Validation of depletion codes for burnup credit evaluation of LWR assemblies

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Abstract

This paper reports the comparison of the CASMO-4E predictions with the radiochemical assay data from assemblies irradiated in Takahama-3 PWR and Fukushima-Daini-2 BWR, and the most recently reported spent fuel data from the VVER-440 assembly irradiated in Novovoronezh 4. Some of the calculations were repeated with the ABURN burnup code, which is a combination of the MCNP4C Monte Carlo code and the ORIGEN2 depletion code. The cross section libraries applied were based on the ENDF/B-VI and the JEF-2.2 data.

KEYWORDS: Burnup credit, validation, isotopic composition, CASMO-4E

1. Introduction

Burnup credit (BUC) means the benefit gained when instead of the fresh-fuel assumption the calculated composition of the irradiated fuel is used in the criticality safety assessment. The use of BUC may result in significant cost savings through more compact fuel storages, by reducing the need for expensive absorber materials, through the use of present containers and storages for new fuel designs, and by allowing more assemblies to be loaded into containers and thereby leading to the reduction in the number of spent fuel shipments.

The level of BUC is defined by the isotopes included in the analysis. In the most conservative case, credit is taken for the integral burnable absorbers and the maximum reactivity of the assembly is estimated with a burnup calculation (Gd credit). In order to apply BUC, a thorough validation of the depletion and criticality calculation codes is required corresponding to the chosen level of BUC. The validation of isotopic calculation codes should primarily be made against experimental data. The lack of suitable data for depletion code validation however is one of the major problems on the way for an efficient use of BUC. The comparison of the codes and the data libraries may be useful especially when experimental data is absent.

This paper reports the comparison of the predictions of the two-dimensional transport theory code CASMO-4E with measured isotopic compositions data obtained from three light-water reactors. The data covered 17x17 PWR assemblies from Takahama-3, a 8x8 BWR assembly from Fukushima-Daini-2, and a VVER-440 assembly from Novovoronezh 4. A burnup code ABURN that combines the MCNP4C Monte Carlo transport code and the ORIGEN2 isotopic depletion code was also applied to some of the problems. Nuclear data from ENDF/B-VI and JEF-2.2 were applied to study the effect of base nuclear data on the isotopic concentrations. Finally, the contribution of the isotopic uncertainties on the uncertainty of k_{eff} in a criticality safety application was studied.

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2. Description of the PIE data

The benchmarks calculated in this paper were based on data from three light-water reactors Takahama-3, Fukushima-Daini-2, and Novovoronezh 4. The basic information of the assemblies and the radiochemical assay data are listed in Table 1. Complete descriptions of the data are available in Reference [1, 2].

PIE data covered all actinides usually included in BUC. In the Takahama-3 and the Fukushima-Daini-2 cases only a few BUC fission products were measured. In the Novovoronezh 4 case the fission products measurements were quite comprehensive. The burnups in the VVER-440 case were determined by matching the calculated Nd-148 concentrations with the measured ones. In the other two cases the burnups were based on the reported values.

Table 1: Characteristics of the post-irradiation examination data.

PIE data	Taka-3 (17x17 PWR)		Fuku-2 (8x8 BWR)		Novo-4 (VVER-440)
	UO_2	UO_2-Gd_2O_3	UO_2	UO_2-Gd_2O_3	UO_2
N. of samples	11	5	8	10	8
U-235 enr. (%)	4.11	2.63	3.90	3.40	3.60
Gd_2O_3 (wt-%)	-	6.0	-	3.0 - 4.5	-
Burnup (a)	$\sim$33	$\sim$21	$\sim$33	$\sim$25	$\sim$38
Actinides (all/buc)	19/12	15/12	19/12	19/12	16/12
FPs (all/buc)	19/7	12/2	19/7	19/7	33/16

(a) Average burnup of the samples (GWd/tU).

3. Codes and methods

CASMO-4 is a two-dimensional transport theory code which can be used for the burnup calculations of PWR and BWR assemblies and the generation of nodal constants for full core calculations. CASMO-4E is an extended version for which JEF-2.2 and ENDF/B-VI based libraries are available. It also has a hexagonal option for calculation of VVER-type assemblies. The calculations were made with the e60201 (ENDF/B-VI) and j20200 (JEF-2.2) libraries.

A burnup code ABURN has been developed at VTT during 2005-2006. The code combines the MCNP4C Monte Carlo transport code and the ORIGEN2 isotopic depletion code for the calculation of fuel depletion in an arbitrary geometry where MCNP4C is used for the calculation of the reaction rates and ORIGEN2 for the subsequent depletion calculation in all depleted zones. The predictor-corrector method is applied to estimate the isotopic concentrations at the end of the depletion step. The main use of the code is the prediction of the isotopic composition of spent fuel. It provides independent means for burnup credit criticality safety and decay heat calculations as well as testing of base nuclear data. The input format is based on the MCNP4C input with additional cards to describe the depletion. An automatic input generation from CASMO-4 input file is being tested to enable easy code-to-code comparisons. ABURN can apply any cross section data compatible with MCNP4C. Preliminary testing has so far been conducted with ENDF/B-VI.8, JEF-2.2, JENDL-3.2 and JEFF-3.1 libraries. The cross section library applied in this paper was prepared at the Royal Institute of Technology (KTH, Sweden) and it was based on the ENDF/B-VI.8 data.

MCNPLINK is a criticality safety analysis script, which combines MCNP4C and CASMO-4/4E for Gd credit analysis of LWR assemblies in various storage geometries. The script performs the depletion calculations with CASMO-4/4E and the criticality calculations with MCNP4C corresponding to the predicted irradiated fuel composition. The script automatically performs also a sensitivity analysis which takes into account the material and fabrication tolerances, depletion code uncertainties, control rod exposure etc. In this paper MCNPLINK was used to estimate the sensitivity coefficients of the BUC isotopes in realistic PWR and BWR spent fuel pools. Also the effect of the isotopic uncertainties on k_{eff} through sensitivity coefficients was studied.

3.1 Depletion calculation - Bias and uncertainty

The fuel depletion was performed in a single assembly geometry with the reflecting boundary conditions. The cycle averaged power and boron histories were used. CASMO-4E was applied in all cases and a single test calculation was performed with ABURN for each assembly type. In the ABURN calculations $1.1 \cdot 10^6$ neutron histories were simulated at each burnup step. The length of the burnup step was set equal to the values used by default by CASMO-4 [3].

The comparison of the calculated and experimental isotopic concentrations were used to calculate the average C/E-value (bias) defined as

$$b_i = (\frac{C}{E})_{i,ave} = \frac{1}{n}\sum_{k=1}^{n}(\frac{\mathrm{C}_{i,k}}{\mathrm{E}_{i,k}}), \tag{1}$$

where i refers to the isotope in question and n is the number of samples. The uncertainty of the bias was estimated with the standard deviation

$$\sigma_i = \sqrt{\frac{1}{n-1}\sum_{k=1}^{n}(\frac{\mathrm{C}_{i,k}}{\mathrm{E}_{i,k}} - (\frac{C}{E})_{i,ave})^2}. \tag{2}$$

3.2 Criticality calculation - Sensitivity coefficients

In case the burnup credit principle is applied, an additional safety margin is needed due to uncertainties in the calculated spent fuel composition. First of all, the isotopic compositions should be corrected for the bias. Typically, a correction that would lead to a decrease of k_{eff} is not allowed. The correction for the bias and the uncertainties can also be done simultaneously by using correction factors

$$c_i = 1/b_i \pm 2 \cdot \sigma_i, \tag{3}$$

where c_i designates the correction factor for isotope *i*. Negative sign is used for the absorbers and positive sign for the fissile isotopes. Each isotopic concentration is multiplied by the corresponding correction factor before criticality calculation takes place. The use of the correction factors shown above however yields a very conservative estimate for the k_{eff}.

A more realistic way to estimate the additional safety margin is to consider the isotopic uncertainties independently and to combine the individual contributions statistically. The procedure can be done in two phases. In the first phase the concentrations are corrected for the bias and the k_{eff} is estimated. This gives the bias contribution to the safety margin. In the second phase the sensitivity coefficients of the nuclides are determined in the problem geometry. The isotopic uncertainties and the sensitivity coefficients can then be combined to obtain

$$\delta k_{\text{eff, stat}} = k_{\text{eff}} \cdot \sqrt{\sum_i (S_i \cdot \delta n_i)^2}, \quad (4)$$

where S_i is the sensitivity coefficient of k_{eff} to the concentration of nuclide i and δn_i is the uncertainty of isotope i estimated as $\delta n_i = 2\sigma_i$. The safety margin is the then the sum of the bias and the statistical terms

$$\Delta k_{\text{eff, best-estimate}} = (k_{\text{eff, bias}} - k_{\text{eff}}) + \delta k_{\text{eff, stat}}. \quad (5)$$

Another approach is to estimate the effect of each isotopic uncertainty directly by determining the $k_{\text{eff, i}}$ after correcting for the uncertainty of isotope i. Then the statistical contribution to the safety margin would be

$$\delta k_{\text{eff, stat}} = \sqrt{\sum_i (k_{\text{eff, i}} - k_{\text{eff}})^2}, \quad (6)$$

MCNPLINK was used to estimate the sensitivity coefficients of the BUC isotopes in some real PWR and BWR spent fuel pools. The square boron steel racks with 1.0 wt-% and 1.6 wt-% boron were used in the PWR and the BWR cases, respectively. The necessary input files were created with MCNPLINK and the criticality calculations were done with CASMO-4E and its ENDF/B-VI-based library.

4. Results and discussion

The time dependence of the calculated isotopic compositions in the BWR case is illustrated in Figures 1 - 3, where the results of the CASMO-4E and ABURN calculations are compared. The calculated infinite multiplication factors (Figure 1) agree very well. The case without burnable absorbers was also added to the figure. It can be observed that the effect of Gd in the beginning of irradiation is about 35 000 pcm. Figures 2 and 3 show the depletion of burnable absorbers Gd-155 and Gd-157 in the BWR assembly. The predictions of CASMO-4E and ABURN are very similar in this respect too. The difference of about 25 % in the calculated Gd-155 concentrations at the maximum reactivity burnup explains the small difference of about 250 pcm in the k_{inf}. The comparison of the end-of-life isotopic compositions calculated with CASMO-4E and ABURN is shown in Table 3.

The biases and standard deviations obtained from the comparison of CASMO-4E predictions and Takahama-3 measurements are shown in Figures 4 and 5, respectively. The results include both UO_2 and UO_2-Gd_2O_3 samples (total 13). The U-235, U-238, Pu-239 and Pu-241 concentrations are predicted with less than 5 % biases. A large overprediction of the Np-237 concentration is seen in the UO_2-Gd_2O_3 samples which also contributed significantly to the uncertainty (standard deviation) of the nuclide. The biases of the Am, Cm and Sm isotopes remained quite large.

The same is depicted for the BWR case in Figures 6 and 7. The results include both UO_2 and UO_2-Gd_2O_3 samples (total 14). The biases in the Pu-239 and Pu-241 concentrations are large. This may be due to inadequate modeling of the void history since the main contribution of the bias came from samples taken from similar heights (void fractions 30 - 65 %). The void history applied was based on Reference [4]. The uncertainties of the important nuclides U-235, Pu-239, Pu-241, and several other actinides and fission products are about twice as large as in the PWR case.

Figures 8 and 9 show the biases and uncertainties obtained in the VVER-440 case. Most of the biases are in an acceptable range. In the case of Np-237, Ag-109, Sm-147 and some other isotopes the deviations from the experimental values are large. The standard deviation of U-234 is almost 40 % which may be due to uncertainties in the initial composition. The uncertainties of Cs-134, Nd-142, Sm-151, and Am isotopes are large. The results of CASMO-4E and ABURN agreed quite well. This implies that the uncertainties in the VVER-440 case are most likely due to the uncertainties in the reported initial composition, operating history and the measurements.

The results of the sensitivity calculations for the PWR and the BWR pools are shown in Table 2. Only the nuclides present in the PIE data and with the largest uncertainty effects are included in the table. It was seen that the uncertainties in the PWR case were quite acceptable. On the other hand the uncertainties in the BWR case remained large. The most important nuclides contributing to the overall uncertainty proved to be Pu-239, U-235, Pu-241, Np-237, Pu-240 and Sm-149. The use of the two different methods (Equations (4) and (6)) yielded similar results.

It seems that it is difficult to demonstrate the accuracy of the single assembly burnup codes reliably enough with the publicly available radiochemical assay data and the simple description of the irradiation. This is true at least in the BWR and the VVER-440 cases where some the isotopic biases and uncertainties remained above acceptable levels to be applied to BUC. A more accurate 3-D modeling of the irradiation must be considered. Also, more data aimed at BUC purposes is needed in order to make necessary trend analyses for the isotopic biases. The predictions of the independent codes were very similar and the differences between the results obtained with different cross section libraries remained quite small. This gives further assurance that the source of the uncertainty lies mainly in the modeling of the irradiation and the quality of the experimental data rather than in the accuracy of the codes and the base nuclear data[a].

References

1) Spent fuel composition database (SFCOMPO), http://www.nea.fr/html/science/wpncs/sfcompo/

2) L. J. Jardine, ”Radiochemical assays of irradiated VVER-440 fuel for use in spent fuel burnup credit activities”, UCRL-TR-212202, (2005).

3) M. Edenius et al, CASMO-4 ”A Fuel Assembly Burnup Program”, STUDSVIK/SOA-95/1, (1995).

4) Y. Hakahara et al, ”Nuclide Composition Benchmark Data Set for Verifying Burnup Codes on Spent Light Water Reactor Fuels”, Nuclear Technology, **137**, (2002).

[a] It must be noted that concentrations of some other nuclides with minor significance to BUC (such as Cm) were sensitive to the cross section library used.

Table 2: The results of the sensitivity calculations made with MCNPLINK.

	PWR pool (38 GWd/tU)			BWR pool (38 GWd/tU)		
Nuclide	S_i(a)	δk_1(b)	δk_2(c)	S_i(a)	δk_1(b)	δk_2(c)
Pu-239	1.88e-01	360	365	1.64e-01	1287	1284
U-235	1.52e-01	210	212	2.00e-01	1285	1288
Pu-241	7.17e-02	152	153	5.71e-02	391	394
Np-237	-4.31e-03	113	112	-3.58e-03	50	50
Pu-240	-6.70e-02	99	61	-5.90e-02	269	179
Sm-149	-7.99e-03	62	60	-6.87e-03	143	142
Sm-151	-7.63e-03	21	21	-6.85e-03	42	42
Nd-143	-1.28e-02	21	20	-1.51e-02	30	30
Am-241	-1.39e-03	21	20	-1.01e-03	13	13
Eu-154	-2.73e-03	10	10	-2.10e-03	21	21
U-234	-7.90e-04	9	8	-1.13e-03	7	7
Sm-152	-3.72e-03	8	8	-3.92e-03	34	34

(a) Sensitivity coefficient $\delta k[\%]/\delta n[\%]$.
(b) $\delta k_1 = \mathrm{k_{eff}} \cdot |S_i \cdot 2\sigma_i|$ (pcm), see Equation (4).
(c) $\delta k_2 = \mathrm{k_{eff,\,i}} - \mathrm{k_{eff}}$ (pcm), see Equation (6).

Figure 1: Infinite multiplication factors as a function of burnup calculated with CASMO-4E and ABURN. The case without burnable absorbers is also included in the figure.

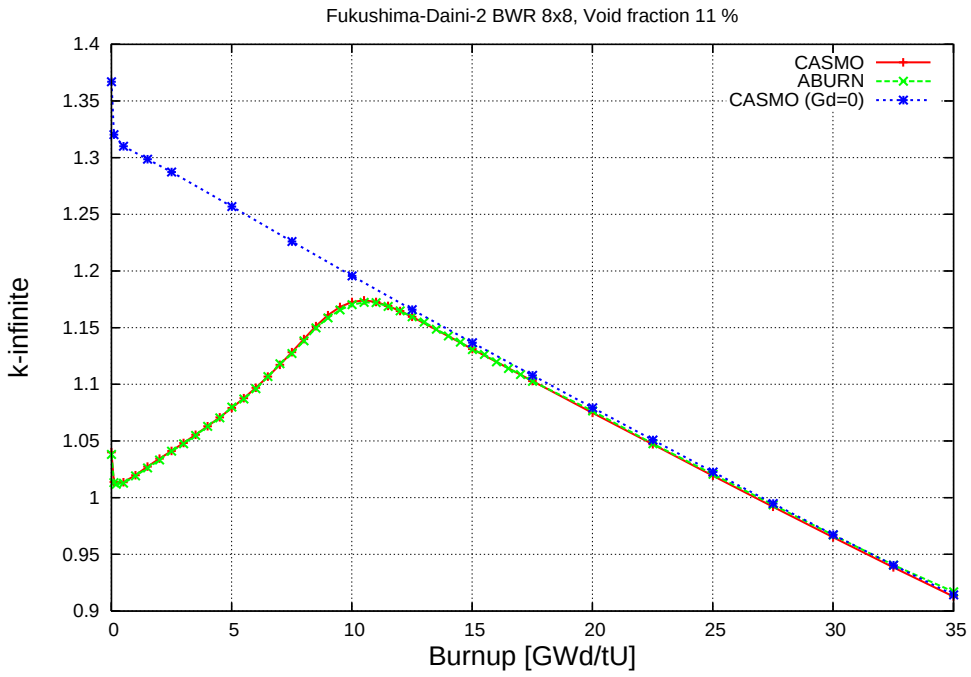

Figure 2: Comparison of the depletion of Gd-155 calculated with CASMO-4E and ABURN. The difference is shown in ten annular regions, where the numbering begins from the center.

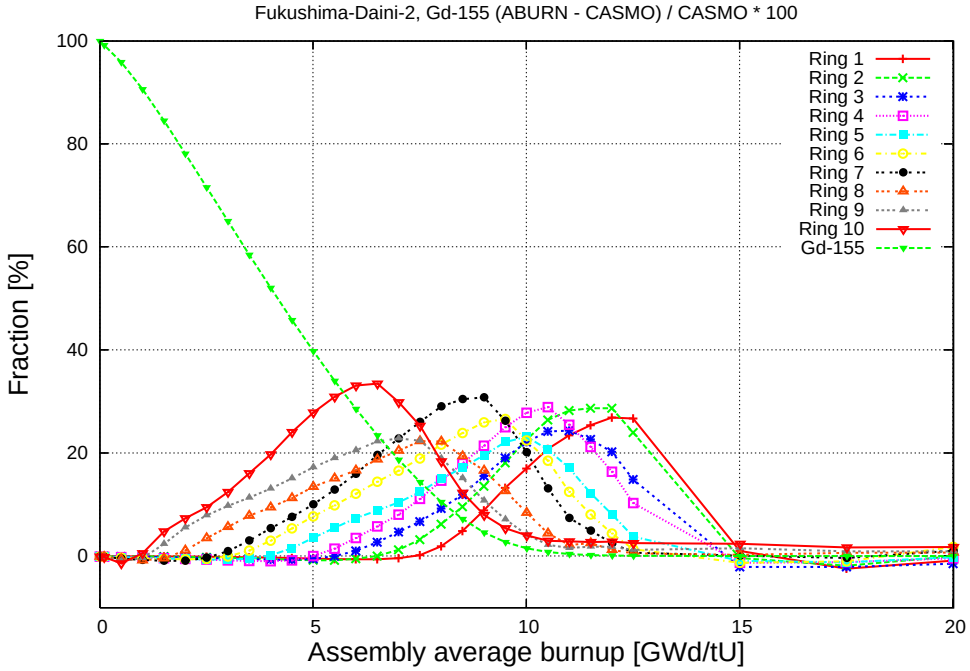

Figure 3: Comparison of the depletion of Gd-157 calculated with CASMO-4E and ABURN. The difference is shown in ten annular regions, where the numbering begins from the center.

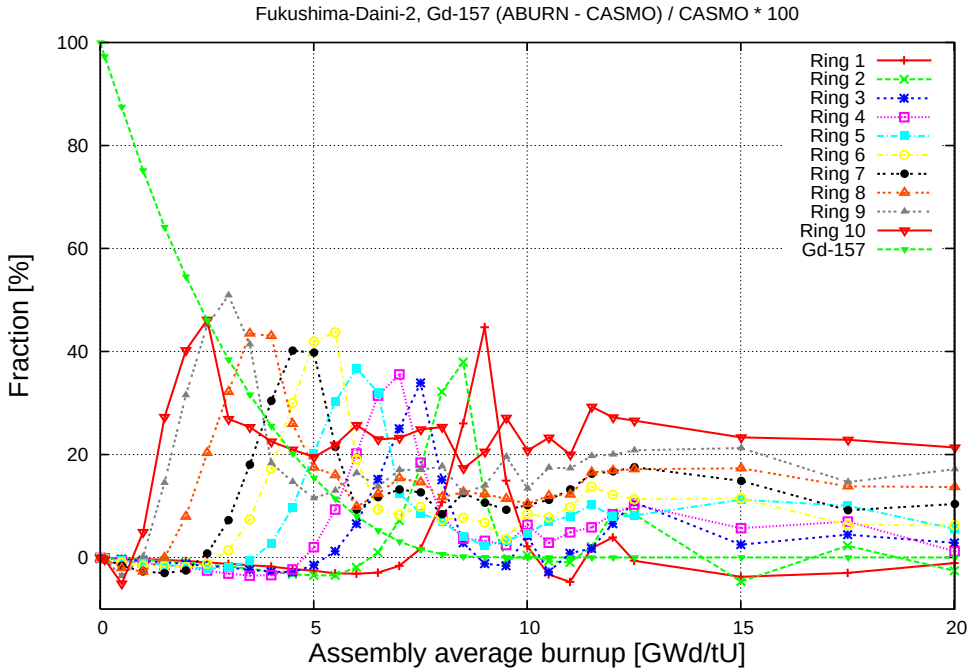

Table 3: Comparison of the calculated end-of-life nuclide concentrations (ABURN/CASMO-4E).

Sample	sf97-4	sf98-4	s149
Reactor	(PWR)	(BWR)	(VVER)
GWd/tU	47.03	42.35	43.41
Nuclide	A/C(a)	A/C	A/C
U-234	0.98	0.99	0.97
U-235	1.01	1.01	0.99
U-236	0.99	1.00	0.99
U-238	1.00	1.00	1.00
Np-237	1.00	1.00	1.00
Pu-238	1.02	1.04	1.02
Pu-239	1.02	1.00	1.03
Pu-240	1.06	1.06	1.07
Pu-241	1.02	1.01	1.01
Pu-242	1.02	1.03	1.02
Am-241	1.04	1.03	1.02
Am242m	1.06	1.07	1.04
Am-243	0.80	0.89	0.78
Cm-242	0.99	1.01	0.98
Cm-244	1.62	1.78	1.57
Mo-95	1.01	1.00	1.01
Tc-99	1.06	1.04	1.07
Ru-101	0.99	0.99	1.00
Ag-109	1.04	1.05	1.04
Cs-133	0.98	0.99	0.98
Cs-134	1.03	1.04	1.03
Cs-135	1.07	1.08	0.98
Cs-137	0.99	0.99	0.99
Sm-147	0.88	0.91	0.88
Sm-150	1.14	1.06	1.14
Sm-151	0.90	0.95	0.90
Sm-152	0.87	0.91	0.88
Nd-143	1.00	1.00	1.00
Nd-145	0.98	0.98	0.99
Nd-148	1.01	1.00	1.01
Eu-153	1.00	1.02	1.00
Eu-154	0.89	0.88	0.88

(a) A/C = ABURN / CASMO-4E.

Figure 4: Biases calculated from the comparison of CASMO-4E with Takahama-3 data.

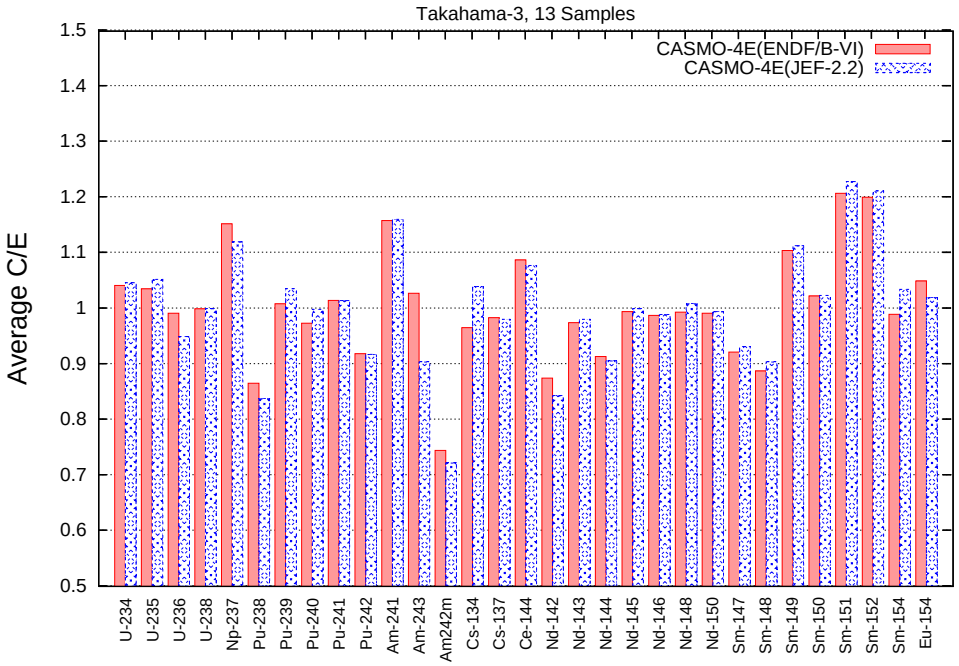

Figure 5: Standard deviations calculated from the comparison of CASMO-4E with Takahama-3 data.

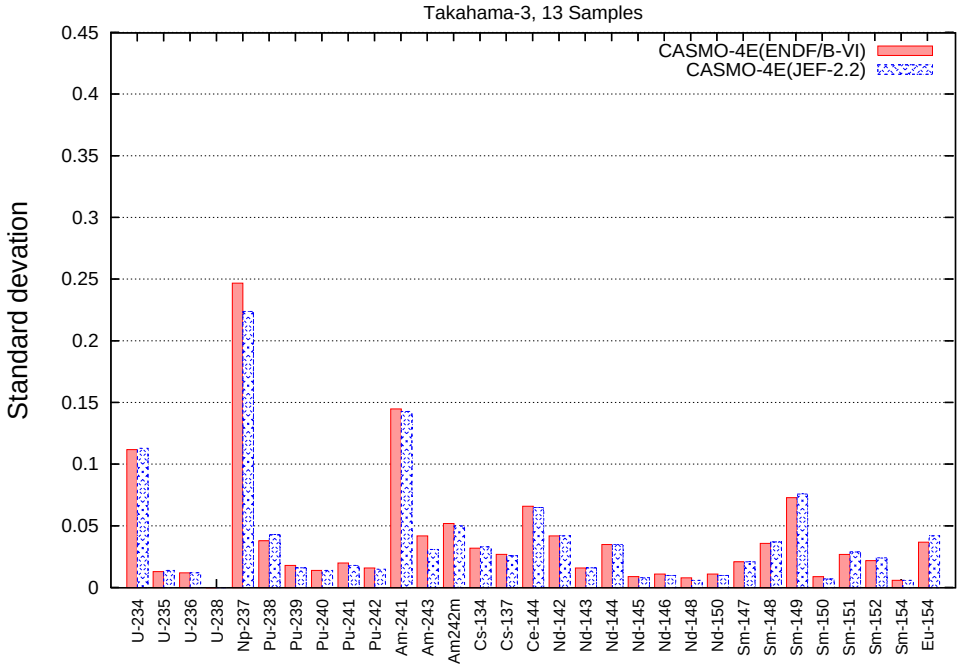

Figure 6: Biases calculated from the comparison of CASMO-4E with Fukushima-Daini-2 data.

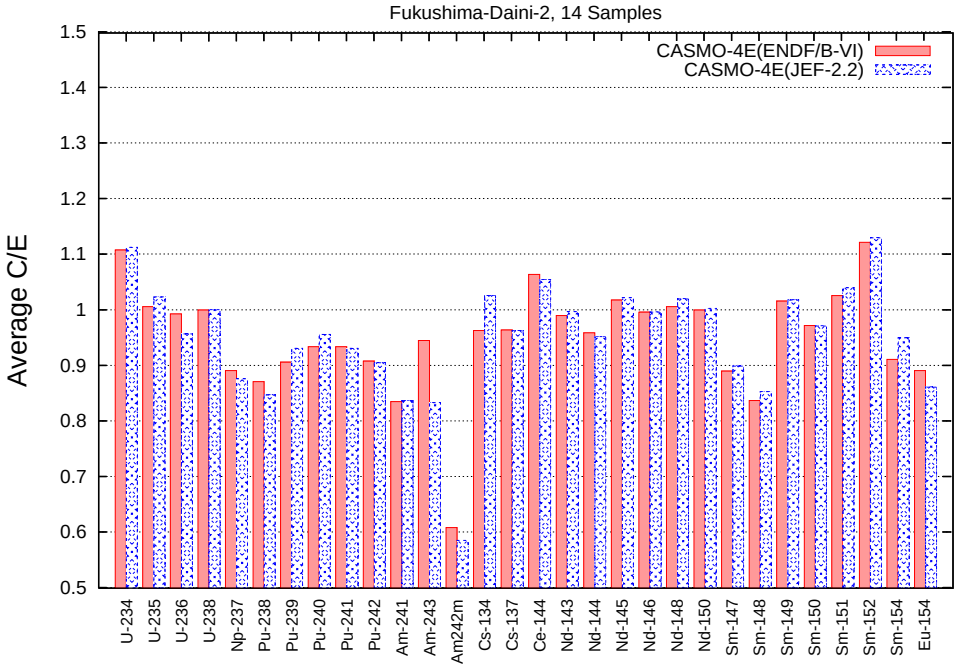

Figure 7: Standard deviations calculated from the comparison of CASMO-4E with Fukushima-Daini-2 data.

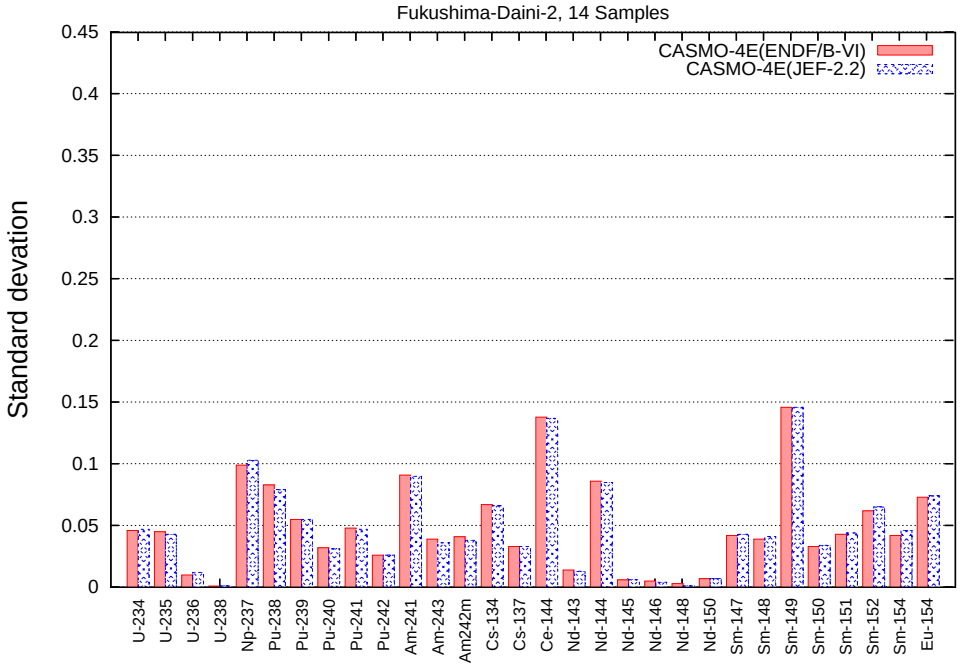

Figure 8: Biases calculated from the comparison of CASMO-4E with Novovoronezh 4 data.

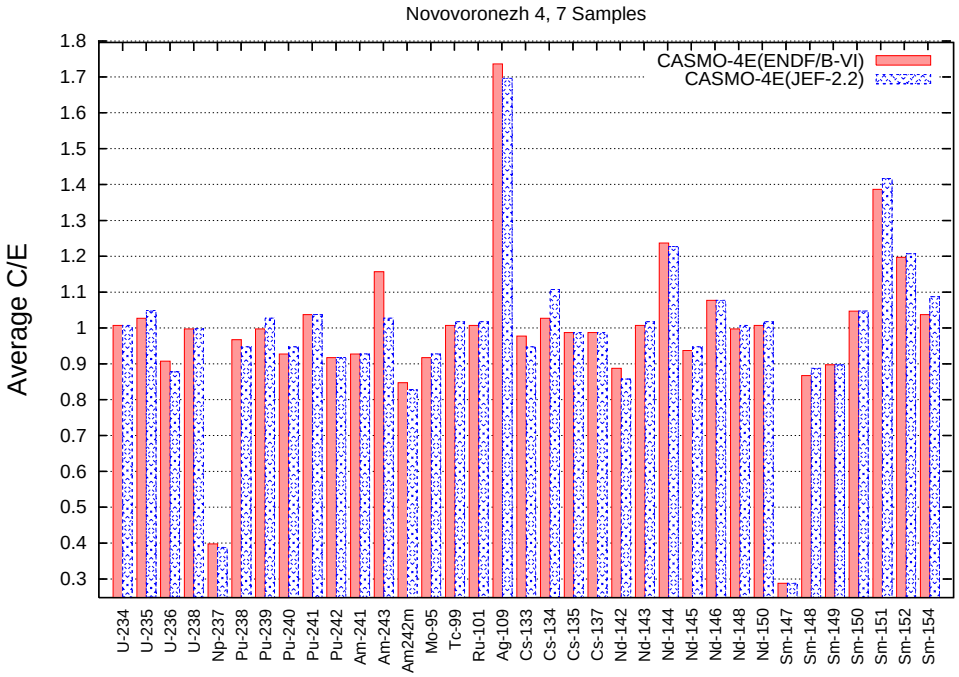

Figure 9: Standard deviations calculated from the comparison of CASMO-4E with Novovoronezh 4 data.