Consistent criticality and radiation studies of Swiss spent nuclear fuel: the CS_2M approach

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Abstract

In this paper, a new method is proposed to systematically calculate at the same time canister loading curves and radiation sources, based on the inventory information from in-core fuel management system. As a demonstration, the isotopic contents of the assemblies come from a Swiss PWR, considering more than 6000 cases from 34 reactor cycles. The CS_2M approach consists in combining four codes: CASMO and SIMULATE to extract the assembly characteristics (based on validated models), the SNF code for source emission and MCNP for criticality calculations for specific canister loadings. The cases considered cover enrichments from 1.9 to 5.0 % for the UO_2 assemblies and 4.7 % for the MOX, with assembly burnup values from 7 to 74 MWd/kgU. Because such a study is based on the individual fuel assembly history, it opens the possibility to optimize canister loadings from the point-of-view of criticality, decay heat and emission sources.

1 Introduction

The future of the spent nuclear fuel (SNF) is the focus of a large number of recent studies regarding intermediate and long-term storage. Countries like Finland, Sweden, Germany or Switzerland have opted for final disposal in deep geological repositories [1–4], and if some questions are not yet unequivocally answered, many advances have been made (*e.g.* on confinement, isolation, radiation or heat) to assure the safety of a repository for tens to hundreds of thousands of years.

For a safe storage of the SNF, three quantities are of prime importance considering the fuel itself: its long-lived radioactivity content (actinides and fission products), heat generation (also called decay heat) and radiation level (or source term). The high radiation level implies the use of shielding and remote handling systems, the heat generation limits the stored amount in a

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given volume of rock, and the radioactivity content means that the criticality safety of a repository can be an issue for the duration of the repository. In the preliminary studies regarding the storage of the SNF in licensing application, these quantities can be calculated and their precise knowledge is important to obtain a high degree of confidence in safety analysis. Some of these quantities can also be measured (decay heat [5], radiation level [6]), but systematic measurements on the thousands of SNF cannot be performed in practice (in Switzerland alone, about 12 000 fuel assemblies are expected to be finally discharged [7]). Therefore the dependence on calculated quantities is capital. These three quantities are linked together as the isotopic content of a SNF is responsible for the produced decay heat, the emitted radiations and the possibility of criticality events: for instance, the correct knowledge of the amounts of ^{235}U, ^{239}Pu, ^{244}Cm, and other isotopes is at the start of the correct assessment of the emitted number of neutrons, gammas, and of the criticality occurrence.

For practical reasons, the studies of the criticality-safety (for instance using burn-up credit methods) and radiation shielding or decay heat can be performed applying different assumptions on the fuel content. One reason is that the fuel content of a specific assembly can be derived from its irradiation history, and this irradiation history is often not precisely known by the institutions responsible for such studies. As a consequence, one of the solutions is to choose for representative studies (see for instance Ref. [8]) which is supposed to cover all different cases of assembly histories. The assumptions at the basis of such representative studies can differ (*e.g.* bounding depletion conditions, axial and horizontal burnup profiles [9], lattice heterogeneities or neutron spectrum representation), leading to possible differences in the isotope inventory for criticality and radiation studies (see examples in Refs. [10,11]).

Such situation can finally result in analyses which are softly linked between criticality-safety and shielding (*e.g.* Refs. [12–15]). There are nonetheless advantages of performing specific assembly analysis for criticality-safety, shielding and radioprotection using a unique source of data: (1) the consistency between these different quantities is assured, (2) in the case of using an in-core fuel management system (ICFM), all cases can be covered, avoiding the difficulty of representative studies, and (3) there is no need to consider assumptions for pinwise isotopic distributions, or assembly-wise burnup profiles.

In this work, we are proposing to calculate the isotopic content, decay heat, neutron & gamma sources and the neutron multiplication factor at once, taking into account the individual history of all fuel assemblies from an ICFM system. Four specific codes will be involved for this study: CASMO and SIMULATE (with validated reactor core models), SNF and MCNP and in the following, the method will be called CS_2M (based on the initials of each code). The example of a real Swiss Pressurized Water Reactor (PWR) will be considered over a duration of 34 cycles. To cover a large range of possibilities in terms of assemblies, enrichment and burn-up, each assembly at the end of a cycle (being discharged or reloaded in the next cycle) will be considered

in this study. This represents a total of 5378 UO_2 assemblies and 640 MOX assemblies, as presented in Fig. 1.

The results will be presented in terms of loading curves for a specific canister,

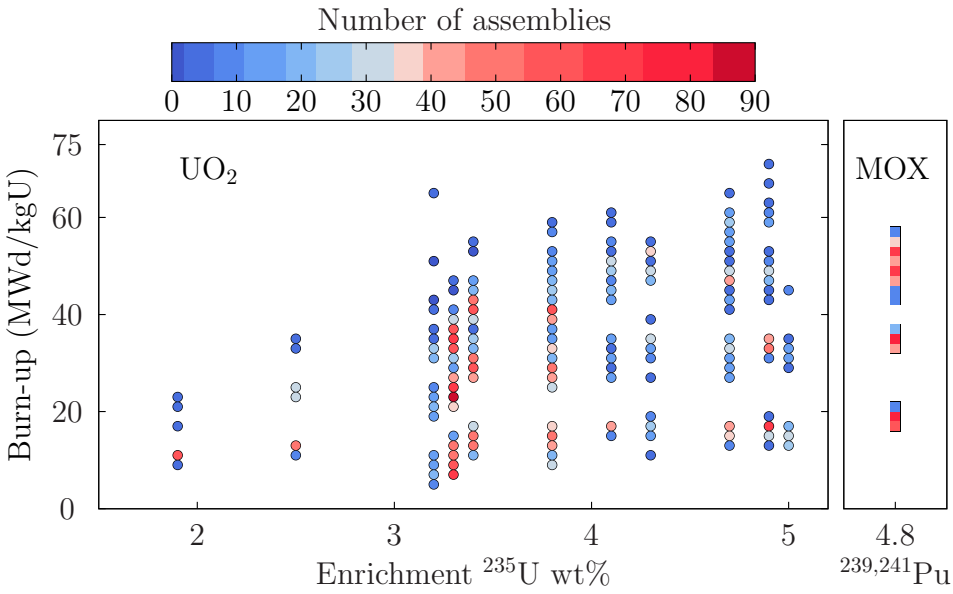

Fig. 1. Total number of fuel assemblies considered in this work from a specific Swiss PWR power plant, over 34 cycles (5378 UO_2 and 640 MOX assemblies). All assemblies are considered at the end of each cycle, being discharged or not.

together with the corresponding decay heat, and neutron/gamma source terms for individual assemblies. For this specific demonstration study, no uncertainty propagation will be considered and the burnup credit will be performed with the "*actinide only*" approach.

2 Method

The method proposed in this work is summarized in Fig. 2, allowing to consistently calculate criticality quantities (being the neutron multiplication factor k_{eff}), emission source (neutron and gamma) and decay heat. Such approach can provide these quantities considering the individual history of each irradiated fuel assembly and is articulated around the four following steps:

(1) each assembly, core and cycle is represented by validated models and data,
(2) these CASMO/SIMULATE models are applied to reconstruct the lattice data file with isotope concentrations and power history data file,
(3) the *SNF* code is using the previous information to provide individual isotopic concentrations for each pin and segment, together with the assembly neutron/gamma source and decay heat quantities, for different

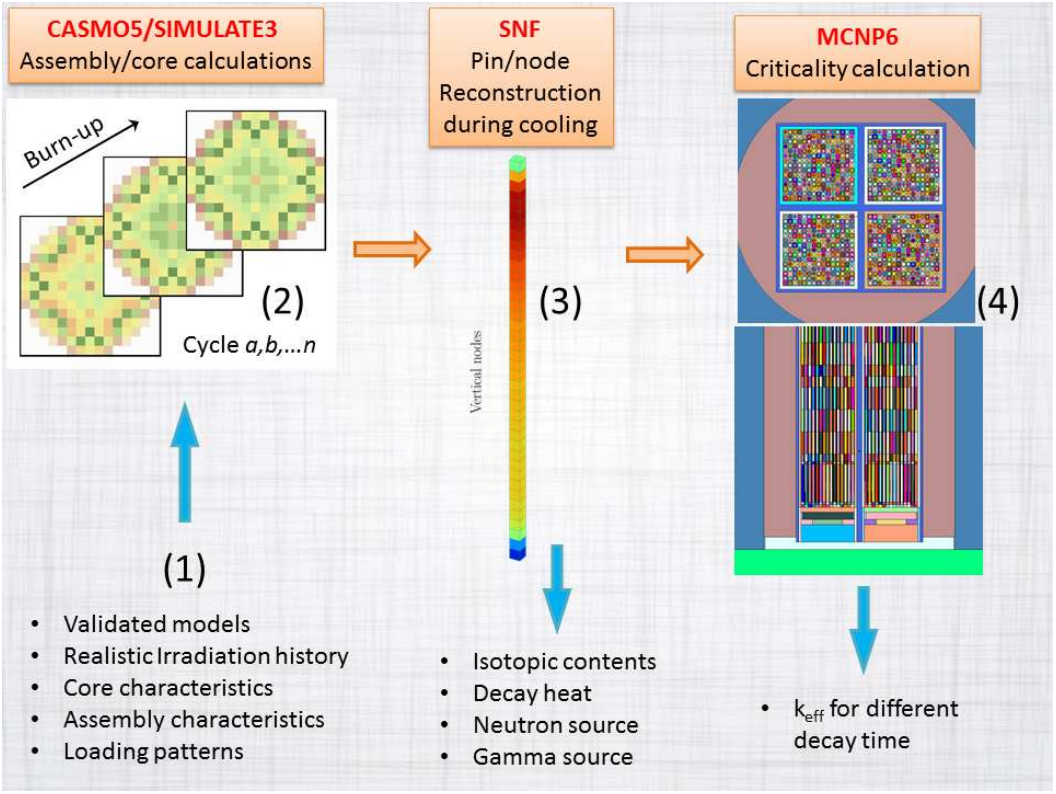

Fig. 2. General overview of the consistent calculation scheme based on CS_2M method (with CASMO/SIMULATE/SNF/MCNP models). The four steps are detailed in the text.

cooling times,

(4) the isotopic concentrations for each cooling time is passed to a MCNP model (for each pin and vertical segment) for a specific canister, allowing to calculate k_{eff}.

Details are given in the next sections.

2.1 Step 1: Validated models

The starting point of this calculation scheme is the large database of all reactor cycles for the Swiss nuclear power plants [16]. It is a PSI in-house code and model integrated system based on the Studsvik suite of codes and is referred to as the CMSYS database in the following. It includes predictive and validated models for the existing and next operating cycles for independent verifications of all Swiss reload licensing submittals in support of the national regulator ENSI. Some additional details can be found in Ref. [17].
The extraction of the suitable data from the CMSYS database is represented in Fig. 2 by the arrow with the index (1). In the context of the present work, the quantities of interest are the assembly characteristics (fuel, enrichment,

geometry), the cycles in which they are burned, their calculated burnup values at the end of each cycle, the core cycle characteristics (core power, boron concentration, cycle length, shutdown period...), and the fuel loading pattern.

2.2 Step 2: CASMO/SIMULATE calculations

The foreseen irradiation history, the core loading patterns, the assembly characteristics and other parameters are received from the plant operator in the context of the reload licensing verification. They are then used with CASMO-5 [18] and SIMULATE-3 [19]. After the cycle operation, the real irradiation histories are used in the CMSYS database and predictions are compared with in-core measurements (boron concentration and Traversing In-Core Gamma & Neutron Probes, or TIPS), allowing to obtain validated models and procedures. The present work is based on these validated models and is represented in Fig. 2 by the index (2).

2.3 Step 3: Addition of the SNF code

The use of the *SNF* code (denoted in italics to differentiate it from the "SNF" term for Spent Nuclear Fuel) allows to calculate different quantities for any assemblies included in our CMSYS database. It is using the irradiation history from the 3D reactor simulator (SIMULATE) and the isotopic library from the lattice data (CASMO). Both information are included in the CMSYS database.
Different isotopic concentrations for each pin of each assembly, as well as a function of the segment height can be calculated by *SNF*, at the time of discharge and for different cooling times. In the present study, a UO_2 assembly is made of 15×15 pins (225) with 20 control rod guide tubes. For the MOX assemblies, one additional empty channel (the central one) is considered. Typically, the number of vertical segment for each pin is 40, leading to a total of 8200 zones per assembly. The current *SNF* version in place is 1.6.4, which allows to extract all the important actinides, but is restricted in the case of fission products. Therefore the actinide-only burnup credit approach is considered here and the list of isotopes for the criticality-safety calculations is given in Table 1.

An example for a specific UO_2 assembly enriched at 4.9 % used for 5 successive cycles is presented in Fig. 3. This assembly was located between the center and the border of the core for these cycles. One can see the distributions of the burnup values at the end of cycle as a function of vertical nodes for each

Table 1
List of the considered isotopes, extracted from *SNF* and used in the criticality-safety calculations.

^{16}O	^{234}U	^{235}U	^{236}U	^{238}U	^{237}Np
^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
^{242m}Am	^{243}Am	^{242}Cm	^{244}Cm		

reactor cycle. Local minima are due to spacers. It can be noticed that the distributions are not flat between the nodes 5 and 30, but present a weak slope due to the asymmetrical vertical power distribution in the core. In addition to the burnup distributions, the inserts in Fig. 3 present the relative ^{235}U concentrations at the pin level for the assembly at the end of the 5 cycles. One can see that the pin distribution is becoming more heterogeneous as the burnup of the assembly increases.
In parallel to the isotopic contents, *SNF* provides other quantities as a function of cooling time, such as the total activity, decay heat, neutron and gamma sources. They will be presented for a specific assembly in section 3. The extraction of the suitable data from the CMSYS database is represented in Fig. 2 by the arrow with the index (3).

2.4 Step 4: Canister model with MCNP

Once the *SNF* code is used for all the pins and vertical segments in a given assembly, the isotopic contents for the isotopes in each volume of fuel material is passed to MCNP6 [20]. Given the large number of volumes with specific isotopic information for a single assembly, such a step is automatized and performed by the in-house tool called COMPLINK [21]. The different zones in the MCNP model are represented in Fig. 2 right by different colours.
The model for the canister considered in this work was used in Refs. [22,23]. It corresponds to a preliminary Swiss disposal canister design: the cask is a carbon steel cylinder, almost 5 meters height, where 4 PWR fuel assemblies in 4 separate carbon steel boxes can be inserted and welded [24]. No strong neutron absorbers are included in the design.
In the following, the same assembly will be loaded in the four available spots for simplicity.

3 Application to a Swiss reactor

In this paper, a demonstration of the method is presented. To illustrate the capabilities of this approach, as mentioned, a specific Swiss PWR core is considered, with UO_2 and MOX fuel assemblies. This study spans over 34 reactor

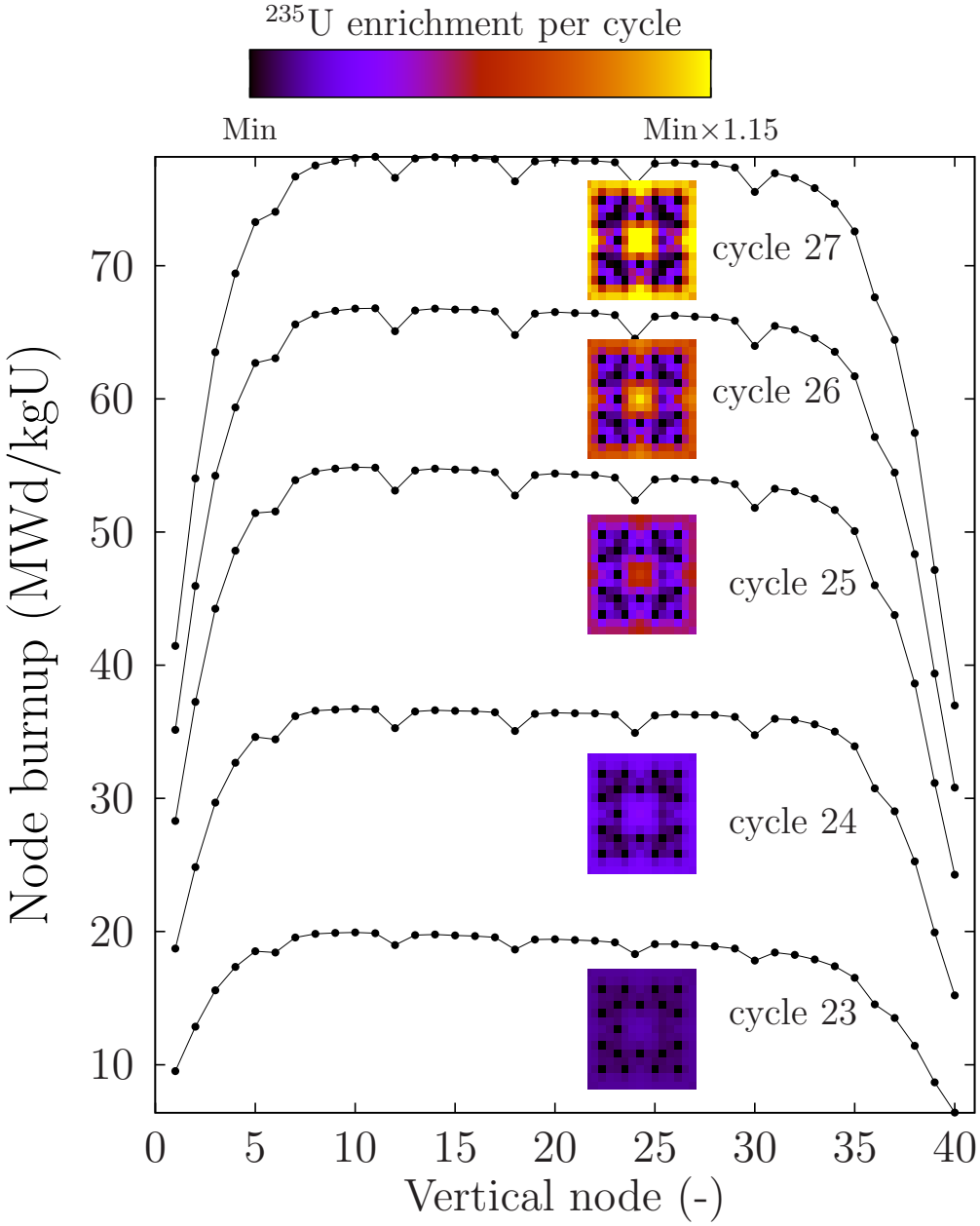

Fig. 3. Example of burnup distributions at the end of cycle as a function of the vertical nodes for a specific UO_2 assembly used for 5 different cycles. The inserts indicate the relative ^{235}U concentration for the node 22.

cycles, representing about 35 years of operation. In total, approximately 1790 assemblies were used in multiple cycles, being more than 6000 assembly-cycles (5378 UO_2 and 640 MOX). To cover a large range of assembly burnup values, the present criticality and radiation source calculations are performed for each assembly at the end of each cycle, being discharged for final disposal or not (see Fig. 1). As observed, the UO_2 enrichment is spread from 1.9 % to 5 % and the MOX enrichment (fissile Pu content) is 4.8 %, see Table 2. As presented, the same isotopic content from Table 1 is considered for the MCNP calcu-

Table 2
Characteristics of the spend nuclear fuels considered in this work. The MOX enrichment is given for the fissile Pu isotopes. The last column presents the assembly average burnup values.

Fuel	Enrichment %	Cycles	Assembly-cycles	Burnup MWd/kgU
UO_2	1.9-5.0	34	5378	7-74
MOX	4.8	15	640	18-58

lations (criticality) and the radiation source calculations (neutron & gamma emission, activity and decay heat). As an illustration, examples for two specific assemblies (one UO_2 and one MOX) are presented in Fig. 4. As mentioned for the criticality calculation, it is assumed that the canister is loaded with four of the same assemblies. The calculations of criticality and radiation sources are also presented for different cooling times. These assemblies have a similar enrichment for fissile isotopes (4.7 and 4.8 %) and were used in multiple cycles. It can be observed that as expected, the criticality values of the loaded canister depend on the burnup of the assemblies. In the UO_2 example, the lowest burnup value leads to k_{eff} clearly higher than 1, for all decay time. When considering a single k_{eff} curve, it can be observed that the highest k_{eff} value is obtained either at very short cooling time, or between 10^4 and 10^5 years. This is also characteristics of the "actinide only" approach. Same remark can be applied to the MOX assembly, although the MOX assembly is less reactive in terms of criticality than the UO_2 assembly.
Regarding the radiation sources, the gamma emission and decay heat are very similar for both assemblies and at different burnup values. On the other hand, the neutron emission is changing as a function of burnup values and is higher in the MOX case, due to the higher content of minor actinides.

3.1 Loading curves

A loading curve is defining the limit between an acceptable canister loading (with a k_{eff} value lower than a given limit), and a loading leading to a k_{eff} value higher than the limit. In the following, the limit if fixed at $k_{eff} = 0.95$. To calculate the loading curve, the MCNP model as described in section 2.4 is used to calculate k_{eff}, with four similar assemblies. All assemblies as presented in section 3 are used for the MCNP calculations, being equivalent to about 6000 calculations at a specific cooling time. Additionally, the k_{eff} values need to be obtained at many cooling times, as presented in Fig. 4. As observed in this figure, the k_{eff} at discharge is the highest, or almost the highest compared to longer cooling times. For the purpose of the demonstration, the calculations at different cooling times are performed for a limited number of assemblies.

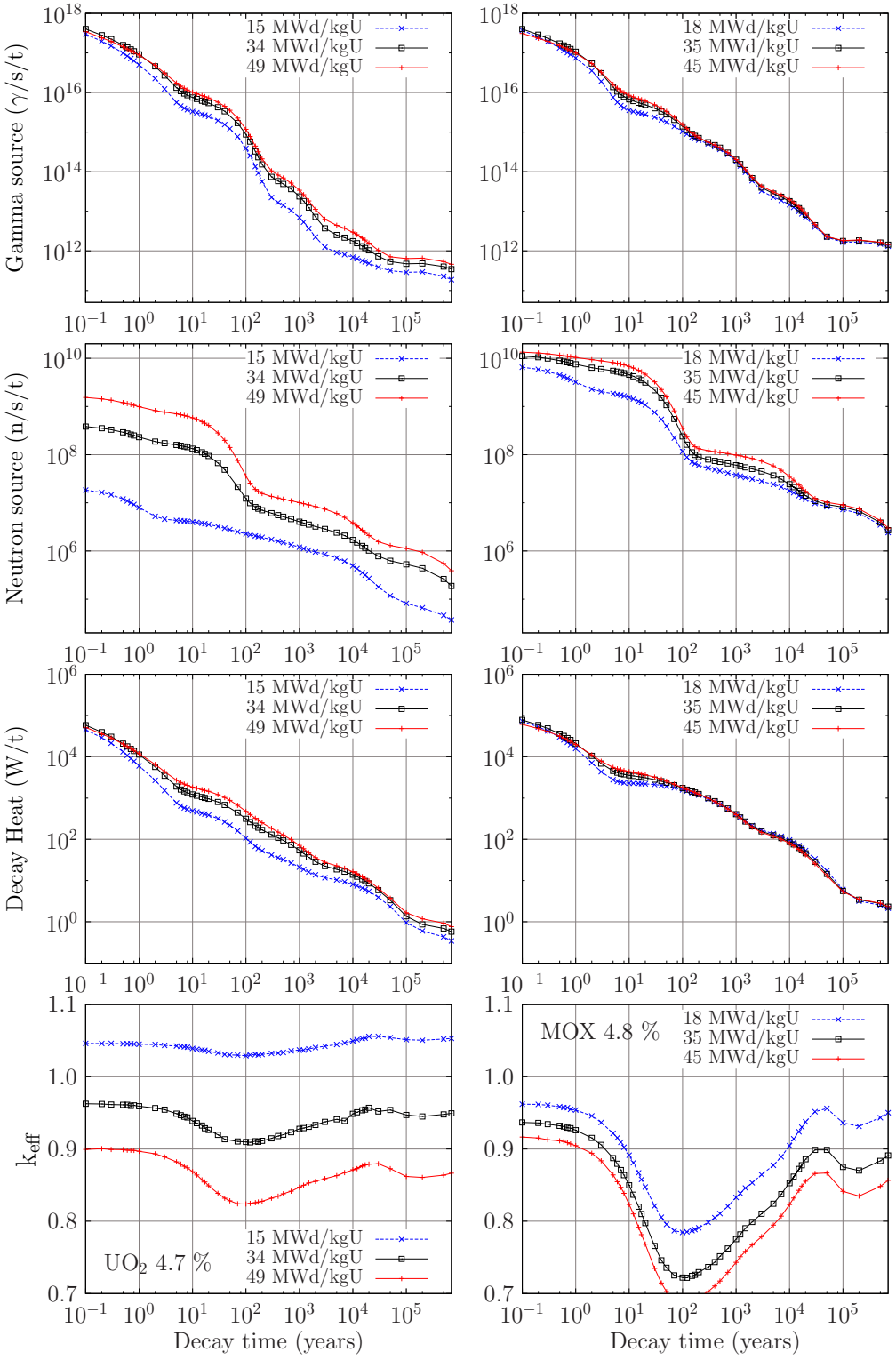

Fig. 4. Examples for a UO_2 (left) and MOX (right) fuel assembly for k_{eff}, decay heat, neutron and gamma source as a function of cooling time. The same assembly is used for 3 successive cycles, represented in the figure by three burnup values.

But for the majority of them, the k_{eff} calculations are done only at 0.1 year cooling time. This allows to drastically reduce the number of MCNP calculations, but does not affect the conclusion of this demonstration.
Considering only the k_{eff} calculations at 0.1 year, the loading curve can be calculated at a specific fuel enrichment by comparing the k_{eff} for each assembly canister loading for this fuel enrichment. An example for all UO_2 assembly-cycles with an enrichment of 4.7 % is presented in Fig. 5. In this figure, the

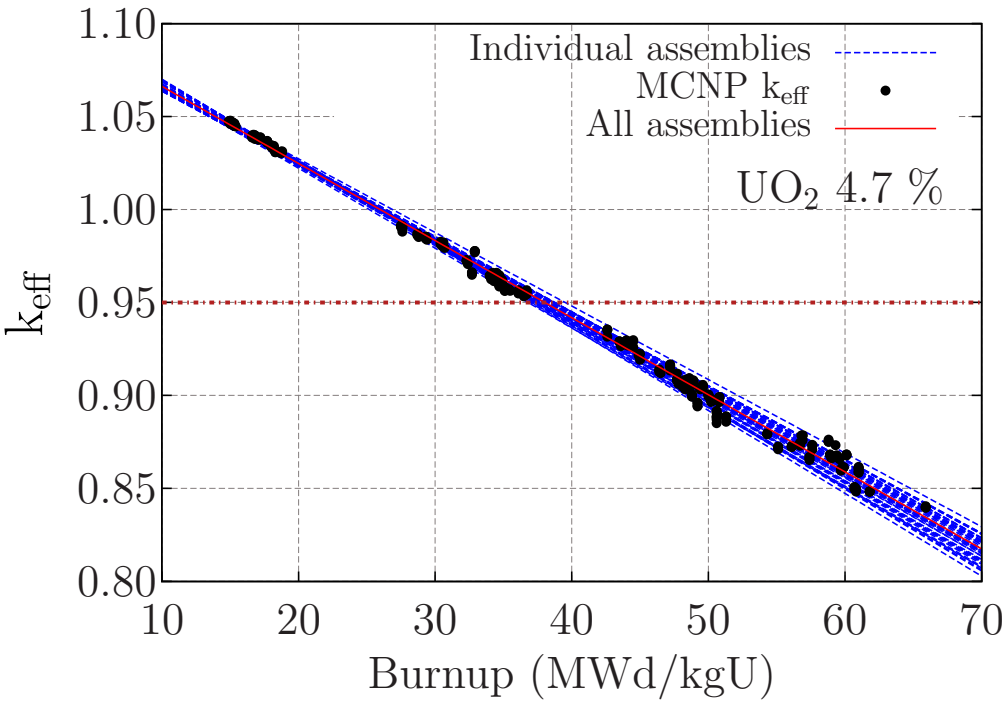

Fig. 5. Example of the calculated k_{eff} for the UO_2 assemblies with an enrichment of 4.7 %. The blue and red lines represent fits of the data. The dark horizontal line is the limit of $k_{eff} = 0.95$. Actual assembly values are represented by black dots.

points represents the actual MCNP k_{eff} calculations (88 unique assemblies, representing 356 assembly-cycles). The k_{eff} values higher than 0.95 indicate that such loading patterns are not allowed from the criticality point-of-view. In order to find the limit in terms of assembly burnup, one can either fit all the assembly-cycles together, or each assembly individually. In the first case, a unique average value for the limit is obtained (38.02 MWd/kgU, represented by the red line in Fig. 5), and in the second case, several values are obtained, one for each assembly. By repeating such study for all the enrichments, the loading curve can be obtained, as presented in Fig. 6. In this figure, the loading curve is obtained by fitting all assembly-cycles together for a specific enrichment. The gray band in Fig. 6 represents the region where the loading of such assemblies can lead to a k_{eff} higher than 0.95. The MOX assemblies, leading to lower k_{eff} values compared to the UO_2 assemblies with similar fissile enrichments are plotted separately. As only one enrichment of MOX is considered, the slope of the MOX loading curve in Fig. 6 is presented as an indication and

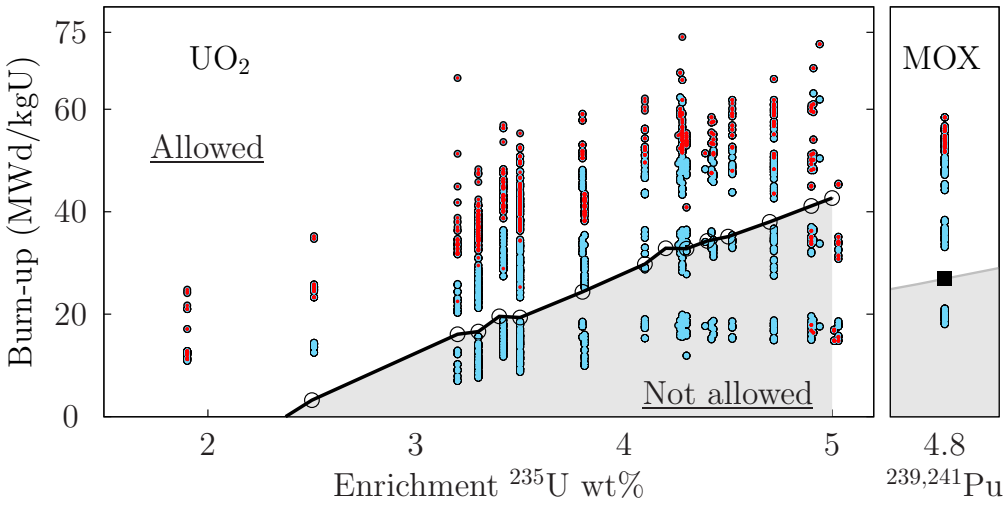

Fig. 6. Loading curves (one line for UO_2 and one for MOX) calculated with $k_{eff} = 0.95$, taking into account all assembly-cycles. The lines denotes the limit between the allowed and not allowed zones for the canister loading with 4 identical assemblies. The blue dots are all the assembly-cycles considered (assemblies being discharged or reloaded) and the red dots indicate the assemblies used in their last cycles.

is similar to the UO_2 cases. On this figure, the blue and red dots represent the 6012 assembly-cycles (among which 640 MOX) used to calculate the loading curve. The red dots represent the assemblies in their last cycles. For the majority of them, it corresponds to their end of life, but as this study is using data up to a specific cycle, some assemblies will be used again (especially the ones with a high enrichment and low burnup values, likely to be used in cycles posterior to this study).

As presented in Fig. 5, one can also use the burnup limit for each assembly instead of an average value representing all assemblies. In this case, a distribution of burnup limits (for a given enrichment) is obtained, characterized by its average, standard deviation and extreme values. Such values are presented in Fig. 7. As observed, the bands defined by one standard deviation or by the extreme values (minimum and maximum) are not very large in the case of UO_2 fuel, slightly decreasing with the enrichment value. In the case of the MOX fuel, the spread is more pronounced: 8.9 MWd/kgU compared to 2.8 for the UO_2 at 4.7 %. This indicates a stronger variability of the loading limit in the case of the MOX fuel, showing that the individual assembly history is important in the determination of the loading curve.

3.2 Comparison with radiation sources

As the radiation sources are calculated for each assembly-cycle, the quantities such as activities, decay heat, and the neutron & gamma emissions can be

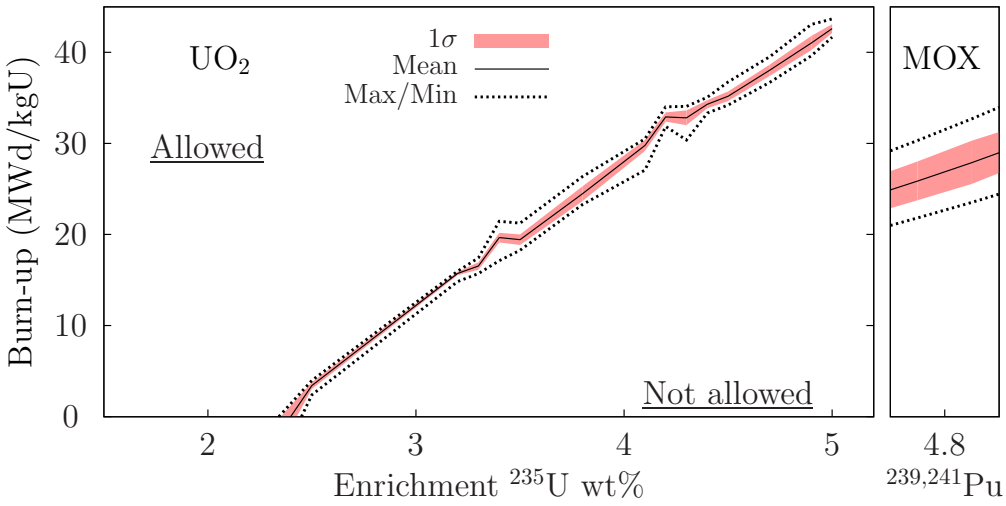

Fig. 7. Maximum, minimum and average loading curves calculated with $k_{eff} = 0.95$, taking into account all assembly-cycles.

compared with the k_{eff} values. Such comparisons are presented in Figs. 8 to 10, allowing to assign radiation values to the loading map, or in different terms to know the k_{eff} and the radiation values for each assembly-cycle. Note that the quoted activity, neutron and gamma emission are given for a single assembly and not for the canister.

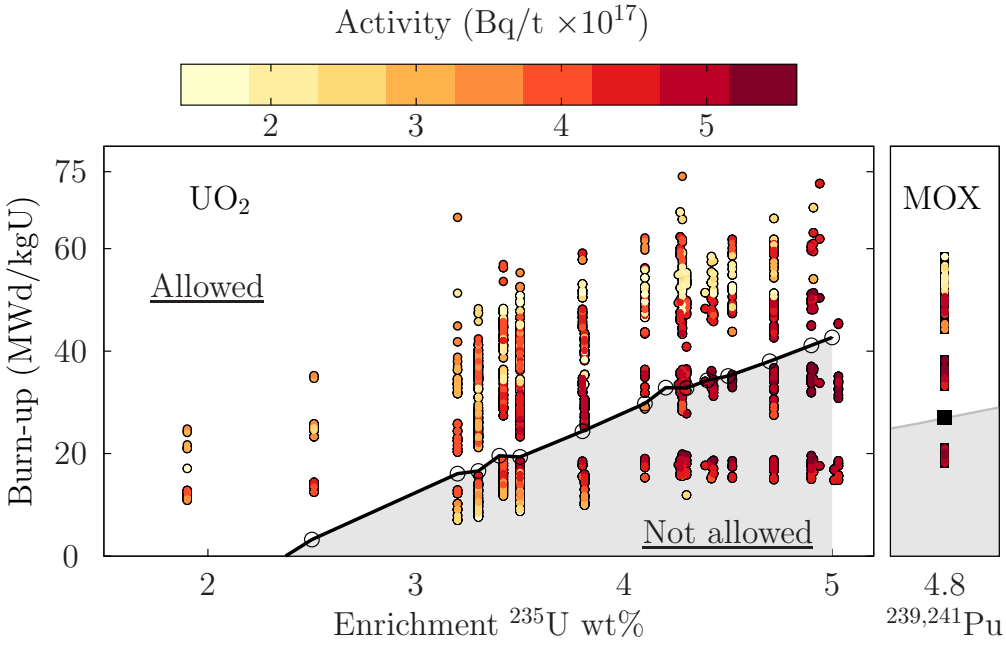

Fig. 8. Loading curve (with $k_{eff} = 0.95$) and activity map, taking into account all assembly-cycles. A very similar plot is obtained for the decay heat (in W/t).

One can see that for the activity and the decay heat (not represented, but its distribution is very similar to the one for the activity), high values are obtained in both the allowed and not allowed zones. It can be seen that for

the same burnup values (and enrichments), a difference of a factor 2 can appear for the assembly activity. Examples are provided in Table 3 for three sets of similar assembly-cycles. For the six assemblies presented, the ones which were

Table 3
Characteristics of some assemblies with very similar burnup values, but different activities, for the UO_2 case with 4.28 % enrichment. The "␣" mark indicates cycles for which the specific assembly was not used.

Assembly name	Cycle	Activity W/t×10^{17}	Burnup MWd/kgU	Previous cycles
A1	25	16.6	48.1	20, 21,␣,23,␣
A2	22	45.1	48.2	20, 21
B1	27	18.4	49.1	22, 23, 24,␣,␣
B2	21	44.8	49.1	19, 20
C1	30	25.3	59.3	21,␣,␣,24,25,␣,␣,␣,29
C2	22	42.0	61.3	19, 20, 21

not used in consecutive cycles present lower activity values. This indicated that the irradiation gap (in terms of cycle) before the last cycle is important, and that the irradiation gaps during the assembly irradiation history are also important. In the case of assemblies not used in consecutive cycles, the shorter lived fission products have decayed, leading to lower assembly activity. Again, this indicates that the specific assembly irradiation history is important to obtain correct values.
The neutron source is presented in Fig. 9. In this case, the neutron emission does not represent an heterogeneous distribution as a function of the assembly burnup, contrary to the activity. On the contrary, in the case of the UO_2 fuel, the number of emitted neutron is increasing, globally following a parabolic function:

$$n_{\mathrm{UO2}} = a + b \times \mathrm{BU} + c \times \mathrm{BU}^2 \quad (1)$$

with $a = 1.9 \times 10^9$, $b = -3.02 \times 10^7$ and $c = 1.26 \times 10^6$ and BU is the assembly burnup (n_{UO2} in n/s/t and BU in MWd/kgU). For the MOX fuel, the neutron emission is linearly increasing:

$$n_{\mathrm{MOX}} = a + b \times \mathrm{BU} \quad (2)$$

with $a = 1.02 \times 10^9$ and $b = 2.80 \times 10^8$.

The difference between UO_2 and MOX fuel in terms of neutron emission comes from the fact that the main neutron emitters (such as ^{244}Cm) are produced at a faster rate for the MOX fuel, directly from the built-up from ^{239}Pu and not ^{238}U.
A similar remark as for the activity can be made for the gamma emission.

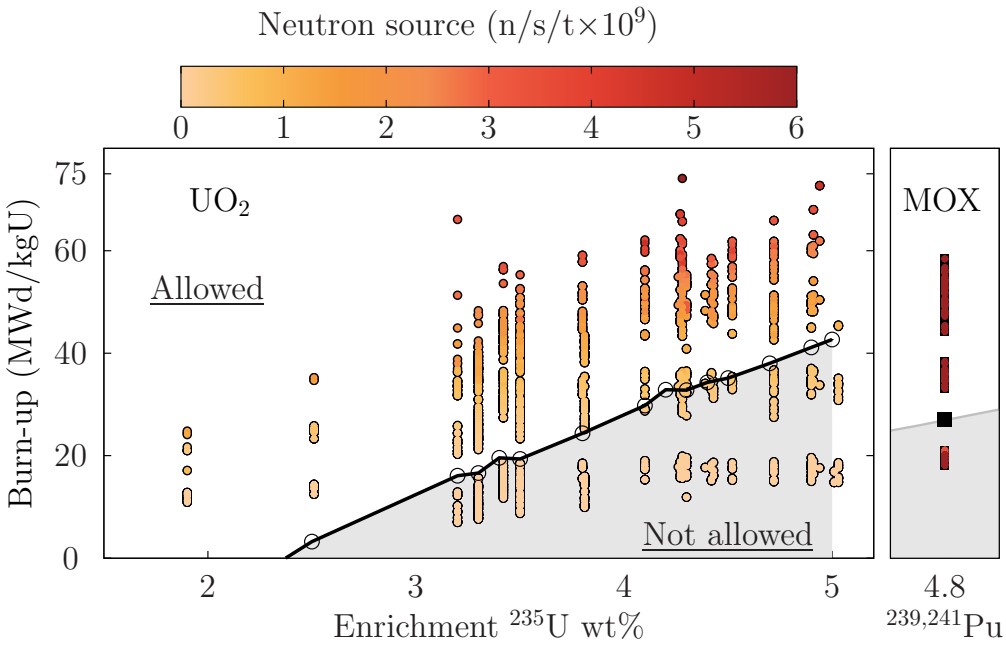

Fig. 9. Loading curve (with $k_{eff} = 0.95$) and neutron emission map, taking into account all assembly-cycles.

There is indeed a strong correlation between the activity and the gamma emission. Here again, the specific assembly histories (with cooling time before being reused in later cycles) strongly influence the gamma emission, as some shorter lived fission products can decay.

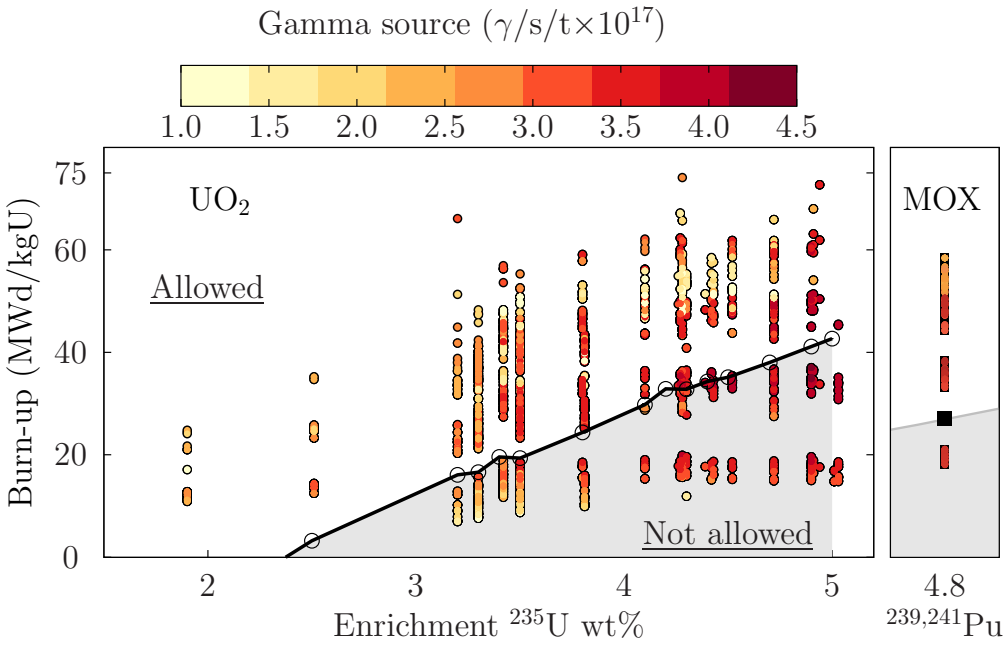

Fig. 10. Loading curve (with $k_{eff} = 0.95$) and gamma emission map, taking into account all assembly-cycles.

Such general observations indicate that there is a possibility of optimization of the canister loading for assemblies with the same burnup values: if well chosen, the decay heat and gamma emission can be minimized while using assemblies of the same enrichment and burnup for the canister loading. This can only be achieved while considering the individual assembly histories. Alternatively, the distribution of the neutron emission was seen to increase with the assembly burnup (see Fig. 9): the region of allowed loading corresponds to the cases with the highest neutron emission (for both UO_2 and MOX). Therefore, from the neutron emission point-of-view, a canister loaded with assemblies having high burnup values will also have a high neutron emission rate, independently of the individual assembly history.

4 Conclusion

This study presents the possibility to calculate radiation quantities for spent fuel assemblies and criticality values for their handling and storage, based on their specific assembly isotopic inventory. Such inventory comes from an in-core fuel management system, validated core models and on four state-of-the-art codes: CASMO, SIMULATE, SNF and MCNP6. The importance of the assembly irradiation history was illustrated, notably of the cooling time between cycle for the assembly activity, decay heat and gamma emission. By considering the specific assembly histories, optimization for canister loading can be done for criticality, activity and decay heat in order to minimize risks and environmental irradiation.

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