Inter-code Comparison for Nitrate Plutonium Solutions using JEF2.2

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ABSTRACT

Recent JEF2.2 benchmark studies have shown important discrepancies between calculations and experiments. The general tendencies observed in independent studies by GULLIFORD et al. [1, 2] and by NOURI et al. [3, 4] are summarised here :

For uranium oxide and mixed oxide lattices, good agreements between calculations and experiments have been found. This confirms the previous benchmark studies performed in the framework of reactor physics [5,6]

For homogeneous fuel (uranium, plutonium, or mixed U+Pu solutions or UO_2 powder) , the agreements are less satisfactory and a systematic over-prediction has been observed.

Although the general conclusions of these studies are consistent, important discrepancies have been pointed out for the configurations containing homogeneous fuel. It has therefore been decided to carry out comparisons between different calculation schemes which use JEF2.2 nuclear data in order to understand the origin of the discrepancies. We believe that this work is required before providing feedback to the nuclear data in JEF, to avoid the possibility of calculation inaccuracy distorting the conclusions.

In this paper, we focus our investigations on configurations containing nitrate plutonium solutions. We have selected a set of experiments for which the calculation results obtained by MONK and APOLLO-1 + MORET-3 are first presented and compared. In the second paragraph, a reduced set of configurations have been selected in order to better understand the origin of the discrepancies between the two calculation schemes. The most important parameters describing such media have been taken into account (various Pu concentrations in the solutions and proportions of ^{240}Pu in the Pu). Simplified configurations have been defined and code comparisons have been commenced. The participants involved in these code comparisons are summarised in paragraph four. A first attempt to analyse the results is presented in the fifth paragraph, followed by a provisional conclusion.

I- EXPERIMENTAL BENCHMARKS AND CALCULATION RESULTS

Two experimental configurations, involving nitrate plutonium solutions have been studied. Both are part of the recently evaluated benchmarks selected by the OECD working group « International Criticality Safety Benchmark Evaluation Program » [7].

The first serie of experiments was performed in VALDUC with plutonium containing 19 % of ^{240}Pu. The solution was contained in a large cubic tank (130 x 130 x Hc cm^3, where Hc is the critical height) reflected by 20 cm of water. The plutonium concentration ranged from 13.2 g/l to 105 g/l.

The second serie of experiments was performed in Battle Pacific Northwest Laboratories (PNL). The ^{240}Pu proportion varies from 3 % to 5 % and the plutonium concentration ranged from 27 g/l to 105 g/l.

These benchmarks have been calculated using JEF2.2 based libraries with the UK criticality Monte-Carlo MONK code (12630 groups) and the French criticality system of codes APOLLO-1 + MORET-3 (99 groups + self-shielding). The results are presented in the figures 1 and 2 as a function of the slowing-down current, q, calculated by APOLLO-1 as the number of neutrons which become thermal for one emitted fission neutron.

The following remarks could be drawn :

Both codes over-predict the keff values for VALDUC experiments (0.8 % higher than unity for MORET and 1.9 % over-prediction for MONK).

For PNL configurations, MORET results are consistent with the experiments while MONK over-predicts by 1.4 %.

The difference between the two codes ranges from 0.5 % to 2.3 %. MONK obtains systematically higher results. An average discrepancy of 1.4 % is observed for PNL experiments and 1.1 % for VALDUC experiments. No clear tendency with the spectrum softness is observed.

II- REDUCED SET OF NITRATE Pu BENCHMARKS
AND SUBMISSION SPECIFICATIONS

A reduced set of experiments has been selected for a more detailed inter-code comparison. Three steps have been adopted for de-coupling and evaluating the different sources of discrepancies. The codes will first be compared for infinite media; then the effect of leakage calculation will be evaluated in bare sphere configurations; finally, the inter-code comparison for the experimental configurations will give the effect of computing the water reflection. This reduced set considers six configurations where the plutonium concentration ranges from 13 g/l to 119 g/l and the ^{240}Pu proportion varies from 3 % to 19 %. The following table gives the atomic number density (in 10^{24} atoms / cm^3) and the radius for the bare spheres calculations obtained by critical buckling conversion where the material buckling was obtained by APOLLO-1.

Case #	1	2	3	4	5	6
Reference[1]	001-C003	002-C02	006-C03	012-C05	012-C06	012-C08
C(Pu) g/l	119	51.42	26.97	13.2	105	52.7
^{240}Pu % in Pu	4.6	3.1	3.1	18.9	18.9	18.9
^{238}Pu	1.8063^E-8	-	-	-	-	-
^{239}Pu	2.8482^E-4	1.2549^E-4	6.5822^E-5	2.4713^E-5	1.9658^E-4	9.8665^E-5
^{240}Pu	1.3935^E-5	4.0246^E-6	2.1109^E-6	6.2620^E-6	4.9811^E-5	2.5000^E-5
^{241}Pu	9.0672^E-7	-	-	1.8562^E-6	1.4766^E-5	7.4109^E-6
^{242}Pu	2.6645^E-8	-	-	3.7496^E-7	2.9827^E-6	1.4970^E-6
^{241}Am	-	-	-	2.0116^E-7	1.6001^E-6	8.0310^E-7
N	2.4117^E-3	1.5890^E-3	2.0666^E-3	1.3717^E-3	2.5139^E-3	1.7850^E-3
O	3.0741^E-2	3.5895^E-2	3.6223^E-2	3.5301^E-2	3.7250^E-2	3.5957^E-2
H	6.1482^E-2	6.3356^E-2	6.1880^E-2	6.3594^E-2	6.0760^E-2	6.2402^E-2
Fe	-	2.9331^E-6	1.0352^E-6	3.5584^E-6	2.4802^E-5	1.2185^E-5
Cr	-	-	-	1.1443^E-6	7.9755^E-6	3.9184^E-6
Ni	-	-	-	8.1104^E-7	5.6527^E-6	2.7772^E-6
Radius for bare sphere (in cm)	17.91	18.47	22.09	53.52	21.41	22.25

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[1] all these experiments are described in reference 7. For instance, 001-C03 is the experiment described in the chapter Pu-SOL-THERM-001 for experimental case number 03.

In order to make the inter-code comparison easier, participants are asked to follow the form described below for the submission of results.

⇨ For the 2 groups comparison the energy cut-off is 2 eV.

⇨ The energy boundaries (in MeV) in the 15 groups energy structure are :

19.64 (or the upper energy limit) - 6.06 - 2.23 - 1.35 - 0.498 - 0.183 - 0.0674 - 0.0248-0.00912 - 0.00203 - 454.E-06 - 22.6E-06 - 4.E-06 - 0.53E-06 - 0.1E-06 -0.00011E-06 (or the lower energy limit)

⇨ Infinite media : the results required for 6 cases are : kinf, fluxes, reaction rates in both 2 groups and 15 groups.

Results for each case may be sent in the following form :

kinf
------
flux group 1
flux group 2
------
^{239}Pu absorption group 1
^{239}Pu absorption group 2
^{239}Pu production group 1
^{239}Pu production group 2
^{239}Pu fission group 1
^{239}Pu fission group 2
^{240}Pu absorption group 1
^{240}Pu absorption group 2
------
flux group 1
....
flux group 15
------
^{239}Pu absorption group 1
....
^{239}Pu absorption group 15
^{239}Pu production group 1
....
^{239}Pu production group 15
^{239}Pu fission group 1
....
^{239}Pu fission group 15
^{240}Pu absorption group 1
....
^{240}Pu absorption group 15

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⇨ Bare sphere : those who are carrying out calculations with codes using macroscopic cross section (and do not calculate the separate reaction rates) need to submit only the following information for each case :

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keff
proportion of leakage

For the others contributors, the following form maybe used for the submission of the results of each case :

-----
keff
proportion of leakage
-----

flux, ^{239}Pu absorption, production and fission and ^{240}Pu absorption reaction rates in 15 groups structure are also required. Please follow the form specified for infinite media.

⇨ Experimental configurations : Only keff and proportion of leakage are required.

III- PROVISORY LIST OF PARTICIPANTS

Up to now, the following participants have submitted complete or partial results. Since the comparison is still running, other participations are welcomed.

APOLLO-1 (99 groups), APOLLO-2 (99 or 172 groups) and MORET-3 have been used by A. NOURI, G. POULLOT and G. COURTOIS (Fontenay-aux-Roses),

MONK-7 (12630 groups), WIMS-7 (69 or 172 groups) have been used by N. SMITH, C. DEAN and D. HANLON (Winfrith)

TRIPOLI-4 (pointwise) and APOLLO-2 + TRIMARAN-2 (99 groups) have been used by J-P. BOTH, Y. PENELIAU AND Y. K. LEE (Saclay)

IV - RESULTS AND ANALYSIS

A) Infinite media

In the following table, some neutronic characteristics of these configurations calculated in infinite media are presented. The reactions rates are integrated in the whole energy range and was calculated by APOLLO-1 in infinite media. We see that the most important nuclides are those considered in this table and that the two first isotopes of Pu are the most important especially in cases 1 to 3. The table also provides, as a spectrum indicator, the slowing-down current, defined as the number of neutrons which become thermal for one emitted neutron source. These configurations have a very important thermal component. A more detailed spectrum comparison is presented in figure 3. The average flux (the normalisation is 1 neutron emitted by fission) is calculated in infinite media with 172 groups. Notice that case # 4 has the most important thermal component, case # 5 has the most important contribution to the ^{240}Pu first resonance absorption and case # 1 the most important contribution to the ^{239}Pu first resonance absorption.

Case #	1	2	3	4	5	6
slowing-down current	0.91	0.95	0.97	0.98	0.92	0.95
Pu239 A	0.8763	0.8495	0.7591	0.4957	0.7145	0.6878
Pu239 P	1.6733	1.6636	1.5084	0.9983	1.3710	1.3487
Pu240 A	0.0640	0.0283	0.01883	0.0637	0.1542	0.1216
Pu241 A	0.0029	-	-	0.0453	0.0574	0.0582
Pu241 P	0.0065	-	-	0.0988	0.1258	0.1273
H$_2$0 A	0.0438	0.1051	0.1843	0.3477	0.0512	0.1071
N A	0.0120	0.0165	0.0370	0.0439	0.0144	0.0191
Others A	0.0010	0.0006	0.0008	0.0037	0.0083	0.0062

The infinite multiplication factors are presented in the following table. ↑ indicates the highest value and ↓ indicates the lowest one.

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Case #	1	2	3	4	5	6
slowing-down current	0.91	0.95	0.97	0.98	0.92	0.95
MONK-7 12630 g	1.6809	1.6687↑	1.5165↑	1.1097↑	1.5002	1.4809↑
WIMS-7 69 g	1.6812↑	1.6645	1.5079	1.0946	1.4991	1.4766
WIMS-7 172 g	1.6806	1.6642	1.5081	1.0951	1.4997	1.4771↓
TRIPOLI-4 pointwise	1.6819	1.6640	1.5091	1.0954	1.5016↑	1.4783
APOLLO-1 99 g	1.6805	1.6638	1.5085	1.0976	1.4997	1.4775
APOLLO-2 99 g	1.6793	1.6618	1.5082	1.0948	1.5004	1.4782
APOLLO-2 172 g	1.6778↓	1.6609↓	1.5077↓	1.0943↓	1.4989↓	1.4772

These results show good agreements between WIMS, TRIPOLI, APOLLO-1 and APOLLO-2. The maximum discrepancy is lower than 0.3 % for the six cases. However, the MONK results are in general higher than the others specially for case # 4 (more than 1 % difference). To analyse these results, let us consider the following definitions :

A_9^i : ^{239}Pu absorption rate in the group i

P_9^i : ^{239}Pu production rate in the group i

A_0^i : ^{240}Pu absorption rate in the group i

A_{others} : total absorption rate of other nuclides

P_{others} : total production rate of other nuclides

A : total absorption rate of all nuclides

P : total production rate of all nuclides

For an infinite media, we have :

$$k_\infty = \frac{P}{A}$$

where A = 1 and $k_\infty = P$

$$P_{others} = k_\infty - \sum_i P_9^i$$

$$A_{others} = 1 - \sum_i A_9^i - \sum_i A_0^i$$

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$$\frac{\delta k_{\infty}}{k_{\infty}} = \sum_i \left(\frac{\delta P_9^i}{P}\right) + \frac{\delta P_{others}}{P} - \sum_i \left(\frac{\delta A_9^i}{A}\right) - \sum_i \left(\frac{\delta A_0^i}{A}\right) - \frac{\delta A_{others}}{A}$$

For a finite media, we have the following definition (L is the proportion of leakage) :

$$k_{eff} = \frac{P}{A+L}$$

where $A+L=1$ and $k_{eff} = P$

$$P_{others} = k_{eff} - \sum_i P_9^i$$

$$A_{others} = 1 - L - \sum_i A_9^i - \sum_i A_0^i$$

$$\frac{\delta k_{eff}}{k_{eff}} = \sum_i \left(\frac{\delta P_9^i}{P}\right) + \frac{\delta P_{others}}{P} - \sum_i \left(\frac{\delta A_9^i}{A+L}\right) - \sum_i \left(\frac{\delta A_0^i}{A+L}\right) - \frac{\delta A_{others}}{A+L} - \frac{\delta L}{A+L}$$

The following table presents a 2 groups discrepancy analysis between APOLLO-1 and MONK

where $\dfrac{\delta X}{X}$ is calculated as $\dfrac{X(APOLLO-1) - X(MONK)}{X(MONK)}$ and is given in pcm (10^{-5})

Case #	1	2	3	4	5	6
$\dfrac{\delta k_{\infty}}{k_{\infty}}$	-24	-292	-526	-1091	-34	-230
$-\dfrac{\delta A_9^1}{A}$	-50	-68	-137	45	18	-18
$-\dfrac{\delta A_9^2}{A}$	107	302	72	523	-57	208
$\dfrac{\delta P_9^1}{P}$	16	131	287	-29	16	-16
$\dfrac{\delta P_9^2}{P}$	-98	-424	-135	-1217	-61	-444
$\dfrac{\delta P_{others}}{P}$	-12	-3	-9	-14	-86	-38
$-\dfrac{\delta A_0^1}{A}$	-27	24	126	79	-9	-3
$-\dfrac{\delta A_0^2}{A}$	-17	-254	-51	-633	134	-149
$-\dfrac{\delta A_{others}}{A}$	57	1	-678	155	11	230

9

Also, the following table presents a 2 groups discrepancy analysis between APOLLO-2 and

MONK where $\dfrac{\delta X}{X}$ is calculated as $\dfrac{X(APOLLO-1) - X(MONK)}{X(MONK)}$ and is given in pcm (10^{-5}).

Case #	1	2	3	4	5	6
$\dfrac{\delta k_\infty}{k_\infty}$	-184	-467	-582	-1390	-88	-253
$-\dfrac{\delta A_9^1}{A}$	-55	-70	-139	45	11	-20
$-\dfrac{\delta A_9^2}{A}$	244	396	71	655	-19	144
$\dfrac{\delta P_9^1}{P}$	28	136	290	-28	29	-11
$\dfrac{\delta P_9^2}{P}$	-292	-617	-198	-1483	30	-446
$\dfrac{\delta P_{others}}{P}$	79	14	-674	122	-86	204
$-\dfrac{\delta A_0^1}{A}$	-13	-3	-9	-14	-89	-39
$-\dfrac{\delta A_0^2}{A}$	-122	-104	75	-132	0	14
$-\dfrac{\delta A_{others}}{A}$	-4	-218	2	-554	97	-99

These data clearly indicate that the thermal range (and specially [239]Pu and [240]Pu sections) is responsible of a large part of the difference. A more detailed picture could be obtained with the 15 groups discrepancy analysis using the same definitions. Figures 4 to 9 give for each case the contribution of the multigroup reaction rate (absorption and production for [239]Pu and absorption for [240]Pu) to the k-infinity discrepancy. The data shown are :

$-\dfrac{\delta A_9^i}{A}, \dfrac{\delta P_9^i}{P}$ and $-\dfrac{\delta A_0^i}{A}$ as a function of the energy group i (note that the negative sign has been

taken into account for absorption reaction rates). If one considers only discrepancies greater than 300 pcm (because of the statistical uncertainty of the Monte-Carlo) only groups # 14 and 15 (energy below 0.53 eV) have a significant contributions to the discrepancies. In particular, we notice that the error in group 14 has systematically the opposite sign of the error in group

15 which certainly leads to compensations. Also, we notice that $\dfrac{\delta A_9^i}{A}$ and $\dfrac{\delta P_9^i}{P}$ have the same

sign, which indicates that problems do exist for both capture and fission data.

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Comparisons between APOLLO, MONK and WIMS cross sections have been performed (data not shown). Preliminary results suggest that the approximate weighting function used in NJOY to produce the MONK application library cross-sections at low energies (below 0.3 eV) is inadequate for the number of groups employed. Re-generation of the MONK library using a more appropriate weighting function and possibly more groups is now proposed. This paper will be updated to report revised results when they become available.

B) Bare spheres

The results of bare spheres configurations are reported in the following table together with the spectrum indicator q. All codes used are Monte-Carlo ones and the statistical uncertainty on the keff is about 0.001 (1 σ) for each case.

Case #	1	2	3	4	5	6
slowing-down current[2]	0.56	0.61	0.69	0.91	0.63	0.68
MONK keff	1.0066↑	1.0071↑	1.0105↑	1.0123↑	1.0076↑	1.0103↑
MONK Leakage	0.3993	0.3946	0.3328	0.0872	0.3258	0.3155
TRIPOLI-4 keff	1.0004	1.0003	1.0016	1.0019	1.0031	1.0036
TRIPOLI-4 Leakage						
A2 + TRIMARAN-2 keff	1.0006	0.9966↓	1.0001↓	1.0004	1.0036	1.0027
A2 + TRIMARAN-2 Leakage	0.4025	0.3979	0.3357	0.0872	0.3290	0.3195
A1 + MORET keff	0.9990↓	0.9979	1.0000	0.9981↓	1.0011↓	1.0016↓
A1 + MORET Leakage	0.4030	0.3990	0.3357	0.0900	0.3297	0.3195

As for the infinite media, the results show very good agreements between TRIPOLI-4, APOLLO-2 + TRIMARAN and APOLLO-1 + MORET. The MONK results are the highest ones (about 0.7 % to 1.2 % greater than the others). The conclusions reported in the previous

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[2] calculated by MORET-3, in the real geometry, as the number of neutrons which become thermal (energy cut-off equal to 2.76 eV) for one emitted fission neutron

section should explain at least one part of the discrepancies. In fact, case # 4 and 3 where the highest discrepancy was observed for infinite media give also the maximum difference for bare spheres. Nevertheless, leakage calculation could have a contribution to the discrepancies since the difference for the other cases is more important in bare spheres configurations than in infinite media. The following table gives some comparisons between MONK and TRIPOLI-4 where the 2 groups structure has been considered. The conclusions are similar to those found in the previous section except that the leakage component adds additional differences. Further investigations are needed and more detailed comparisons will be undertaken as soon as the revised MONK results will be available.

Case #	1	2	3	4	5	6
$\dfrac{\delta k_{eff}}{k_{eff}}$	-616	-675	-881	-1027	-447	-663
$-\dfrac{\delta A_9^1}{A+L}$	75	5	10	0	45	-10
$-\dfrac{\delta A_9^2}{A+L}$	220	393	270	510	170	210
$\dfrac{\delta P_9^1}{P}$	-169	988	-14	-20	6	20
$\dfrac{\delta P_9^2}{P}$	-616	-1018	-688	-1225	-349	-406
$\dfrac{\delta P_{others}}{P}$	169	-646	-179	217	-103	-277
$-\dfrac{\delta A_0^1}{A+L}$	-17	-10	0	-20	-65	-30
$-\dfrac{\delta A_0^2}{A+L}$	56	47	61	110	38	40
$-\dfrac{\delta A_{others}}{A+L}$	36	5	-51	-230	51	190
$-\dfrac{\delta F}{A+L}$	-370	-440	-290	-280	-240	-400

In this table, $\dfrac{\delta X}{X}$ is calculated as $\dfrac{X(TRIPOLI-4) - X(MONK)}{X(MONK)}$

C) Experimental configurations

The results of the experimental benchmarks as described in [7] are reported in the following table. Up to now only APOLLO-1 + MORET-3 and MONK-7 results are available. The statistical uncertainty associated with the keff values is about 0.001 (1 σ) for each case.

Case #	1	2	3	4	5	6
MONK-7 12000 g	1.0148↑	1.0133↑	1.0143↑	1.0209↑	1.0152↑	1.0140↑
A1 + MORET 99 g	1.0031	0.9988	0.9998	1.0143	1.0082	1.0086
$\ln(\dfrac{k_{MONK}}{k_{MORET}})$ pcm	1160	1441	1440	649	692	534

The discrepancy between the two set of results ranges from 0.5 % to 1.4 % which is important. It is interesting to notice that the discrepancies obtained for the experimental configuration are different from those obtained in the bare sphere cases which are again different from those obtained in infinite media (Figure 10). In particular, configurations where the differences was very low in the infinite media have important ones in the reflected spheres. This suggests that more investigations are needed to explain the differences obtained for the benchmark configurations. The difference in the leakage and reflection calculation (anisotropy treatment for instance) could be a source of discrepancies.

V- CONCLUSIONS

This paper describes calculation results for nitrate plutonium experiments and investigates a progressive inter-code comparisons for a selection of configurations. In particular, we show important discrepancies between MONK-7 results and the others codes for well thermalised solutions. The discrepancies were analysed and a potential source of error was proposed and confirmed by preliminary cross-section comparisons. It seems that the weighting function used in NJOY to produce the MONK cross-section library is not appropriate for very thermal energies (below 0.3 eV). Work is in progress to re-generate new application libraries and to provide revised results. This exercise is still open and new contributions are welcomed.

ACKNOWLEDGEMENTS

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[7] : International Handbook of Evaluated Criticality Safety Benchmark Experiment, NEA/NSC/DOC(95)03/I

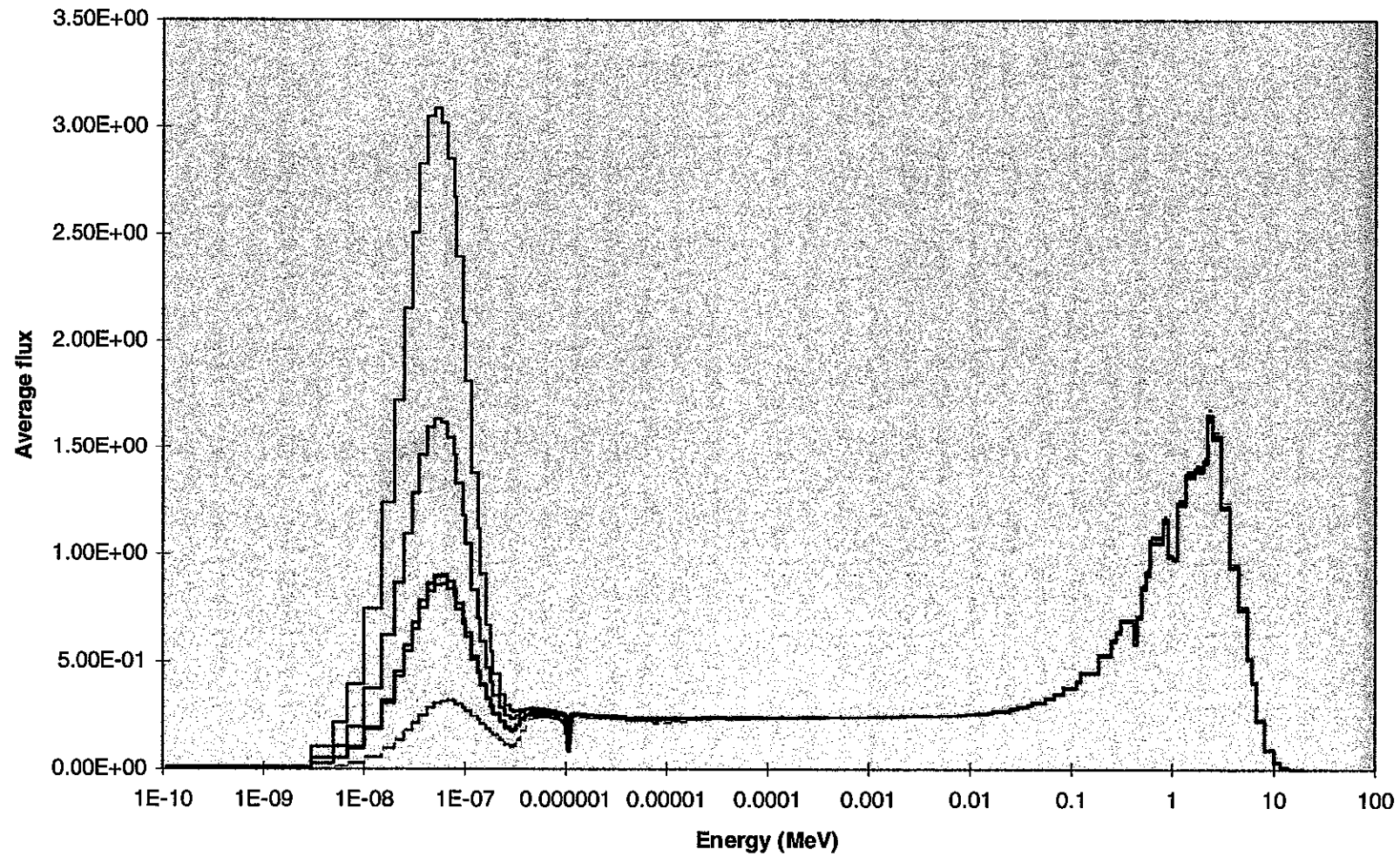# Figure 1 : comparison between MONK-7 and APOLLO-1 + MORET-3 results

PNL experiments
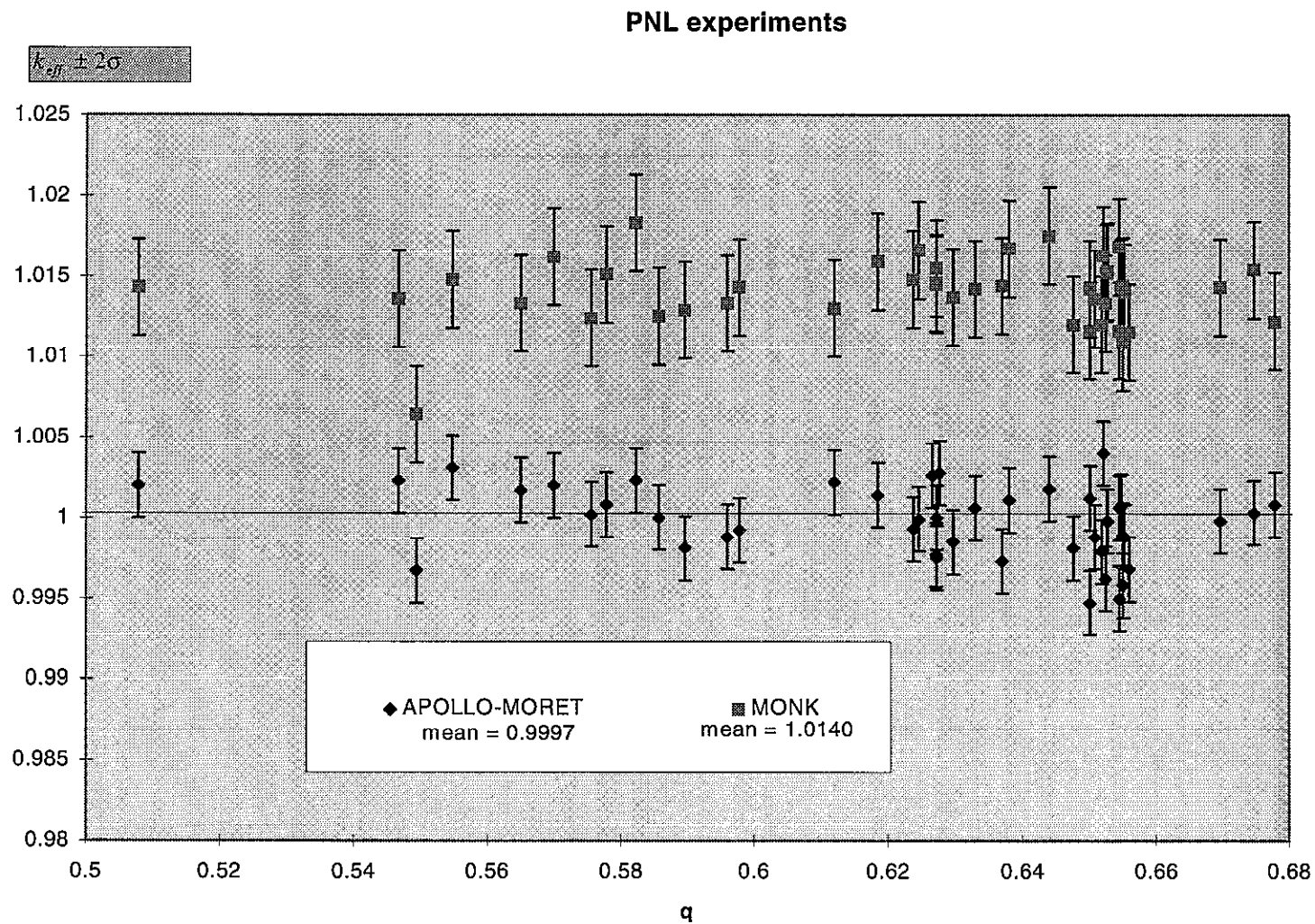
Figure 2 : comparison between MONK-7 and APOLLO-1 + MORET-3 results

VALDUC experiments
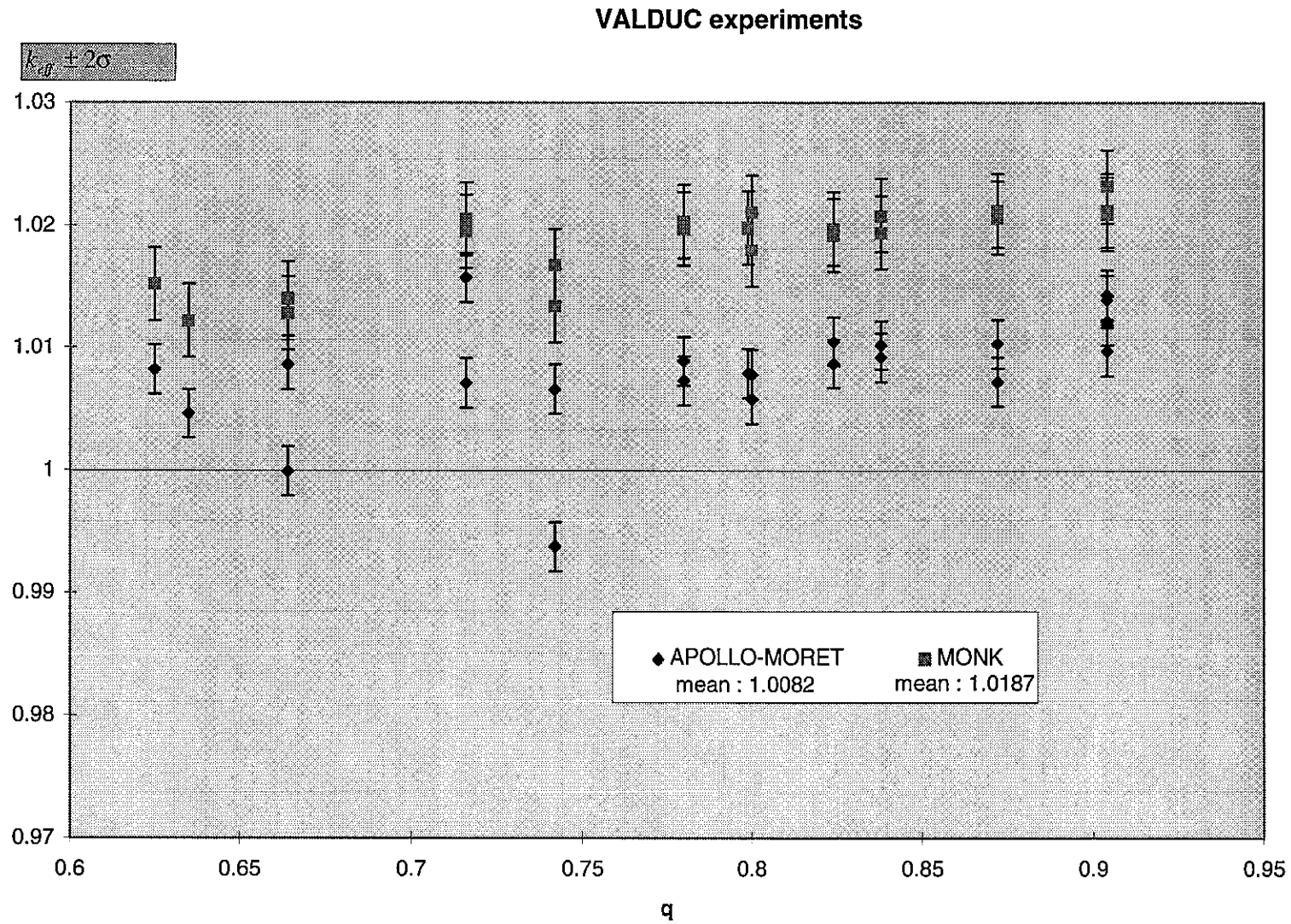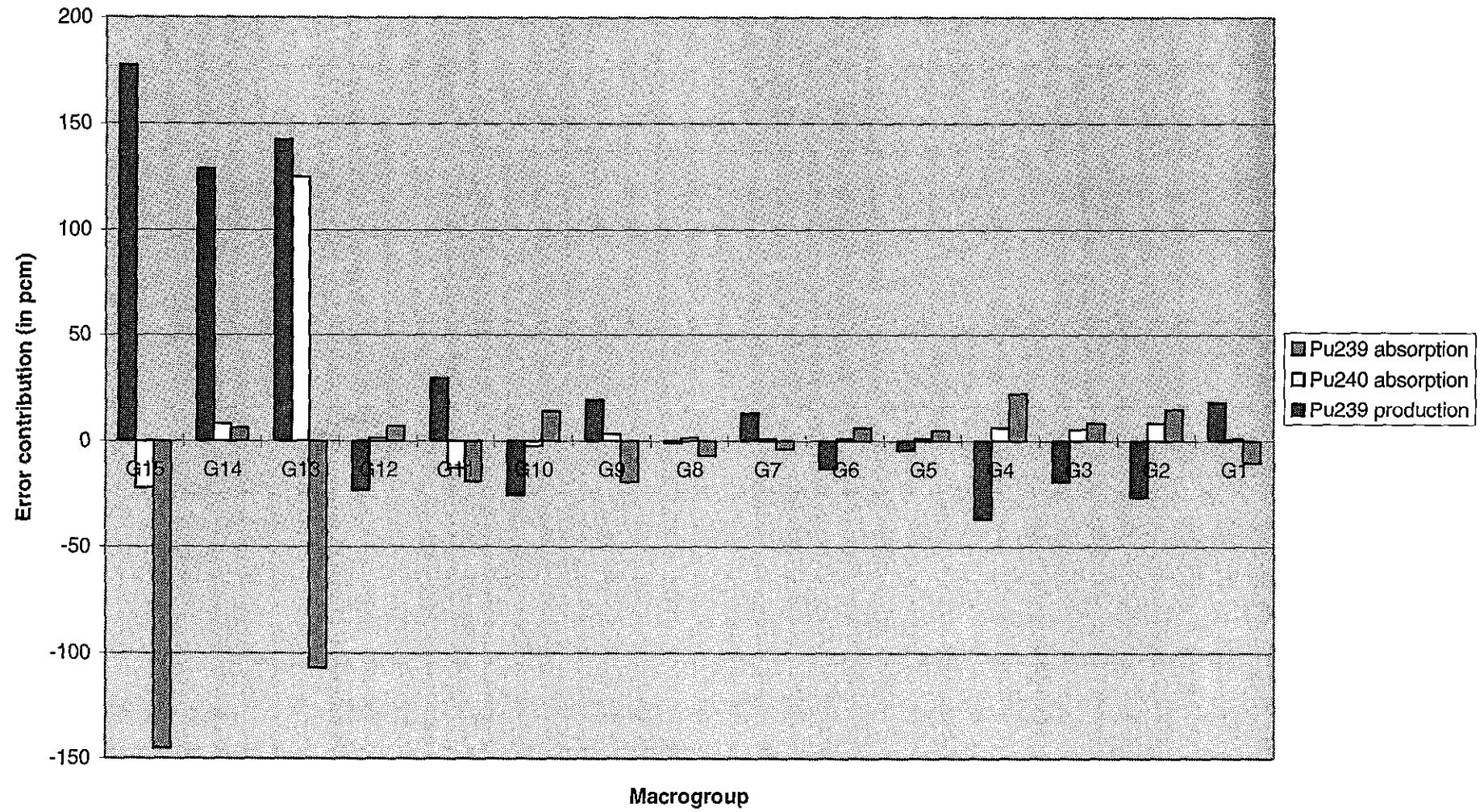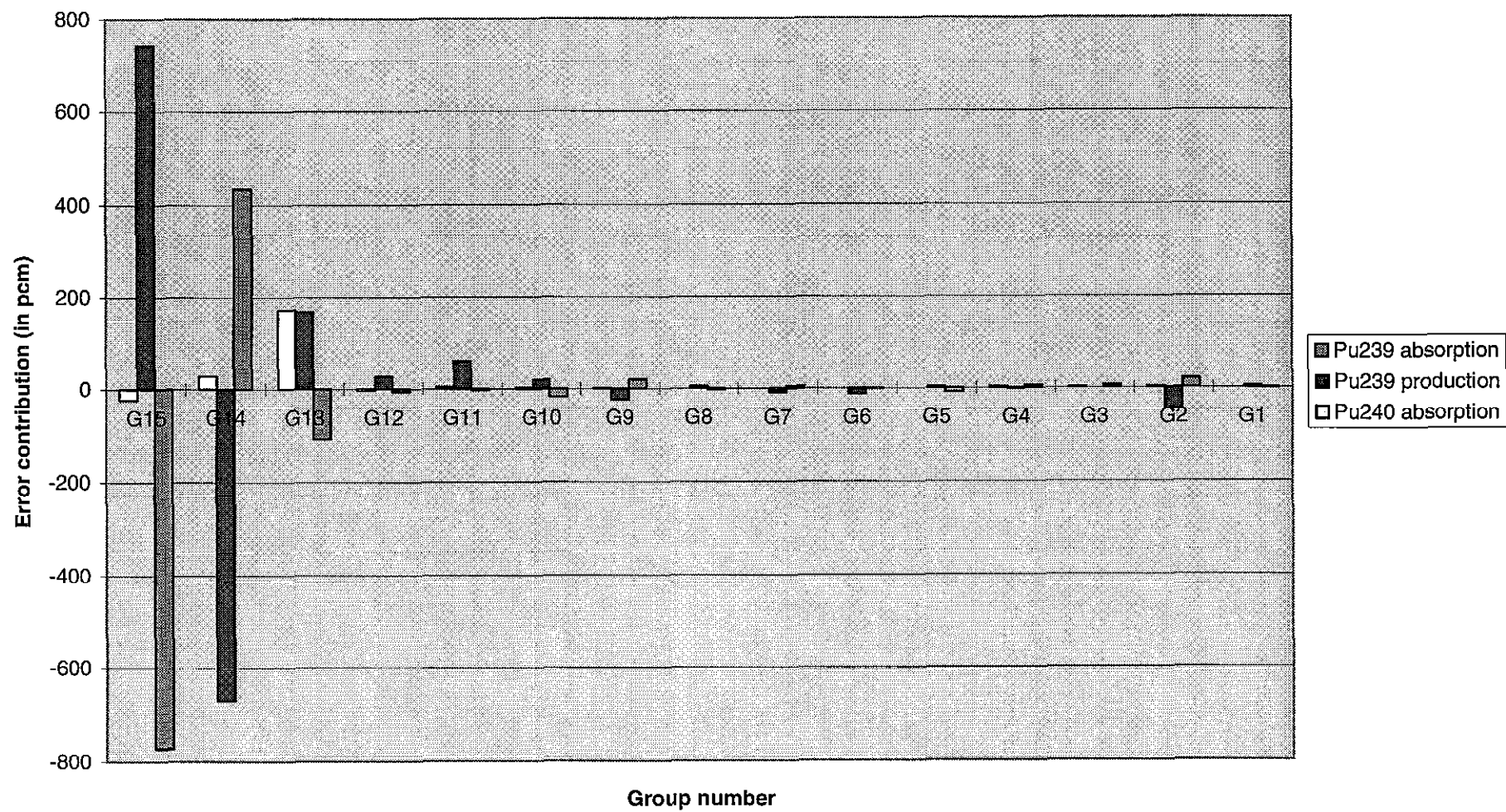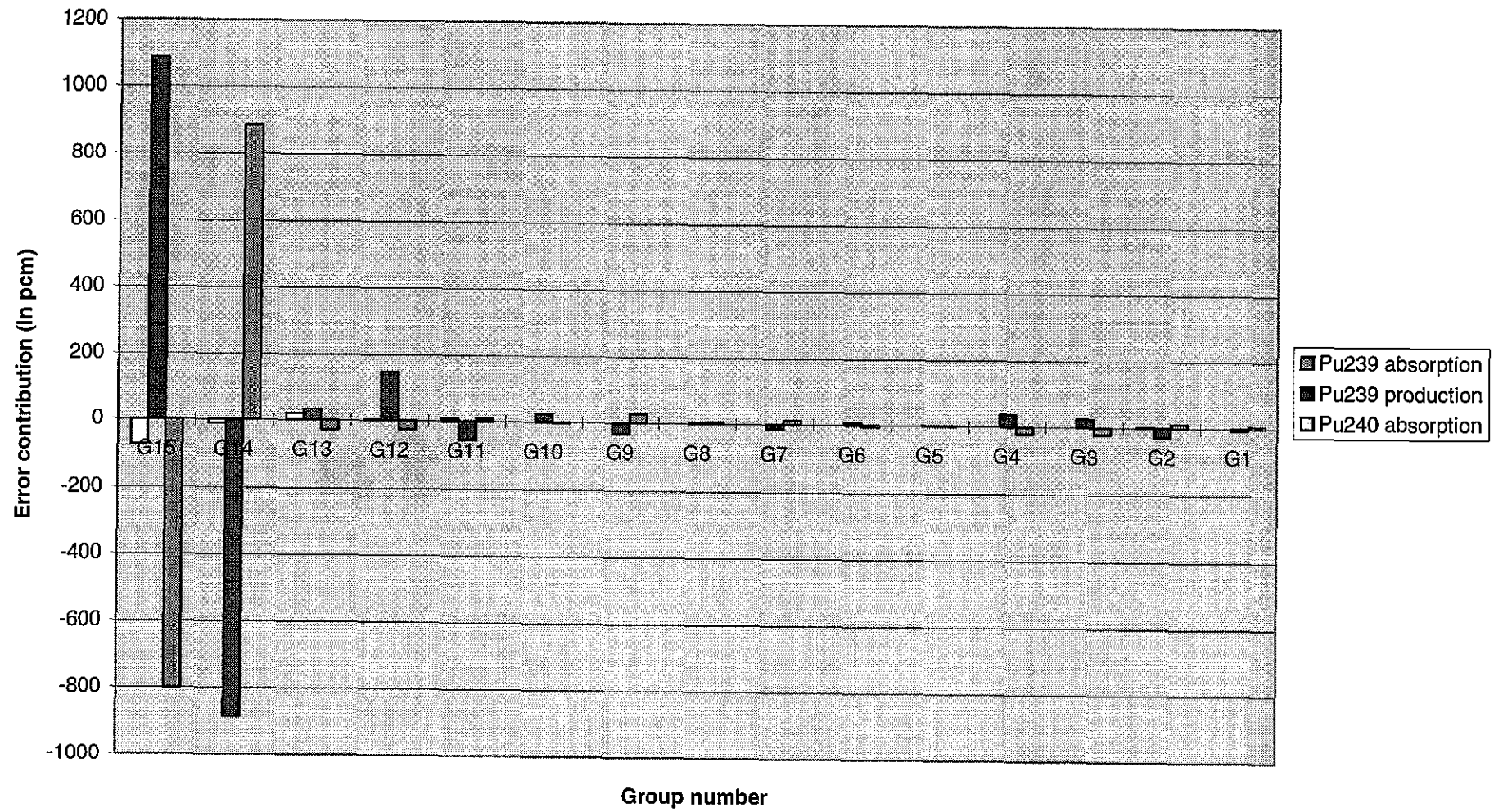
Figure 3 : Neutron spectrum in the infinite configuration for the selected nitrate plutonium solutions
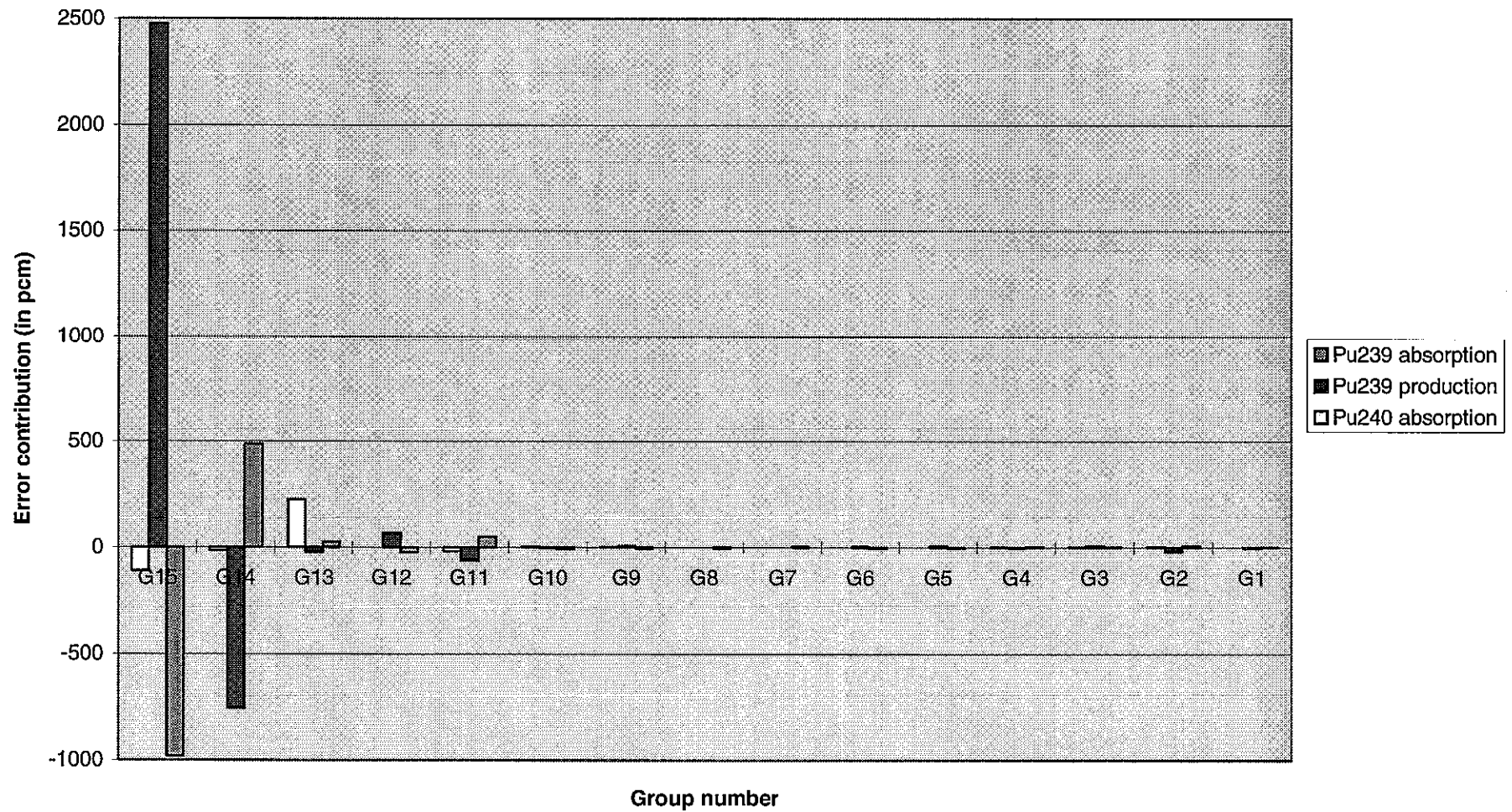
Neutron spectra

Figures 4

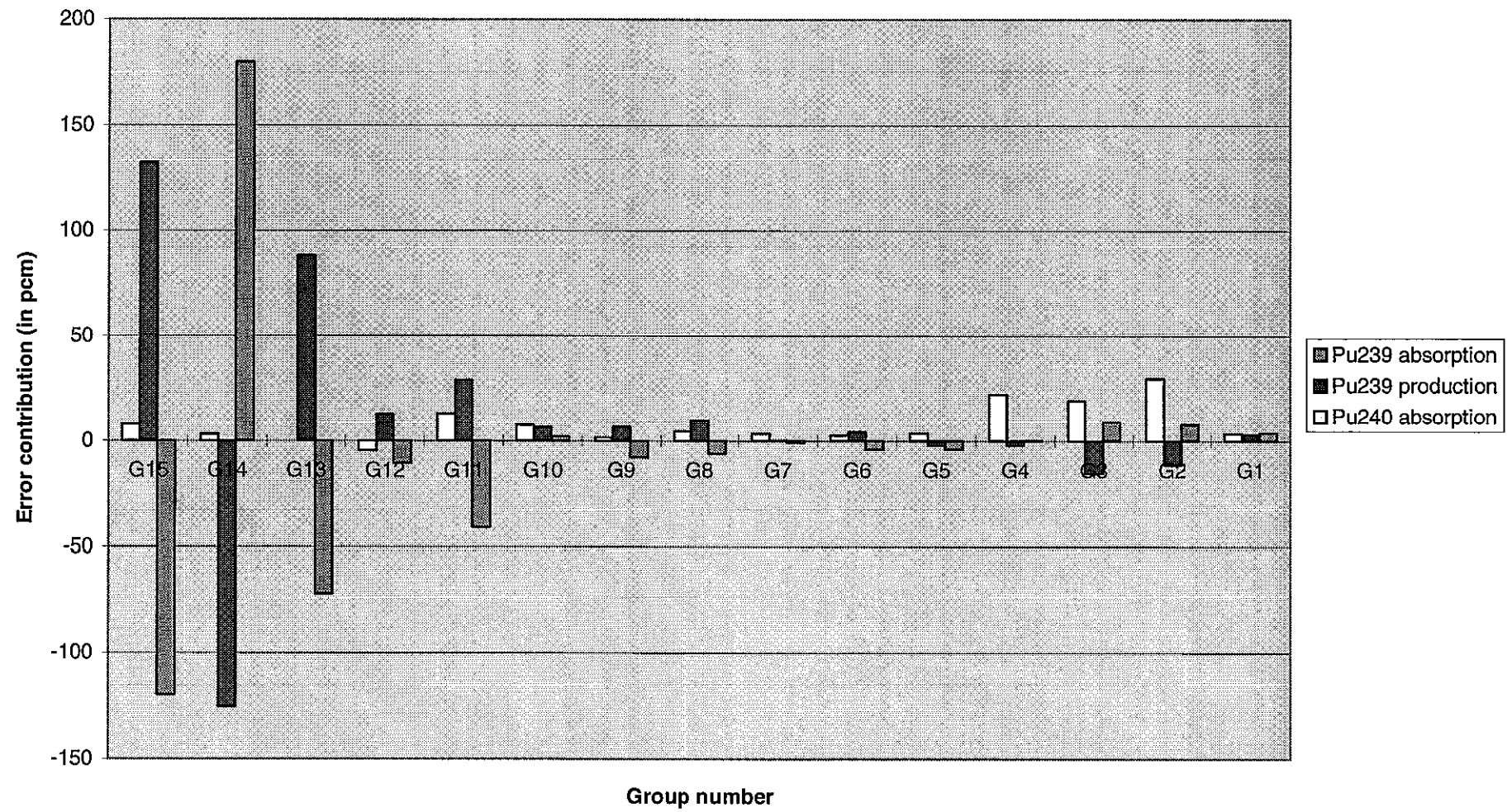## Case # 1 : multigroup contribution to the keff discrepancy

Figures 5

Case # 2 : multigroup contribution to the keff discrepancy

Figures 6

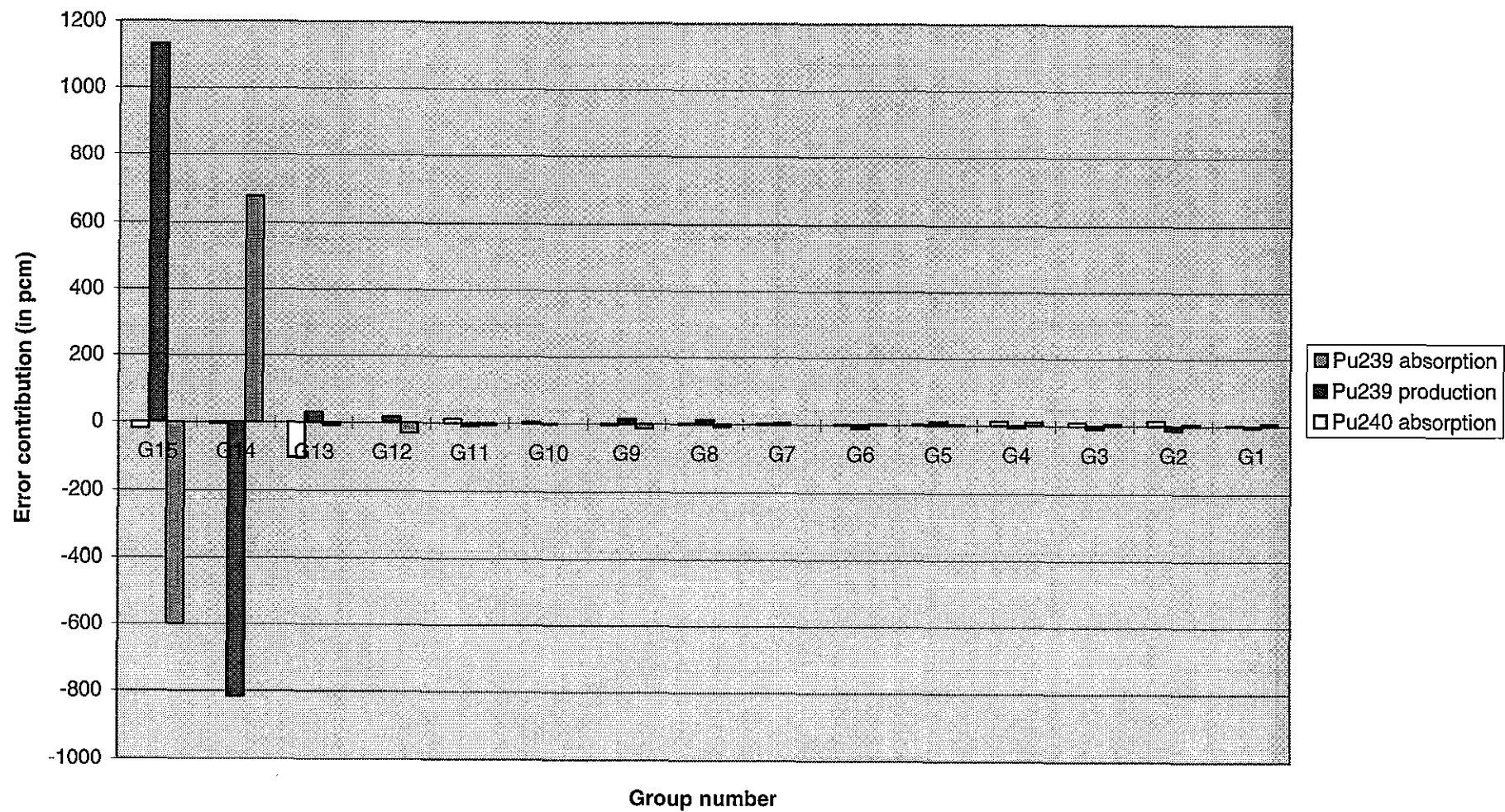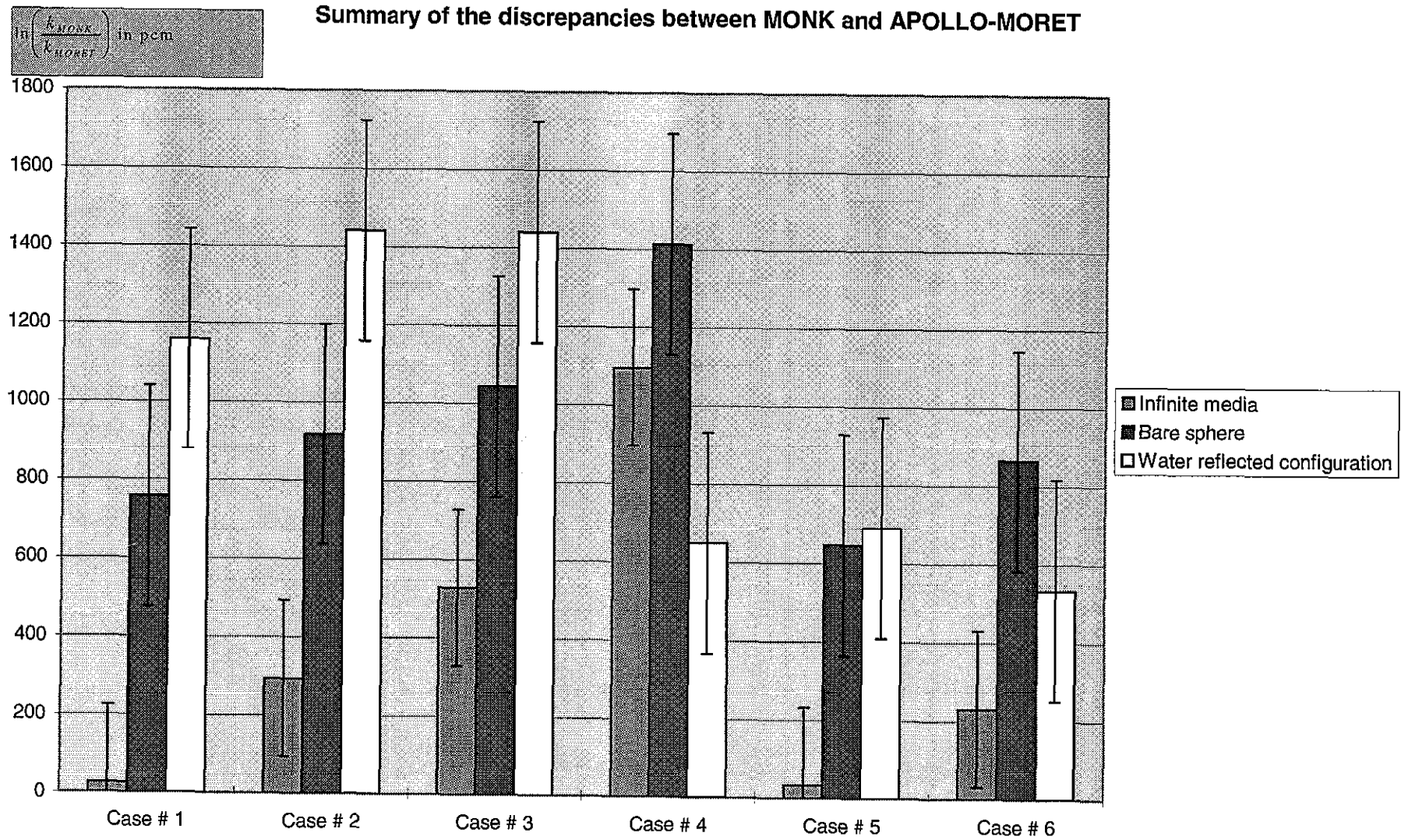### Case # 3 : multigroup contribution to the keff discrepancy

Figures 7

Case # 4 : multigroup contribution to the keff discrepancy

Figures 8

Case # 5 : multigroup contribution to the keff discrepancy

Figures 9

Case # 6 : multigroup contribution to the keff discrepancy

Figure 10

Summary of the discrepancies between MONK and APOLLO-MORET

$\ln\left(\frac{k_{MONK}}{k_{MORET}}\right)$ in pcm

Legend:
- Infinite media
- Bare sphere
- Water reflected configuration

24