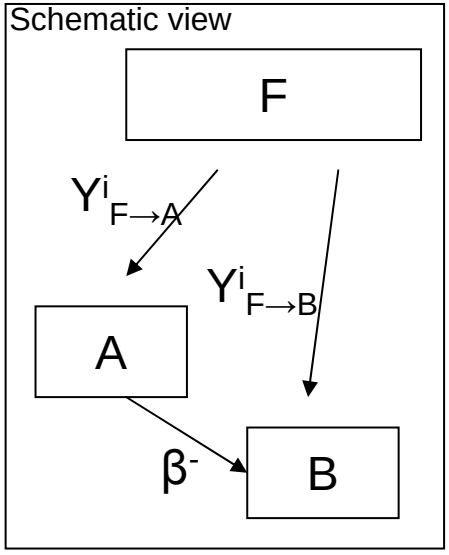


Possible discrepancy in the case of a use of cumulative fission yields

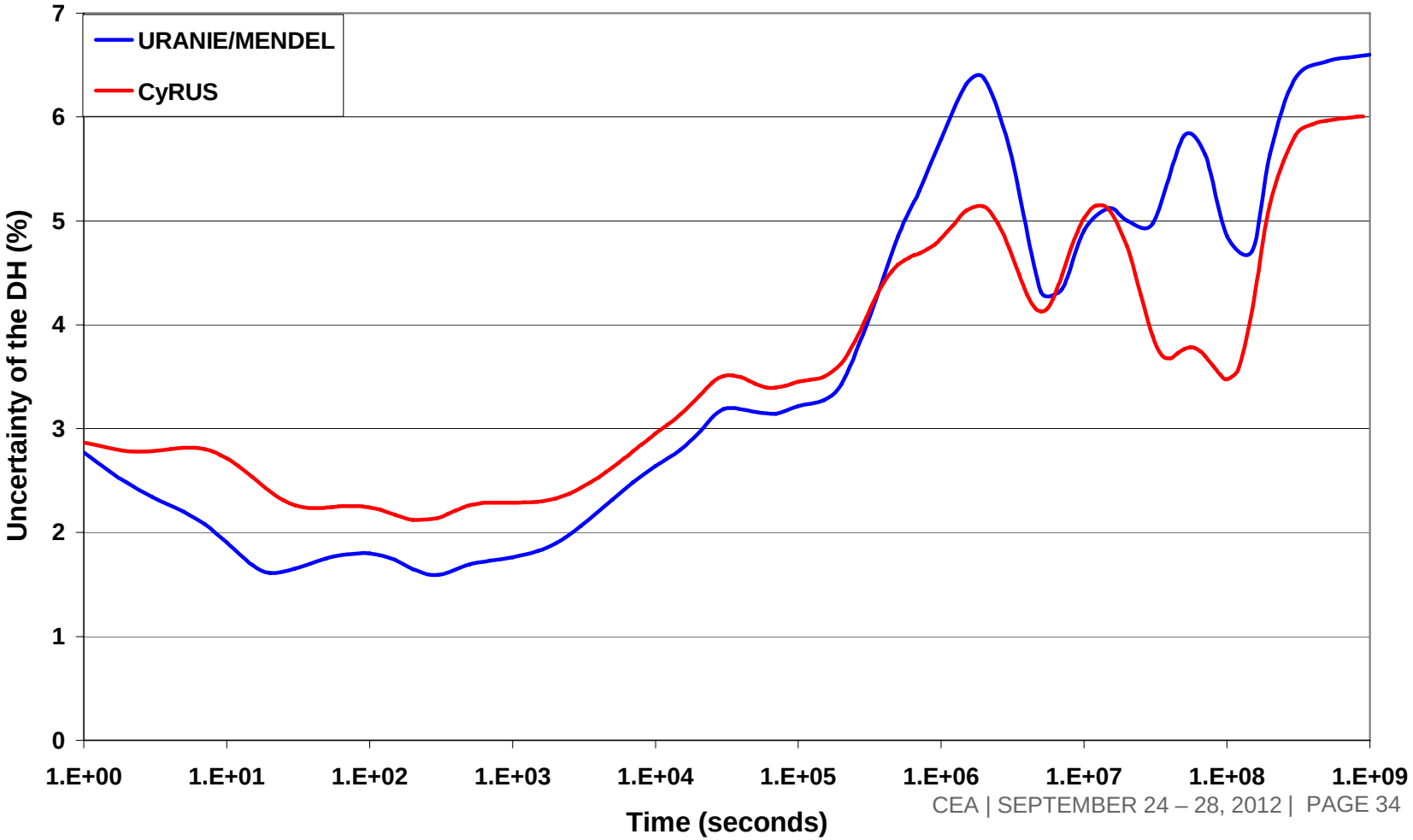
① $$N_B(t) = \frac{\sum_F \left(Y^i_{F\to A} + Y^i_{F\to B}\right)\sigma^f_F \phi N_F}{\lambda_B}\left(1 - e^{-\lambda_B t}\right) + \frac{\sum_F Y^i_{F\to A}\sigma^f_F \phi N_F}{\lambda_B - \lambda_A}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right)$$

② $$N_B(t) = \frac{\sum_F Y^c_{F\to B}\sigma^f_F \phi N_F}{\lambda_B}\left(1 - e^{-\lambda_B t}\right)$$

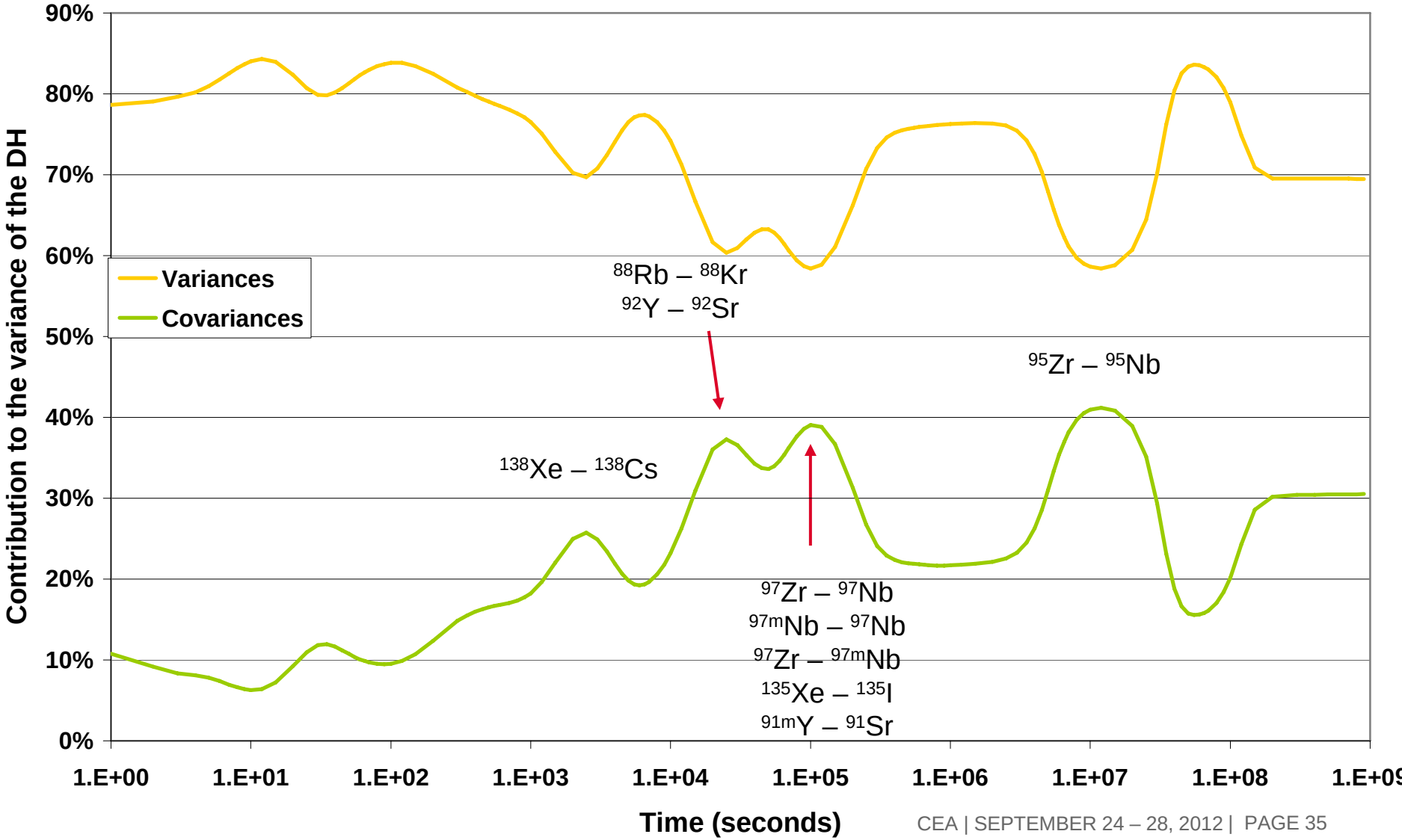




Comparison : Probabilistic - determinist



RESULTS


90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Contribution to the variance of the DH
Variances
Covariances
88Rb – 88Kr
92Y – 92Sr
95Zr – 95Nb
138Xe – 138Cs
97Zr – 97Nb
97mNb – 97Nb
97Zr – 97mNb
135Xe – 135I
91mY – 91Sr
1.E+00
1.E+01
1.E+02
1.E+03
1.E+04
1.E+05
1.E+06
1.E+07
1.E+08
1.E+09
Time (seconds)