

**Impact of Storage Time on the Needed Capture Efficiency  
for Volatile Radionuclides – 13369**

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**ABSTRACT**

During the processing of used nuclear fuel (UNF), volatile radionuclides will be discharged from the facility stack if no recovery processes are in place to limit their release. The volatile radionuclides of concern are <sup>3</sup>H, <sup>14</sup>C, <sup>85</sup>Kr, and <sup>129</sup>I. There are three key regulations that address the release of these radionuclides to the environment — 40 CFR 61, 40 CFR 190, and 10 CFR 20. These regulations apply to the total radionuclide release and establish dose limits for the maximum exposed individual (MEI) in the public both in terms of whole body dose and dose to specific organs such as the thyroid. Each radionuclide released to the environment contributes to the total dose to some degree. In this paper we attempt to evaluate the efficiency requirements for the capture processes to limit the doses to the MEI to regulatory levels. Since the total amount of each volatile radionuclide present in the UNF changes with the age of the fuel, the respective capture requirements also change with time. Specifically, we are interested in the impact of the decreasing contribution of <sup>3</sup>H and <sup>85</sup>Kr, which have relatively short half-lives, 12.32 y and 10.76 y, respectively, with the increasing age of the fuel (i.e., time between when the UNF is removed from the reactor and the time it is processed) on the capture requirements.

In this paper we examine the capture requirements for these four radionuclides for three fuel types (pressurized water reactor [PWR] with uranium oxide fuel [UOX], PWR with mixed oxide fuel [MOX], and an advanced high temperature gas-cooled reactor [AHTGR]), several burnup values, and time out of reactor extending to 200 y. We calculate doses to the MEI with the EPA code CAP-88 and look at two dose contribution cases. In the first case, we assume that the total allowable dose is attributed to only the four volatile radionuclides. This establishes the lowest capture efficiency value possible. Since this is unrealistic, because it assumes zero dose contribution from all other radionuclides, we also examine a second case, where only 10% of the allowable dose is assigned to the four volatile radionuclides. We calculate the required decontamination factors (DFs) for both of these cases for the three fuel types, multiple fuel burnups, and fuel ages and determine whether or not the dose to the whole body or to the thyroid that drives the capture requirements would require additional effluent controls for the shorter half-life volatile radionuclides based on dose considerations. This analysis indicates that the principal isotopes of concern are generally <sup>3</sup>H and <sup>129</sup>I, the latter requiring the highest DFs. The maximum DF value for <sup>129</sup>I is 8000 for the evaluated cases and assumptions used.

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~60 for fresh fuels. The DF for  $^{14}\text{C}$  could be as high as 30 for certain fuels. These values are based on just meeting the regulatory limits, and additional engineering margins (perhaps  $3\times$  to  $10\times$  or higher) should be applied to provide a safety factor for compliance. However, by assuming less conservative dose allocations, taller stacks, different radionuclide speciation, fuel aging, and other reprocessing facility design and location parameters, the DF requirements could be significantly reduced.

## INTRODUCTION

Atmospheric releases of radionuclides during the reprocessing of used nuclear fuel (UNF) must be controlled to levels that comply with air emissions regulations. Radionuclides that tend to form gaseous species that evolve into reprocessing facility off-gas systems are more challenging to efficiently control compared to radionuclides that tend to be contained in solid or liquid phases. Radionuclides that have been identified as “gaseous radionuclides” are noble gases (most notably isotopes of krypton and xenon, tritium, radiocarbon, and radioiodine):  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ , and  $^{129}\text{I}$ . (Note that all radioactive xenon isotopes have very short half-lives relative to the time frames for this study and have decayed to the extent that they do not contribute to the dose.) In aqueous reprocessing, these radionuclides are most commonly expected to evolve into off-gas streams as tritiated water [ $^3\text{H}_2\text{O}$  ( $\text{T}_2\text{O}$ ) and  $^3\text{HHO}$  ( $\text{THO}$ )], radioactive  $\text{CO}_2$ , noble gases, and gaseous  $\text{HI}$ ,  $\text{I}_2$ , or volatile organic iodides. The amount of each radionuclide depends on the fuel burnup and the storage time (age) since its removal from the reactor. The concentration of these radionuclides in the various gas streams from which they must be removed depends on the fuel throughput, the time since the fuel was discharged from the reactor, and the process equipment designs, which ultimately determine the total flow rates of the off-gas streams (Fig. 1). Detailed information on the fate of these radionuclides from a nonaqueous fuel reprocessing plant is not available at this time, but active investigations are taking place. Therefore, we have limited our scope to aqueous reprocessing of UNF. The factors that play an important role in the capture requirements or the required decontamination factors (DFs) for the volatile radionuclides are fuel age (time since end of irradiation), plant size (fuel throughput), and fuel burnup.

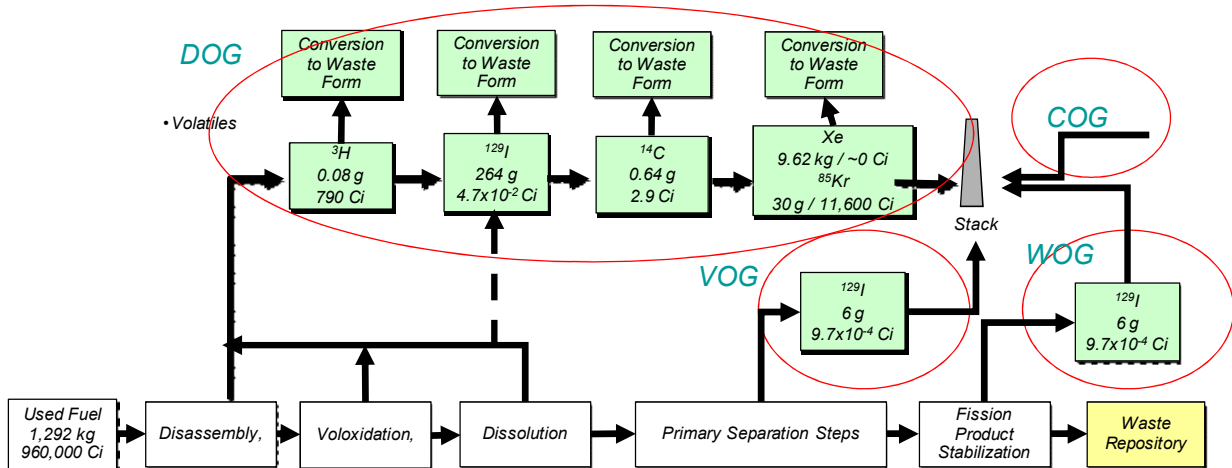


Fig. 1. Volatile fission / activation products from processing 1 t of spent nuclear fuel at 60 Gwd/tHM with 5 years of cooling.

Krypton capture and storage has been discussed as a potentially very costly portion (up to 15–20%) of the total costs for a separations facility.\* Because tritium readily exchanges for hydrogen in water, the recovery of tritium is expensive as well. Fuel aging prior to separations allows decay of short-lived gaseous fission products  $^3\text{H}$ ,  $^{85}\text{Kr}$ ,  $^{131}\text{I}$ , and the radioactive isotopes of xenon. In the context of this study, “fuel aging” refers to the storage time between removal from the reactor core and the processing of the UNF. The  $^{131}\text{I}$  and radioactive isotopes of xenon have such short half-lives (< 37 d) that if the fuel ages up to a year after reactor discharge, then these isotopes are decayed to negligible levels in the UNF. Further aging of the fuel before reprocessing allows the  $^3\text{H}$  ( $t_{1/2} = 12.32$  y) and  $^{85}\text{Kr}$  ( $t_{1/2} = 10.76$  y), to decay, hence reducing the potential emissions of these radionuclides and their overall dose impact.

In this study, we assess the implications of fuel age and the need for capturing the short-lived fission products  $^3\text{H}$  and  $^{85}\text{Kr}$ . Although the  $^{14}\text{C}$  and  $^{129}\text{I}$  do not significantly decay within reasonable fuel aging time periods, the potential need to control emissions of these radionuclides during UNF reprocessing can also affect the cost and complexity of the off-gas system and the cost and complexity of managing waste streams that are generated. An insignificant amount of  $^{14}\text{C}$  comes from fission; however, significant quantities can come from the activation of trace nitrogen in the fresh fuel, or from  $^{13}\text{C}$  if present in fresh fuel for high-temperature gas reactors. This is discussed in sufficient detail to justify including  $^{14}\text{C}$  in this study.

Ultimately letting the fuel age before it is reprocessed may allow the short-lived radionuclides to decay to inventories that may be released without any removal or with removal efficiencies that are much easier to attain in an operating plant. Recent studies have shown that the optimum storage time is 30 to 70 y [1, 2]; we expand on these studies.

The question we attempted to answer was what capture efficiency is needed for each of these radionuclides so that the off-gas emissions from aqueous used fuel reprocessing comply with U.S. regulatory limits. To answer this question, we examined the three regulations that may impact the degree to which these radionuclides must be reduced before process gases can be released from the facility—40 CFR 190 [3], 40 CFR 61 [4], and 10 CFR 20 [5]. The regulations apply to the total radionuclide release for some radionuclides, and to the aggregate dose from all emitted radionuclides to the whole body and to specific organs, in particular the thyroid.

We performed calculations to estimate gaseous radionuclide capture efficiencies that could be needed for assumed used fuels, a range of fuel aging (also called cooling times), and doses estimated using air dispersion calculations. We began by calculating the radionuclide inventories for the radionuclides of concern for this study. These inventories were based on three reactor and fuel types and a range of fuel burnups from 20 to 100 GWd/tIHM. We then looked at the effects of storing these fuels for up to 200 y. We then used the CAP-88 code [6, 7] (an EPA code for calculating dose at locations from an emission stack) to evaluate the dose to the maximum exposed individual (MEI) and determine the required DFs for the radionuclides of concern to meet regulations. We determined the dose from the four isotopes ( $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ , and  $^{129}\text{I}$ ) over a 200-y time span. We soon found that control efficiencies needed to meet whole body dose limits bounded the control efficiencies needed to meet limits for individual organs except for the thyroid, so we included both whole body and thyroid dose calculations. These calculations showed the impact on the whole body dose and the thyroid on a by-isotope basis so that the strategy of aging fuel to meet regulatory releases or provide more “head space” for required DFs could be assessed.

The results of this study can be used as input to broader systems analysis studies in which the implications of fuel age with respect to the fuel cycle as a whole are investigated. The results

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\* Mr. Paul Murray, AREVA, personal communication.

shown here should serve to inform future analyses of the impact of regulatory requirements of the U.S. Environmental Protection Agency (EPA) and the U.S. Nuclear Regulatory Commission (NRC).

## REGULATORY BASIS AND SOURCE TERMS

As part of the advanced fuel cycle effort, reprocessing of UNF is being considered [8]. Since the last time fuel reprocessing was investigated, new regulations have been put in place to govern the release of radionuclides from a reprocessing plant and the rest of the fuel cycle. Within the scope of this study, the volatile radionuclides  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ , and  $^{129}\text{I}$  are, in part, the nuclides that will need to be controlled to meet these regulations. We recognize, however, that other radionuclides are addressed in the regulations for allowable releases, for example, isotopes of plutonium. The scope of the study reported here covers only the four volatile radionuclides mentioned and is the reason that we have included different dose allocations for these radionuclides in this study. Thus, the impact of these regulations on the emissions control efficiencies needed for these radionuclides and the impact of fuel age on the required control efficiencies, expressed as DFs, are the subjects of this paper.

Release of radionuclides from the entire fuel cycle is regulated by 40 CFR 190.10 [3]. This regulation states that

*The annual dose equivalent does not exceed 25 millirems to the whole body, 75 millirems to the thyroid, and 25 millirems to any other organ of any member of the public as the result of exposures to planned discharges of radioactive materials, radon and its daughters excepted, to the general environment from uranium fuel cycle operations and to radiation from these operations...*

and

*The total quantity of radioactive materials entering the general environment from the entire uranium fuel cycle, per gigawatt-y of electrical energy produced by the fuel cycle, contains less than 50,000 curies of krypton-85, 5 millicuries of iodine-129, and 0.5 millicuries combined of plutonium-239 and other alpha-emitting transuranic radionuclides with half-lives greater than one year.*

The latter restriction has the greatest impact on the operation of a reprocessing plant because it regulates the release of the volatile radionuclides, with iodine and krypton being mentioned explicitly. Additionally, 10 CFR 20 [5] and 40 CFR 61[4] regulate the doses to the MEI near a reprocessing plant, primarily from stack releases. To meet these release restrictions for a plant, certain DFs are needed for the removal of these radionuclides from the gaseous effluent.

This study includes the evaluation of releases from the processing of three fuel types—pressurized water reactor uranium oxide (PWR UOX), pressurized water reactor mixed oxide (PWR MOX), and advanced high-temperature gas-cooled reactor (AHTGR)—at several burnup values, and times out of reactor extending to 200 y. Table I provides the quantity of each of the four radionuclides of interest for the PWR UOX case as a function of fuel age and burnup. For purposes of this study, the shortest fuel cooling time considered is 2 y. The terms “fuel age” and “cooling time” refer to the time since the fuel was discharged from the reactor. At extremely short cooling times, dose contributions from  $^{131}\text{I}$  xenon would need to be considered. Also note that the increased amount of  $^{129}\text{I}$  in the 2-y-cooled fuel and beyond compared to that at time of discharge is from the decay of  $^{129}\text{Te}$ .

The dose resulting from the release of these volatile radionuclides was calculated using the Clean Air Act Assessment Package (CAP88-PC Version 3.0) computer model [6, 7]. CAP88-PC is a set of computer programs, databases, and associated utility programs used to estimate dose and risk to members of the public from radionuclide emissions in the air. Version 3.0 of CAP-88 incorporates dose and risk factors from Federal Guidance Report 13 [9], which are based on the methods of the International Commission on Radiological Protection [10]. Emission monitoring and compliance procedures for DOE facilities require the use of the CAP-88 model, or other approved methodologies, to estimate the effective dose to members of the public [6, 7].

**Table I. Source terms, by isotope, for PWR reactor with UOX fuel with time since discharge**  
Isotopic mass (g/tIHM) – 20-GWd/tIHM burnup

	Time since discharge (y)								
	0	2	5	10	20	30	50	70	100
<sup>3</sup> H	0.052	0.047	0.040	0.030	0.017	0.010	0.003	0.001	0.000
<sup>14</sup> C	0.105	0.105	0.105	0.105	0.105	0.105	0.104	0.104	0.104
<sup>85</sup> Kr	22.41	19.71	16.24	11.77	6.18	3.24	0.894	0.246	0.036
<sup>129</sup> I	84.97	85.04	85.04	85.04	85.04	85.04	85.04	85.04	85.04

Isotopic mass (g/tIHM) – 30-GWd/tIHM burnup

	Time since discharge (y)								
	0	2	5	10	20	30	50	70	100
<sup>3</sup> H	0.084	0.075	0.063	0.048	0.027	0.015	0.005	0.002	0.0003
<sup>14</sup> C	0.163	0.163	0.163	0.163	0.163	0.163	0.162	0.162	0.161
<sup>85</sup> Kr	30.79	27.08	22.32	16.17	8.49	4.46	1.23	0.338	0.049
<sup>129</sup> I	134.3	134.4	134.4	134.4	134.4	134.4	134.4	134.4	134.4

Isotopic mass (g/tIHM) – 60-GWd/tIHM burnup

	Time since discharge (y)								
	0	2	5	10	20	30	50	70	100
<sup>3</sup> H	0.184	0.164	0.139	0.105	0.060	0.034	0.011	0.0036	0.0007
<sup>14</sup> C	0.376	0.376	0.376	0.376	0.375	0.375	0.374	0.373	0.372
<sup>85</sup> Kr	48.45	42.59	35.11	25.43	13.35	7.008	1.931	0.532	0.077
<sup>129</sup> I	291.0	291.1	291.1	291.1	291.1	291.1	291.1	291.1	291.1

Two primary dose cases were considered. The first case, perhaps unrealistic, assumed that all of the allowable dose was assigned to the volatile radionuclides. This case disregards any contribution to the total dose from any other radionuclides – it assumes 100% control of all other radionuclides during reprocessing. In lieu of this, for the second case a value of 10% of the allowable dose was arbitrarily selected to be assigned to the volatile radionuclides, which reserves 90% of the total allowable dose for other radionuclides. The required DFs were calculated for both of these cases, including the case for the thyroid dose for which  $^{14}\text{C}$  and  $^{129}\text{I}$  are the main contributors. However, for completeness, for one fuel type and burnup, additional cases were provided, allowing 25% and 50% of the allowable dose to be assigned to the volatile radionuclides.

In this study, we calculated the minimum ages for each fuel type that would not require additional effluent controls for the shorter half-life volatile radionuclides based on dose considerations. With respect to  $^{129}\text{I}$  doses, we find that the highest dose is calculated with iodine as a fine particulate. The dose scaled with the fraction of the total  $^{129}\text{I}$  that was particulate. While we assumed for all of our calculations that 100% of the  $^{129}\text{I}$  was particulate, the users can scale our calculated doses to their needs.

We calculated the required DF values based on the whole body dose limiting case and thyroid dose limiting case and used the more restrictive of the two.

The following is an example case for one fuel type and one burnup.

**Step 1. Calculate whole body dose and maximum plant size requiring no dose-based controls for a fuel type (not yet considering the 40 CFR 190 fuel cycle limits for  $^{85}\text{Kr}$  and  $^{129}\text{I}$ ).** In the case of PWR UOX fuel, the maximum plant size for 30 GWd/tIHM fuel cooled 5 y is 37.6 t/y; if the fuel is cooled 100 y, the plant size is increased slightly to 45.2 t/y (Fig. 2). If the allowable contribution of the volatile components to the total dose is limited to 2.5 mrem/y (i.e., 10% of the total dose), the plant sizes drop to 3.8 t/y and 4.5 t/y for cooling times of 5 and 100 y, respectively. At 5 y cooling,  $^{129}\text{I}$  contributes 82.7% of the total dose and tritium contributes 16.1% of the total dose. For a 100-y-cooling period,  $^{129}\text{I}$  contributes 99.4% of the total dose.

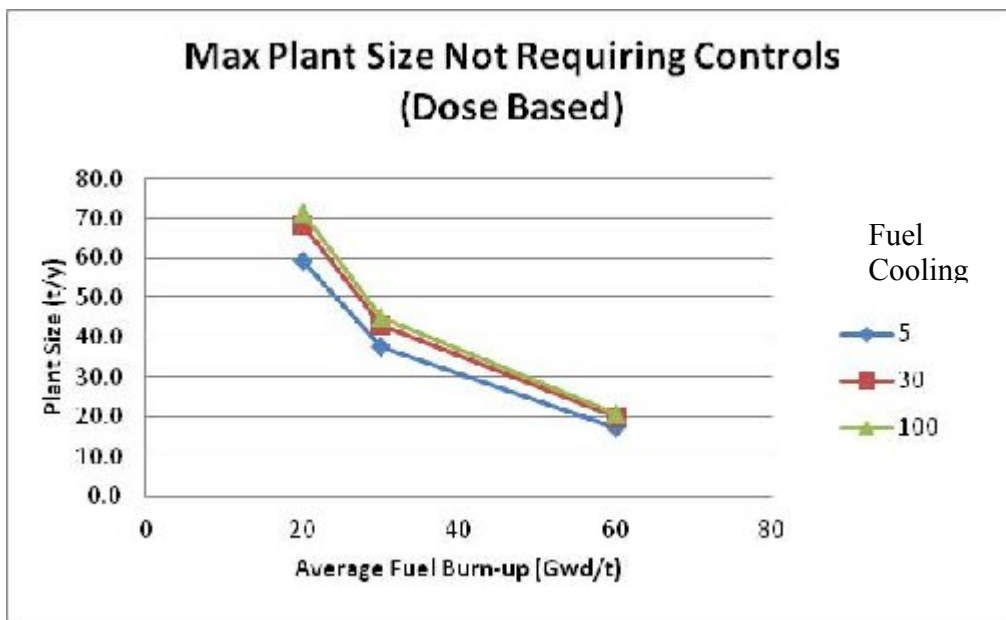
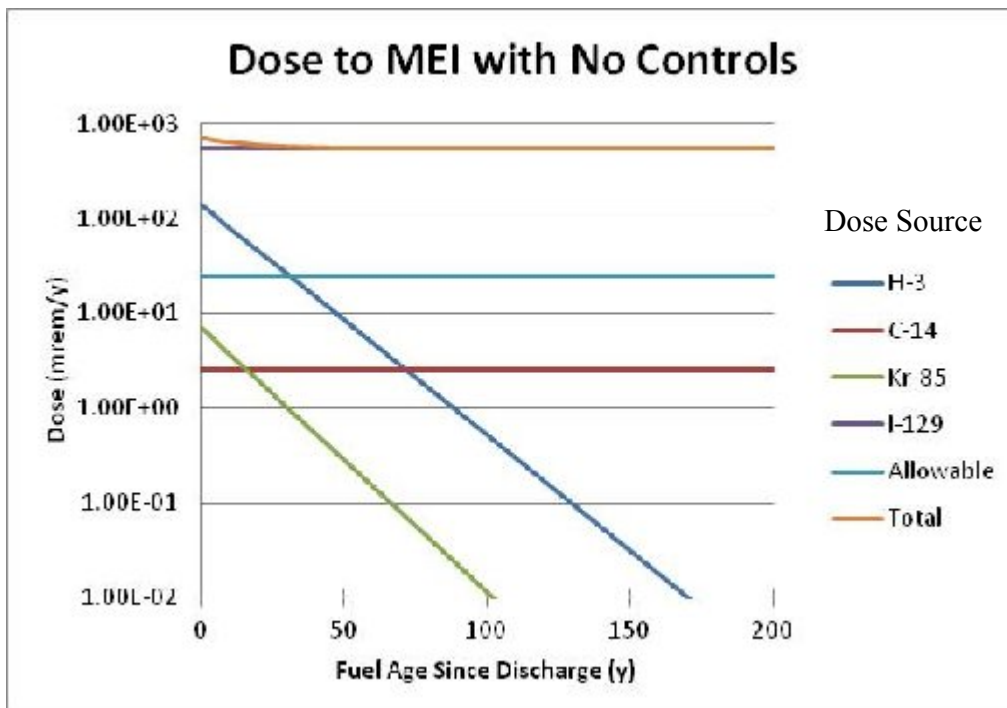


Fig. 2. Maximum plant size for processing PWR UOX fuel that would not require controls to limit exposure to the MEI to  $\leq 25$  mrem/y.

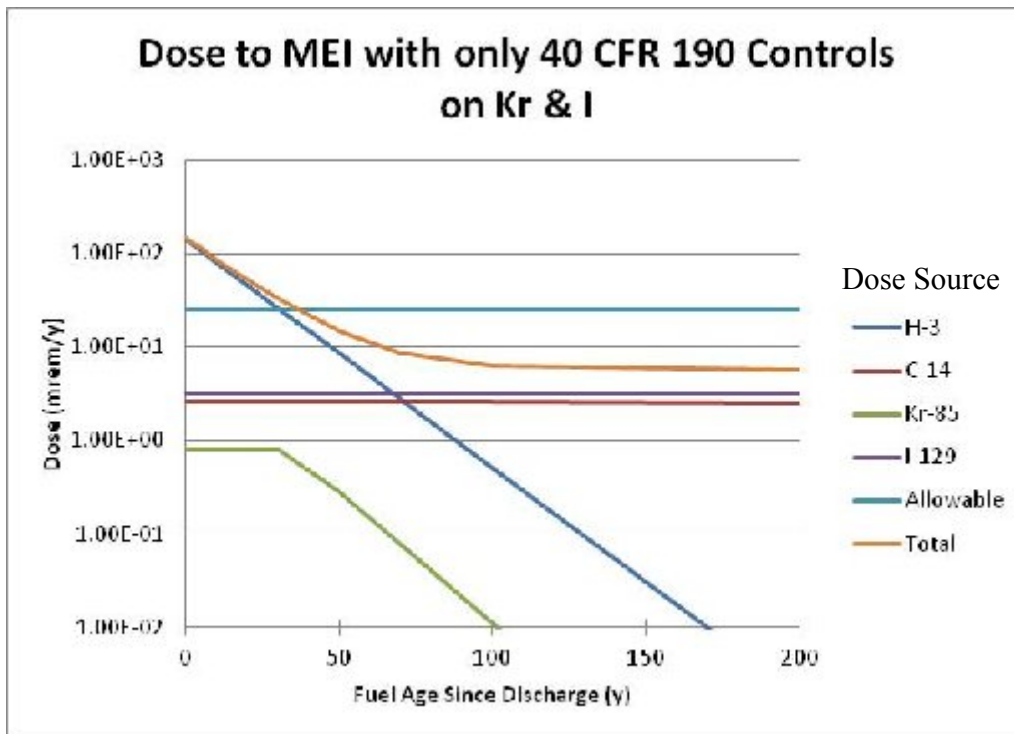
In the second phase of the evaluation, the impact of time since discharge was evaluated in three steps for each fuel type and fuel burnup. Now, focusing only on the case of 30-GWd/tIHM burnup for the detailed calculations of DF requirements, the next steps are as follows.

**Step 2. Calculate uncontrolled dose contributions.** For the case of the 30-GWd/tIHM fuel and a plant size of 1000 t/y, the total dose to the MEI ranges from 686 mrem/y for 2-y-cooled fuel to 553 mrem/y for 100-y-cooled fuel (Fig. 3). Iodine is the major contributor (550 mrem/y). At cooling times up to 30 y, tritium dose contributions exceed 25 mrem/y; <sup>14</sup>C contributes 2.6 mrem/y at all cooling times; and <sup>85</sup>Kr could contribute up to 6 mrem/y at short cooling times.



**Fig. 3. Total whole body dose and dose contributions arising from the volatile radionuclides to the MEI from 1000-t/y recycle plant processing 30-GW/tIHM PWR UOX fuel.** Allowable dose limit is shown at the 25-mrem whole body dose level.

**Step 3. Apply 40 CFR 190 release limits.** If the iodine and krypton release limits imposed by 40 CFR 190 are then applied to the uncontrolled dose contributions (step 2), the whole body dose to the MEI is reduced to 134 mrem/y at 2-y cooling and 6.24 mrem/y for 100-y-cooled fuel (Fig. 4). The iodine contribution is reduced to 3.15 mrem/y, and <sup>3</sup>H becomes the major contributor at cooling times less than 50 y. Carbon-14 again contributes 2.6 mrem/y, but <sup>85</sup>Kr contributions are reduced to less than 0.8 mrem/y. It is clear from Fig. 4 that additional controls at least on <sup>3</sup>H would be required to meet a 25-mrem/y limit. For a 2.5-mrem/y apportioned limit, additional controls (DF) for iodine and carbon would also be required.



**Fig. 4. Total whole body dose and dose contributions arising from the volatile radionuclides to the MEI from 1000-t/y recycle plant processing 30-GWd/tHM PWR UOX fuel with iodine and krypton controls per 40 CFR 190. Allowable dose limit is shown at the 25-mrem whole body dose level.**

**Steps 4 and 5. Apply additional DF requirements as needed to limit dose to MEI to regulatory limits.** Additional controls were imposed as needed on individual isotopes in order to reduce the calculated emissions to the extent required to reduce the dose to the MEI to the regulatory limit or to a portion of the regulatory limit. In general, we attempted to impose control on as few individual radionuclides as possible, since each would likely require a separate process to control and result in a separate waste stream that would need treatment. In selecting the required DF, there is no single correct answer, since the total dose is a sum of the individual contributions. The reduction in one allows more “headroom” for another. A nominal goal was to reduce the dose contribution from an individual radionuclide to somewhere in the range of 10–50% of the total allowable dose. Additional controls were applied first to species that exceeded the allowable limits. For  $^3\text{H}$  and  $^{85}\text{Kr}$ , these were typically applied over the periods of fuel age in which the dose contributions exceeded 30–50% of the allowable dose. Additional controls were typically added to  $^{129}\text{I}$  before  $^{14}\text{C}$ , since  $^{129}\text{I}$  control would be required in all cases anyway.

For the 25-mrem/y limit case, additional controls were imposed for  $^3\text{H}$ . Tritium DF requirements ranged from 6.9 at 2-y cooling to 1.43 at 30-y cooling (Fig. 5). Beyond ~35-y fuel cooling, no additional  $^3\text{H}$  capture would be required if the total allowable dose to the MEI was considered to result from only  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ , and  $^{129}\text{I}$ . If these four radionuclides are apportioned 2.5 mrem/y or 10% of the allowable dose, then additional controls must be placed on  $^{14}\text{C}$  and  $^{129}\text{I}$  for all fuel ages, since these two radionuclides individually contribute more than 2.5 mrem/y and  $^3\text{H}$  contributes more than 2.5 mrem/y until the fuel is aged over 70 y. An additional recovery factor of 10 was applied to  $^{129}\text{I}$  for all fuel ages, raising the total  $^{129}\text{I}$  DF to ~1200. A  $^{14}\text{C}$  DF of 3 was applied to reduce its contribution to the total dose to ~0.8 mrem/y. Tritium DFs of 250 would be required for 2-y-cooled fuel. Beyond ~80 y, no  $^3\text{H}$  recovery would be required. Figure 5 shows



the resulting total dose to the MEI and the individual dose contributions as a function of time, for the 25-mrem/y allocation. Similar calculations were completed to determine the dose contributions for the 2.5-mrem/y allocation and the associated DFs required to limit the dose to the MEI to 2.5 mrem/y. Figure 6 shows the associated DFs as a function of cooling time.

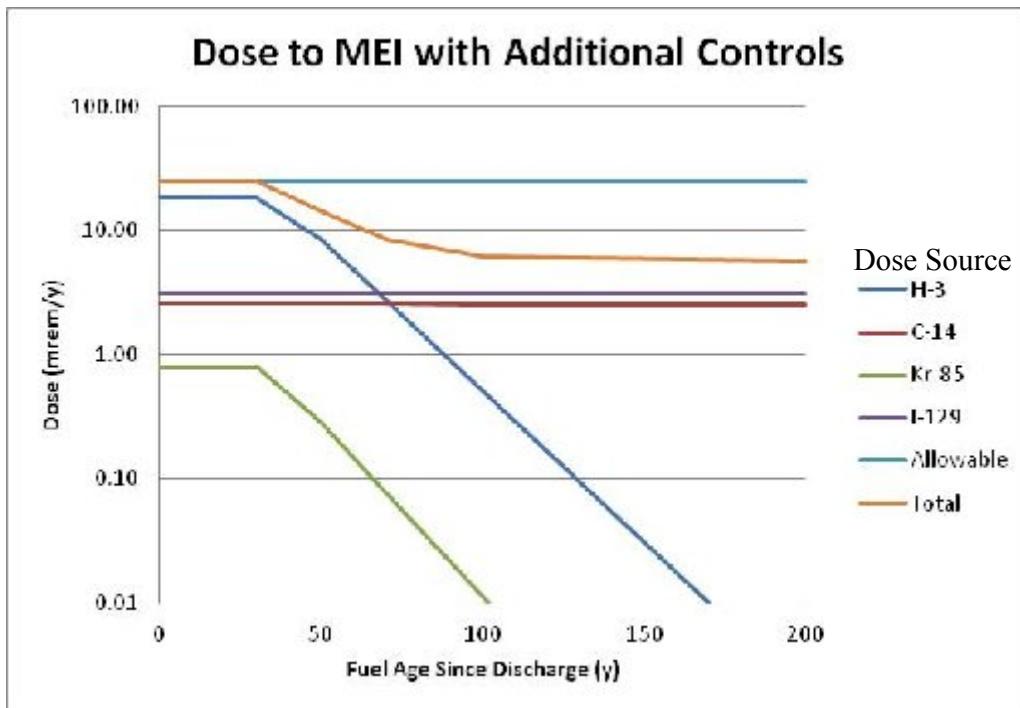


Fig. 5. Dose to MEI total and dose contribution from each of the volatile radionuclides with fuel age for a 1000-t/y recycle plant processing 30-GWd/tHM PWR UOX fuel. Implementing controls required by 40 CFR 190 and additional controls needed to meet an allowable dose limit of 25 mrem.

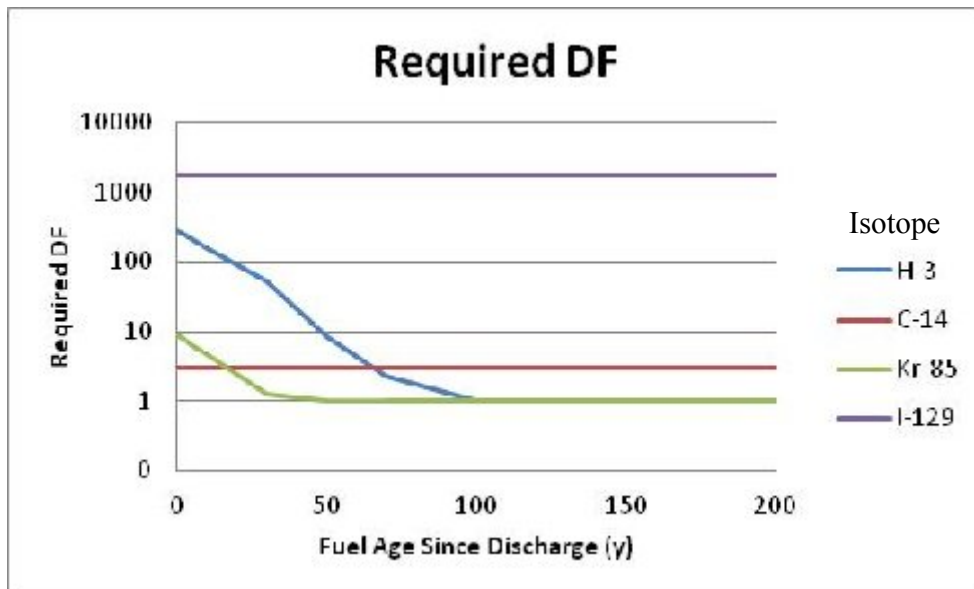


Fig. 6. Required DFs for each of the volatile radionuclides with fuel age for a 1000-t/y recycle plant processing 30-GWd/tHM PWR UOX fuel. Implementing controls required by 40 CFR 190 and additional controls needed to meet an allowable dose limit of 2.5 mrem.

**Step 6. Calculate thyroid dose.** In addition to meeting the whole body dose, there are limits to specific organs. The dose limit to the thyroid is specified in 40 CFR 190 as 75 mrem/y. Applying the same DFs previously used to meet the 40 CFR 190 discharge limits for only iodine and krypton reduces the thyroid dose to 129 mrem/y for the processing of 2-y-cooled fuel, and to 65.6 mrem/y for the processing of 100-y-cooled fuel. The two primary contributors to the dose are  $^3\text{H}$  and  $^{129}\text{I}$  in the case of short cooling times.

Applying the same additional DF requirements as imposed to meet the whole body dose limit of 25 mrem/y, the thyroid dose calculations show that dose to the thyroid is reduced to 74.8 mrem/y. Thus, no additional DF requirements would be needed beyond those needed to meet the 25-mrem/y whole body dose limit.

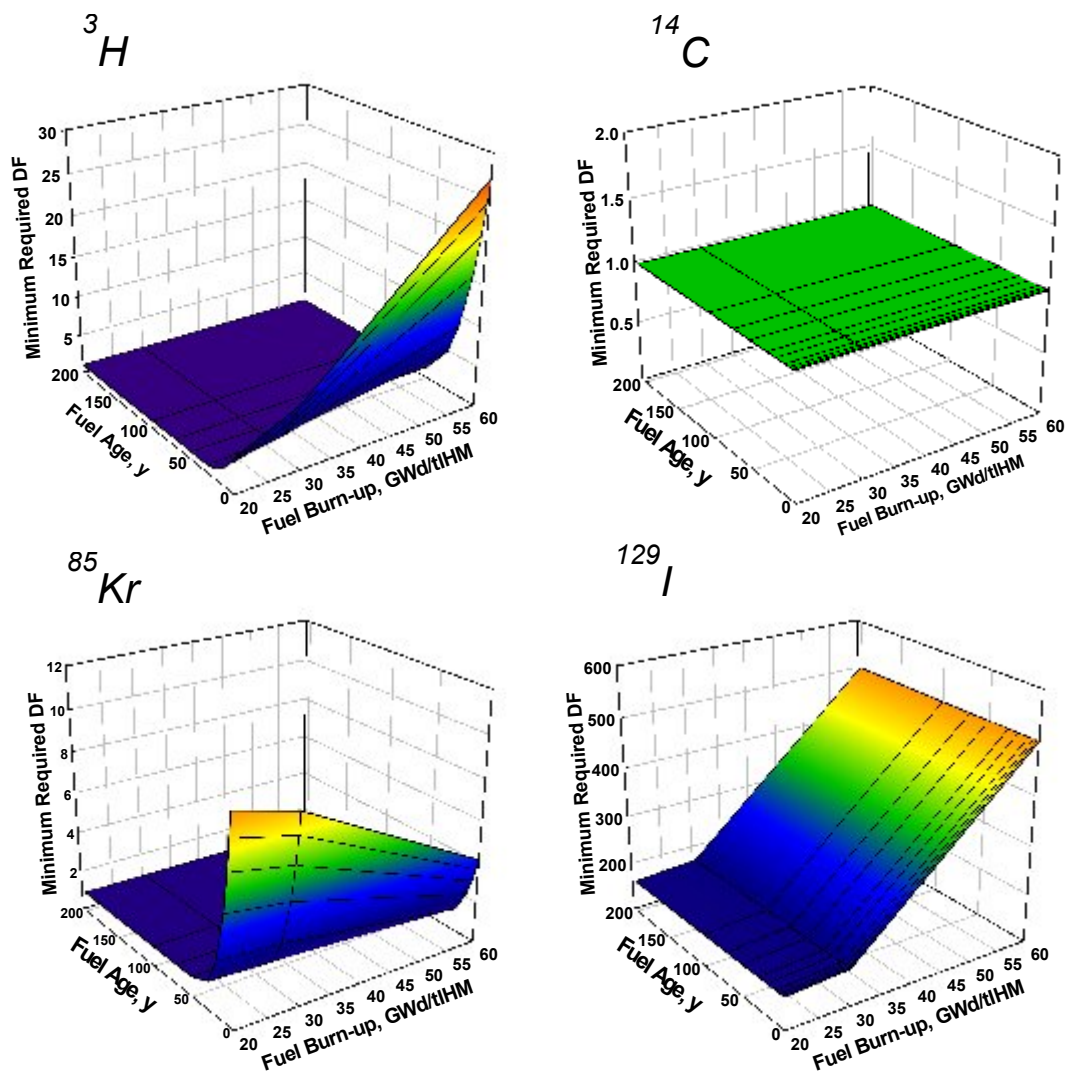
If the same apportionment of 10% is made for the thyroid dose from the four volatile radionuclides as for the whole body dose and if the same additional controls are applied, the resulting thyroid dose ranges from is 7.78–7.38 mrem/y. Iodine-129 accounts for approximately ~80% of this dose, with  $^{14}\text{C}$  the major contributor to the remaining dose. A slight increase in the  $^{129}\text{I}$  DF from ~1750 to 1850 is required to reduce the thyroid dose to <7.5 mrem/y, holding the DFs for the other isotopes as they were.

We also looked at several dose allocation cases to calculate a range of possible DFs. Allowing all of the allowable dose to be used by the volatile radionuclides is one bounding case. Subsets of this bounding case that were evaluated for select cases were (a) allowing the allowable dose to be partitioned somewhat evenly between the four volatile radionuclides and (b) allowing each volatile radionuclide to have up to 97% of the allowable dose, while the other three radionuclides were allocated only 1% of the allowable dose.

A summary of the data for the maximum DF values required to meet regulations is presented in Table II. Figure 7 shows the minimum required DF values for  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ , and  $^{129}\text{I}$ , respectively. These figures were based on the reference PWR UOX cases evaluated assuming that the full dose allocation was assigned to the four volatile radionuclides. The DF values shown in Table II and Fig. 7 contained no engineering margin and could vary by a factor of 2 or more if different site-specific or reprocessing-facility-specific assumptions were used. In terms of establishing practicable target DF values for the capture processes under development, we believed that with a reasonable engineering margin these should be on the order of three to ten times the values that result from a 25% to 50% dose allocation, that is, values that fall between those shown in the two major columns of Table II, recognizing that if the fuel is cooled long enough, then  $^{85}\text{Kr}$  or  $^3\text{H}$  control should be significantly reduced. These DF values were within the range of DFs that are reported for the capture technologies available for the volatile radionuclides [11]. Achieving the required iodine and tritium DFs will be more challenging. While it can be argued that the arbitrary 10% dose allocation may not be appropriate or optimal, the full dose allocation was certainly too optimistic because other radionuclides will contribute to the total dose. The anticipated range was expected to be from 5% to 50%. Further analysis into how potential emissions of other radionuclides in used fuel can be controlled would have been required to refine this value.

**Table II. Maximum DFs required for the volatile radionuclides of concern in this report**

	Full dose allocation to volatile radionuclides			10% dose allocation to volatile radionuclides		
	PWR/UOX	PWR/MOX	AHTGR	PWR/UOX	PWR/MOX	AHTGR
$^3\text{H}$	25 (1 after 57 y)	160 (1 after 90 y)	42 (1 after 66 y)	600 (1 after 114 y)	720 (1 after 117 y)	585 (1 after 113 y)
$^{14}\text{C}$	1	1	4	10	15	30
$^{85}\text{Kr}$	9 (1 after 34 y)	4.2 (1 after 22 y)	9.2 (1 after 34 y)	18 (1 after 45 y)	13 (1 after 40 y)	62 (1 after 64 y)
$^{129}\text{I}$	380	630	650	3800	8000	6550



**Fig. 7. Minimum DFs required for  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ , and  $^{129}\text{I}$  as a function of fuel age and burnup for a 1000-t/y recycle plant processing PWR UOX fuel.**

## SENSITIVITY STUDIES

The initial sensitivity studies showed that several reprocessing facility design, operation, and location parameters can cause variations in the DF values calculated in this analysis.

- Increasing stack height by a factor of 4 (from 37 m to 150 m) results in reducing the dose to the MEI to 1/16 of its original value.
- Changing the speciation of the  $^{129}\text{I}$  from 100% particulate form to 30% to 70%, a range consistent with measurements from the Karlsruhe Reprocessing Plant, would reduce the  $^{129}\text{I}$  dose to 30% to 70% of the original value.
- Changing the stack gas velocity and temperature can affect the dose to the MEI by up to a factor of 2, depending on if the plume was momentum or buoyancy dominated, and if the stack gas velocity was varied by changing the stack diameter or the stack gas flow rate.
- Site-specific meteorological parameters can (a) cause the  $^3\text{H}$  dose to vary by a factor of 2 depending on the humidity, (b) increase the dose from  $^{14}\text{C}$  and  $^{129}\text{I}$  by more than 10% in areas with higher precipitation, and (c) cause the dose from any of the volatile radionuclides to vary by more than 20% for different wind velocities.
- Site-specific agricultural parameters, which vary for different locations in the United States, affect dose from ingestion. Changing from the rural food scenario used in the calculations in this paper to an urban food scenario decreased the doses from those volatile radionuclides that play a role in the food cycle ( $^3\text{H}$ ,  $^{14}\text{C}$ , and  $^{129}\text{I}$ ) by 30–40%. The dose from  $^{85}\text{Kr}$  was unchanged because it does not play a role in the food cycle.

## SUMMARY AND CONCLUSIONS

It was clear from this study that it was possible to decrease the DF for one volatile radionuclide and increase the DF for one of the others while still maintaining the same dose to the MEI. With this in mind, it was not possible to arrive at a single target DF for one volatile radionuclide without taking into account the DFs for the other volatile radionuclides. The DF values required to limit the dose to the MEI were a function of the fuel age at the time of processing.

Allowing the fuel to age before reprocessing allowed a decrease in the volatile radionuclide inventory, but only for the relatively short-lived isotopes  $^3\text{H}$  ( $t_{1/2} = 12.32$  y) and  $^{85}\text{Kr}$  ( $t_{1/2} = 10.76$  y). However, for fuel aging to be an effective method of limiting the release of the volatile radionuclides to the point where little or no control is required, storage times on the order of 30 to 100 y were needed. We calculated the required DF values and doses to the MEI for storage times extending to 200 y so that the user of the calculations provided here can assess this strategy. The cost benefit of long storage of fuel as a strategy for reducing reprocessing costs by reducing or eliminating the need to control emissions of short-lived volatile radionuclides must be evaluated. There may be other impacts of long storage that should be identified and evaluated, such as the value of the recycled fuel. Such a cost-benefit analysis was outside the scope of the work presented in this paper.

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