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Estimated Radionuclide Releases and Collective Doses from the Rokkasho Reprocessing Facility

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Executive Summary

Japan Nuclear Fuel Ltd (JNFL) has indicated it intends to commence operations at the nuclear fuel reprocessing facility at Rokkasho during 2008. If JNFL ever succeeds in putting the plant into full operation, Rokkasho will result in extensive nuclide discharges with significant implications for radiation exposures. These discharges are greater per tonne of reprocessed fuel, than those from French or UK nuclear reprocessing plants.

This report examines official predictions of releases from Rokkasho by comparing them with three yardsticks. First, normalised nuclide releases from the La Hague reprocessing facility in France; second, normalised Kr-85 releases from the Karlsruhe plant in Germany; and third, estimated nuclide releases from Swiss PWR fuel stored for 12 years. The report indicates that, for some nuclides, official release estimates are inconsistent with these yardsticks and therefore may be too low. This is particularly the case with official estimates for liquid iodine-129 releases.

The report also calculates global collective doses for four radionuclides released from Rokkasho which are circulated throughout the world. These are hydrogen-3 (tritium), carbon-14, krypton-85, and iodine-129. Global collective doses from these nuclides are higher than those from French or UK reprocessing plants. Indeed the calculated global doses from Rokkasho releases are likely to be similar in magnitude to the collective dose from the Chernobyl disaster in 1986.

Introduction

1. The Rokkasho reprocessing facility in Japan has been under construction for a number of decades, and it is expected that attempts will be made during 2008 to commence commercial operations. The reprocessing of spent nuclear fuel results in very large releases of long-lived radionuclides. In the past, much concern has been expressed about the large scale of emissions from reprocessing facilities, especially krypton-85 emissions (see for example Mellinger et al, 1984). Therefore this report examines official estimates for future discharges from the Rokkasho plant. These concerns centre on the fact that a number of the released nuclides are exceedingly mobile and have long half-lives, therefore they are distributed throughout the world (IAEA, 1985). These nuclides[1] - H-3, C-14, Kr-85 and I-129 are examined in Appendix A. Exposures to these nuclides will result in **individual** doses to members of critical groups near Rokkasho, and in highly significant **collective** doses to the world's population. This report concentrates on the latter doses.

Nuclide Discharges from Reprocessing

2. Table 1 and 2 below compare emissions/discharges from La Hague in 1999 (a representative year) with expected annual releases at Rokkasho.

Table 1. Annual Releases from Reprocessing Plants – TBq

Nuclide		La Hague Releases in 1999	Estimated future annual Rokkasho Releases

[1] Few data exist for the source terms of Cl-36 and Tc-99 in spent fuel and their subsequent environmental transport and they are not considered further in this report except where data is available. However it is expected that their collective doses would be considerable.

H-3	Aerial	80	1,900
	Liquid	12,900	18,000
C-14	Aerial	19	52
	Liquid	10	-
Kr-85	Aerial	300,000	330,000
I-129	Aerial	0.0074	0.011
	Liquid	1.83	0.043
I-131	Aerial	-	0.017
	Liquid	10	0.17

Data sources

For France: derived from STOA Report – Fairlie, Schneider et al (2001)

For Japan: “Application for the Permit to Modify the Reprocessing Installation" submitted by JNFL to Japan’s Prime Minister in 2001.

Table 2 Nuclide Discharges normalized per tonne of fuel reprocessed - TBq/tonne

Nuclide		La Hague 1999 (divided by 1560 tonnes)	Rokkasho (divided by 800 tonnes)	Rokkasho / La Hague
H-3	Aerial	5.1 E-2	2.4 E+0	X 47
	Liquid	8.3 E+0	2.3 E+1	X 2.7
C-14	Aerial + Liquid	1.8 E-2	6.5 E-2	X 3.6
Kr-85	Aerial	1.9 E+2	4.1 E+2	X 2.2
I-129	Aerial	4.7 E-6	1.4 E-5	X 3.0
	Liquid	1.2 E-3	5.4 E-5	X 0.045
I-131	Aerial	-	2.1 E-5	-
	Liquid	6.4 E-3	2.1 E-4	X 0.033

Comments on Estimated Discharges from Rokkasho

3. Overall, these normalised discharges appear to indicate that both fission products (Kr-85 and I-129) and activation products (H-3 and C-14) in Rokkasho fuel will increase by factors of 2 to 3 compared to 1999 La Hague fuel. These increases appear reasonable in view of the increased burnups in the fuels expected to be reprocessed (see below), but there are some anomalies - highlighted in red in table 2. Therefore, the estimated discharges from Rokkasho cited in table 1 require further discussion. In particular,

- the normalised figure for I-129 liquid discharges (5.4 E-5 TBq per tonne) appears to be a factor of ~20 times lower than La Hague. If the Rokkasho estimate were correct, one explanation would be an iodine precipitation stage at Rokkasho, using silver as iodine precipitant. At present, we are unaware of any such process at Rokkasho, however it is understood that the Tokai reprocessing facility does have an iodine precipitant stage. In addition, the similar predicted reductions for I-131 as for I-129 at Rokkasho would seem to indicate the existence of an iodine reduction stage.
- the normalised figure for estimated tritium aerial emissions at Rokkasho (2.4 TBq per tonne) is ~50 times higher than that at La Hague. The reason for this discrepancy is not known.

High Burnup Uranium Fuel and MOX (ie reprocessed Plutonium) Fuel

4. It is necessary to compare high burnup uranium (U) fuels and MOX (ie reprocessed Pu) fuel because they have similar aims of extracting more energy from the fissions of fissile material. High burnup nuclear fuels are increasingly used throughout the world because they extract more heat energy per tonne of fuel than low burnup fuels. High burnup U fuels use slightly higher enriched uranium than was used in the past, eg 4% to 5% U-235 compared to 2% to 3% in the fuel used in the 1980s and 1990s. Higher burnup fuel was introduced by US utilities in the late 1970s when burnups of 30 GW days/tonne were the norm, and nowadays burnups of up to 50 to 70 GW days/tonne are common in US reactors.

5. The use of higher burnup fuel permits fuel to be left in reactors for longer periods and for it to be “burnt” at higher energy output rates compared to ordinary U fuel. This results in greater operational flexibility, higher fuel productivity and considerable economies for nuclear utilities. High burnup fuel is now used by almost all nuclear utilities throughout the world, except those in countries tied to reprocessing. Even in those countries, utilities are turning to higher burnup fuels because of the significant economies associated with their use. This trend has adverse implications for the continued need for reprocessing, as it is clear that high burnup U fuel is much more economic than plutonium-enriched fuel.

6. As stated above, high burnup U fuel and MOX Pu fuels are similar in their aims of extracting energy from the fission of fissile material. MOX fuel use was initially justified as a method of extracting energy from fissile Pu in spent U fuel, but high burnup U fuel achieves exactly the same aim more economically without the need to extract and reprocess spent fuel. For example, high burnup U fuel in French reactors has consistently achieved higher burnups (44 GW days/tonne) than MOX fuel (36 GW days/tonne) according to the former Cogema executive director, Deroubaix (1999). MOX fuel burnups are restricted to ~36 GW days/tonne by French nuclear regulators because of the reduced safety tolerances of MOX fuel during ramping up and down.

7. The 22% greater electrical energy per tonne obtained from high burnup U fuel represents a large difference in operating economics. It means that EdF suffers a significant financial penalty from using MOX fuel instead of high burnup U fuel (the same is understood to be the case in the declining number of German utilities which continue to use MOX fuel). Because of the need to reduce electricity generating costs due to the liberalisation of the European electricity market, most European utilities now use high burnup U fuel, even in reactors previously reserved for MOX. In France, EdF uses MOX fuel in ten older (900 MW) reactors, but uses high burnup U fuel in its newer reactors despite considerable pressure from Areva to use MOX fuel in them. From the above considerations, it can be seen that MOX fuel is an unattractive policy option for utilities. It is difficult to avoid the conclusion that the main function of MOX burning by EdF is to provide Areva with an excuse for continued reprocessing.

8. For the above reasons, in the UK, the British Government has very recently announced (DBERR, 2008) that future fuel arising from any new nuclear reactors will not be reprocessed. Page 116 of its 2008 White Paper concludes

“Having reviewed the arguments and evidence put forward, and in the absence of any proposals from industry, the Government has concluded that any new nuclear power stations that might be built in the UK should proceed on the basis that spent fuel will not be reprocessed and that plans for, and financing of, waste management should proceed on this basis. We are not currently expecting any proposals to reprocess spent fuel from new nuclear power stations. Should such proposals come forward in the future, they would need to be considered on their merits at the time and the Government would expect to consult on them."

Impact of Increasing Burnups on Nuclide Inventories in Fuel

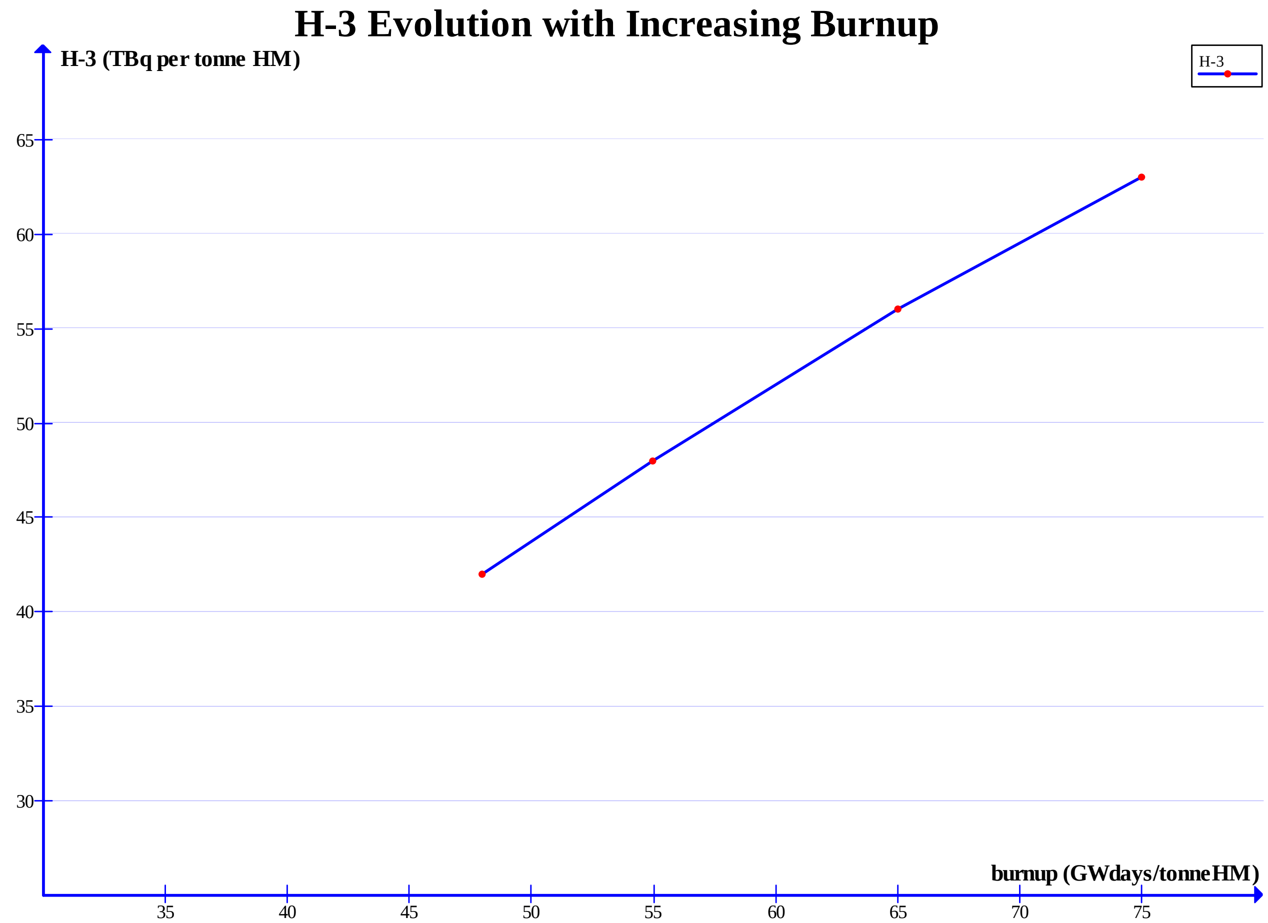
9. Although high burnup fuels offer considerable advantages to nuclear utilities, they also result in higher nuclide inventories in spent fuel. Table 3 and graphs A to E below indicate that the nuclide concentrations in spent PWR fuel increase linearly with burnup. The data in the table and the graphs were obtained from the estimates provided in McGinnes (2002) for 12 year old spent Swiss PWR fuel. The estimates in his report were derived from nuclide inventory calculations derived from the BOXER computer model (Parotte et al, 1996) and the similar ORIGEN-2 model code (Croff, 1980).

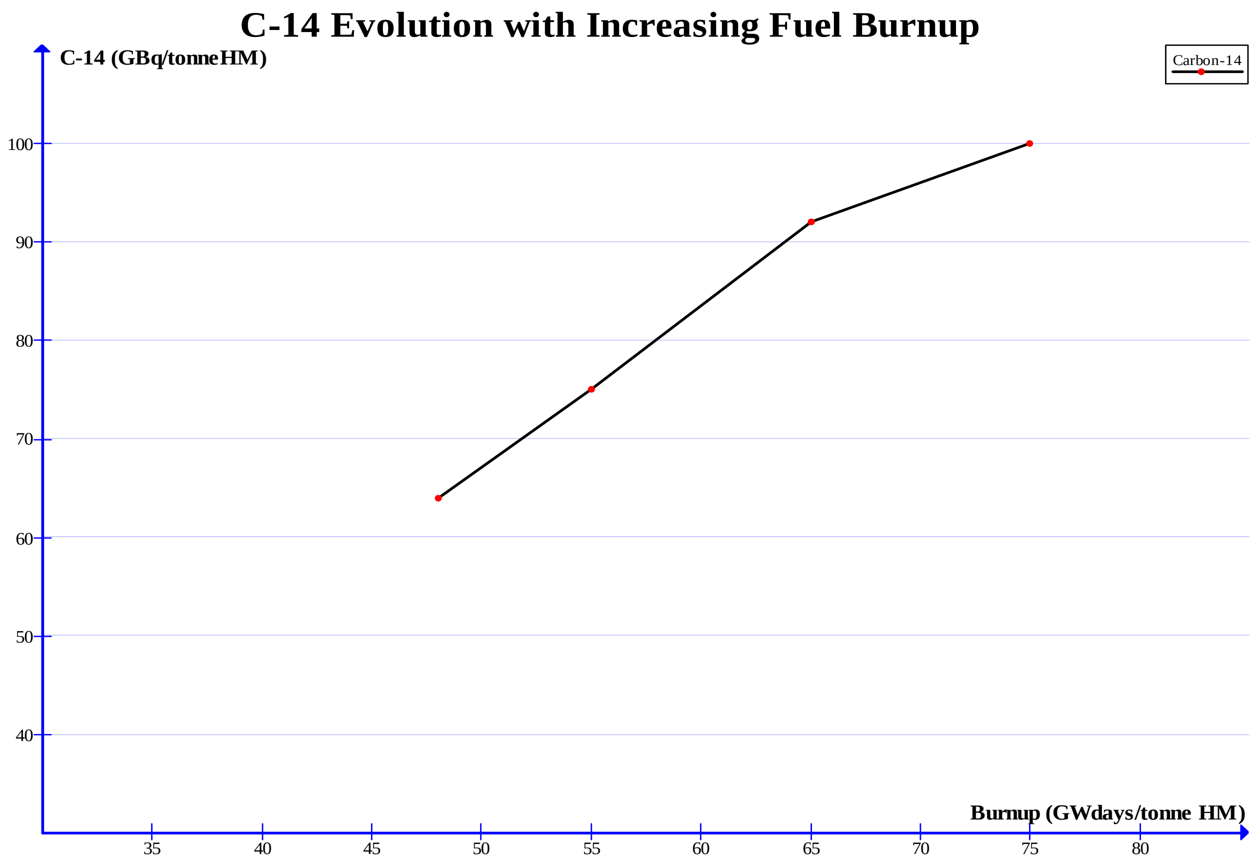
Table 3. Nuclide inventories in PWR fuel at time of exit from reactor at selected burnups Bq/tHM (becquerels per tonne of heavy metal) data from McGinnes (2002)

	48 GWd/ tHM	**55 GWd/ tHM**	**65 GWd/ tHM**	**75 GWd/ tHM**
Nuclide	Bq/tHM	Bq/tHM	Bq/tHM	Bq/tHM
H-3	4.2E+13	4.8E+13	5.6E+13	6.3E+13
C-14	6.4E+10	7.5E+10	9.2E+10	1.0E+11
Cl-36	1.1E+09	1.3E+09	1.6E+09	1.7E+09
Kr-85 (corrected)	4.6 E+14	5.0 E+14	5.6 E+14	6.2 E+14
I-129	1.7E+09	1.9E+09	2.3E+09	2.6E+09

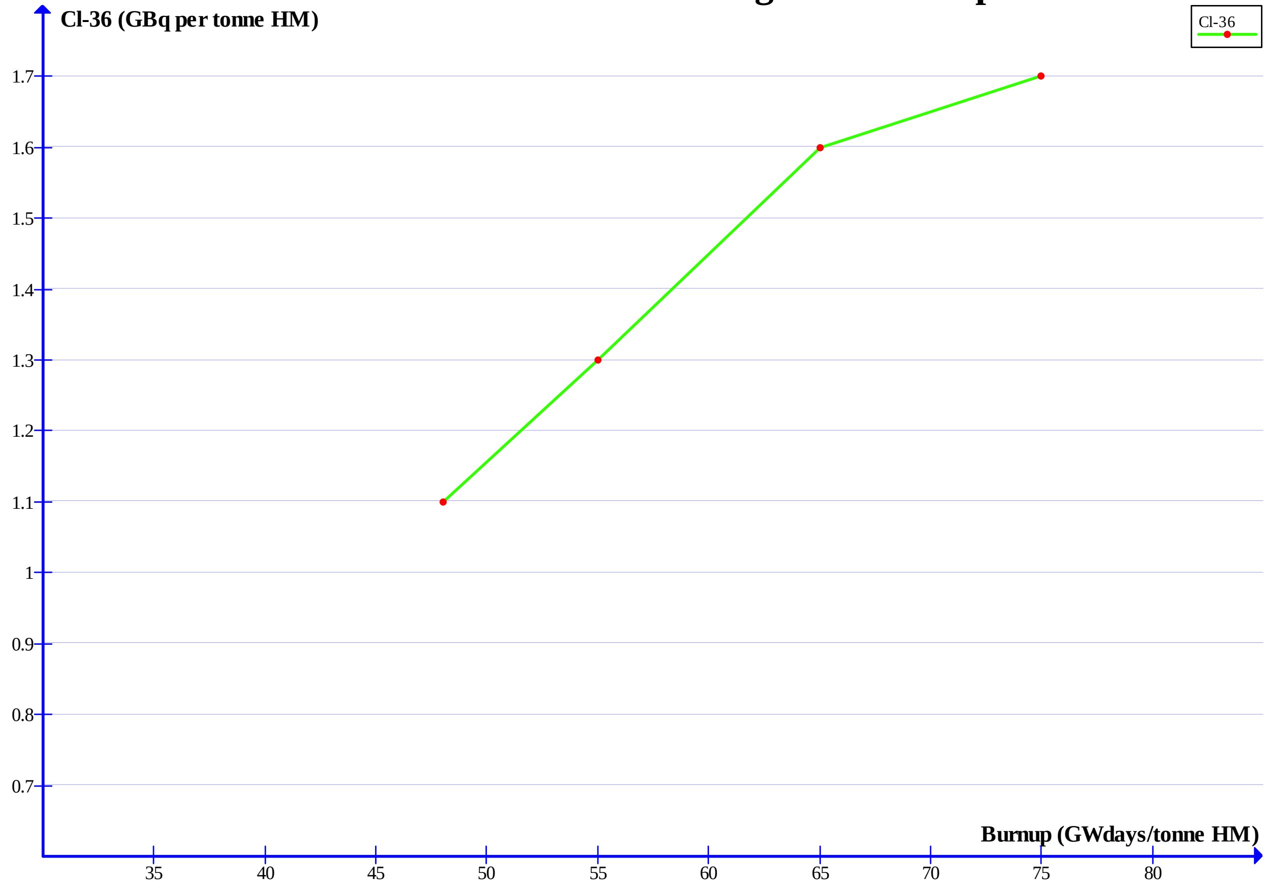
10. Following discussions with German academics, the McGinnes data for Kr-85 have been corrected by dividing by his data by a factor of 5. The reason for this is as follows. McGinnes' data includes the fission yield of the metastable isomere krypton-85m as if all of it decayed to Kr-85. However this is incorrect: only 20% of Kr-85m decays to Kr-85: the remaining 80% decays to stable (ie non-radioactive) Rb-85. This means that McGinnes' data for Kr-85 is too large by a factor of 5: his data have been corrected accordingly in tables 3, 4 and graph D.

Graphs A, B, C, D and E (below)

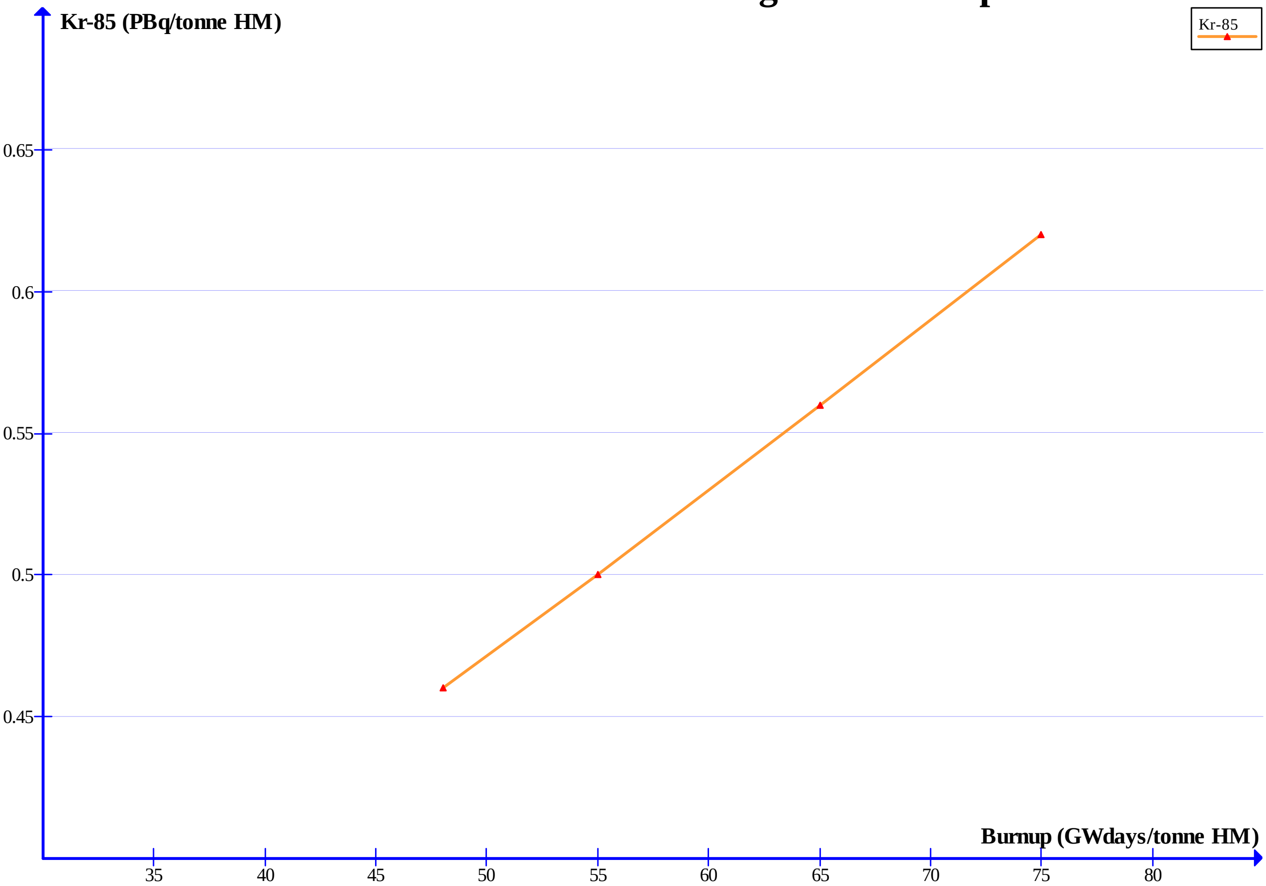

H-3 Evolution with Increasing Burnup
H-3 (TBq per tonne HM)
H-3
burnup (GWdays/tonne HM)
65
60
55
50
45
40
35
30
35
40
45
50
55
60
65
70
75

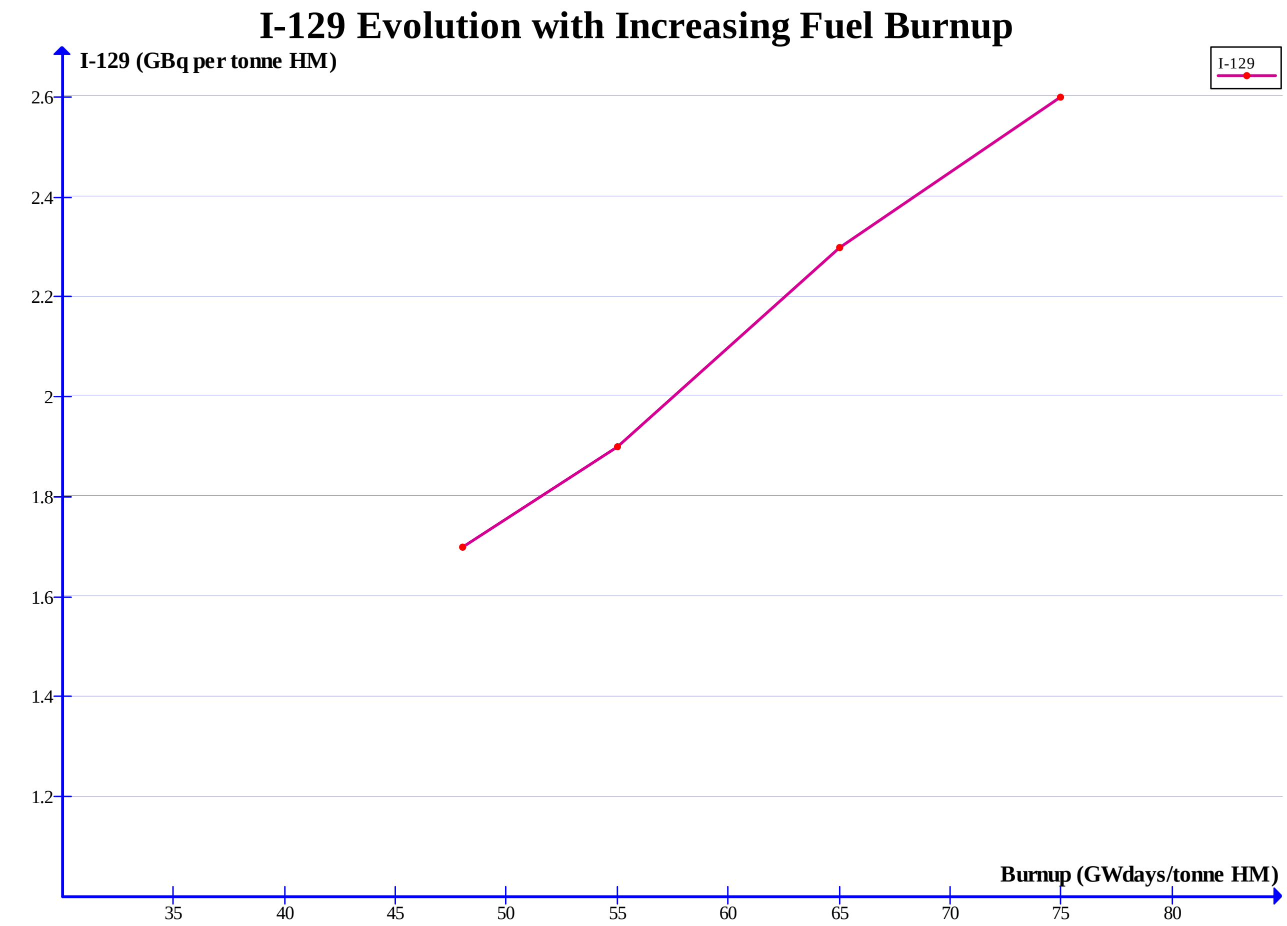

C-14 Evolution with Increasing Fuel Burnup
C-14 (GBq/tonne HM)
Carbon-14
Burnup (GWdays/tonne HM)
100
90
80
70
60
50
40
35
40
45
50
55
60
65
70
75
80

Cl-36 Evolution with Increasing Fuel Burnup



Kr-85 Evolution with Increasing Fuel Burnup





11. It can be seen from the above graphs that the various nuclide concentrations in spent fuel increase approximately linearly with increasing burnup. The above nuclides are almost[2] all released during reprocessing and not during reactor operations, therefore it is possible to make comparisons between the above nuclide concentrations in spent fuel and the amounts discharged as seen in tables 1 and 2.

12. However a number of points must be borne in mind. According to Winger et al (2005), the amount of activity released during reprocessing depends, inter alia, on

a. the type of fuel reprocessed,
b. the irradiation history (ie burnup) of the fuel and
c. the number of years spent in cooling ponds and dry stores before reprocessing

13. Briefly, (a) PWR fuel comprises about half of Japanese fuel, and the rest (BWR fuel) is not significantly different from PWR fuel as regards nuclide generation rates. (b) is dealt with in the above graphs, leaving (c) to be considered. An allowance must be made for nuclide decay during spent fuel storage times of 6 to 20 years being planned by Japanese nuclear utilities. (See appendix B which indicates that the cooling time for most fuel is 6 to 20 years.) This can be taken into account by arbitrarily assuming an average cooling time of 11 years, which is approximately the half-life of Kr-85 (10.7 years) and tritium (12.3 years). During one

[2] A small percentage (<1%) of Kr-85 is understood to escape from nuclear reactors during routine operations. This is neglected in this report, since most estimates here are only made to one or two significant figures.

half-life, one half of the original amount of the relevant nuclide will decay. The use of an 11 year figure will not result in exact comparisons because we do not have the exact fuel cooling times which will be used by Japanese utilities. Nevertheless its use will give us an approximate idea of the expected levels of nuclide emissions, bearing in mind that shorter cooling periods will result in higher emissions and vice versa.

14. Table 4 below compares the McGinnes data for fuel with a burnup of 48 GWdays/tonne (cooled for 11 years), the La Hague normalized emissions, and the predicted Rokkasho emissions. The data for 48 GWdays/tonne are used as this is close to the expected burnup of the fuels to be reprocessed at Rokkasho

Table 4. Comparison of normalized Rokkasho nuclide evolution rates (C) (from table 2) with (A) La Hague 1999 data and with (B) McGinnes (2002) data for burnup of 48GWdays/tonne HM (cooled for 11 years)

Nuclide	(A) La Hague in 1999 (gaseous and liquid emissions added)	(B) McGinnes data for PWR fuel of 48 GWdays/tonne HM burnup (cooled for 11 years)	(C) Rokkasho total (gaseous and liquid emissions added)
H-3	8.3 TBq/tonne	~25 TBq/tonne	25 TBq/tonne
C-14	18 GBq/tonne	64 GBq/tonne	65 GBq/tonne
Kr-85	190 TBq/tonne	~230 TBq/tonne (corrected)	410 TBq/tonne
I-129	1.2 GBq/tonne	1.7 GBq/tonne	0.068 GBq/tonne

15. It can be seen from table 4 that while good agreement exists for H-3 and C-14 nuclide evolution rates, a wide discrepancy exists for I-129: the Rokkasho I-129 figure is about 25 times too low. This may be due to the reduction of iodine releases as a result of a silver precipitation stage at Rokkasho, although a 25 fold reduction is very efficient and may be overly optimistic. JNFL could be asked to compare this rate with the efficiency rate of the silver precipitation stage at the Tokai reprocessing plant.

16. The official Kr-85 estimate of 410 TBq per tonne of reprocessed fuel from Rokkasho needs more explanation as well. In order to understand this, we have examined a report by Kalinowski et al (2004) on Kr-85 emissions from the Karlsruhe reprocessing plant in Germany between 1985 and 1988. From the Kr-85 discharge data from reprocessing spent PWR fuel from the Stade, Neckarwestheim 1 and Obrigheim reactors, the average Kr-85 emissions ranged between 260 and 330 TBq per tonne of PWR fuel. This would be for relatively low burnup fuel, ie ~30 GWdays/tonne, cooled for at least 6 years. This is not dissimilar from the Rokkasho estimate of 410 TBq per tonne, and it means that the official Rokkasho estimate is probably correct as it arises from much higher burnup fuel than used in German reactors in the 1980s.

Collective Doses from Reprocessing

17. The European Commission has constructed models which estimate collective doses to UK, Europe and world populations from the nuclides H-3, C-14, and I-129 (Simmonds et al, 1996). A rudimentary model also exists for Kr-85, which is assumed to remain a gas and not to interact with the biosphere, lithosphere and hydrosphere. Unfortunately no models yet exist for the global transport of Cl-36 and Tc-99. However the collective dose from Cl-36 is

expected to be considerable given its long half-life of 300,000 years: this is a subject for future research. Although no global model has yet been constructed for Tc-99 (whose half-life is also very long - 211,000 years), preliminary estimates by Fairlie and Sumner [2001] indicate that the global untruncated dose coefficient of Tc-99 is about 10 person Sv per TBq. Tc-99 releases from Rokkasho have not been estimated but may be relatively low, as Tc-99 tends to be generated in Magnox rather than in LWR fuel.

18. Untruncated collective doses to the world's population of 6 billion persons have been calculated using the above models and the results are presented here. However users of this information need to be aware of a number of points, as follows.

- Country and regional dose models are specific to their areas; but, global dose models apply to all areas, ie they do not depend on where the discharge takes place.
- The practice of truncating doses after 500 years which is followed by some authors is considered impractical given the much longer half-lives of most of the nuclides under consideration, including C-14 (5,730 years) and I-129 (16 million years) – see Fairlie and Sumner (2001). Therefore collective doses are not truncated in this report.
- For shorter-lived nuclides, such as Kr-85 and H-3 (half-lives of 10.7 and 12.3 years respectively) more than 95% of their global collective doses will be delivered within the first 50 years after discharge, mostly within the first decade.

19. Carbon-14, iodine-129, krypton-85 and hydrogen-3 (tritium) are described in the Appendix to this report, together with some description of their releases from the reprocessing plants at Sellafield, UK and La Hague, France.

20. Tables 5, 6 and 7 below compare the collective doses from releases at Sellafield and La Hague in 1999 (a representative year) with those from expected annual releases at Rokkasho. Source: "Reprocessing Plant - Application for the Permit to Modify the Reprocessing Installation" submitted by JNFL to Japan's Prime Minister in 2001".

Table 5. Estimated Global Collective doses from 1999 discharges – Sellafield

Nuclide		Global Dose Coefficient Person Sv/TBq	Sellafield Releases - TBq	Global Dose Person Sv
C-14	Aerial	115	2.9	330
	Liquid	115	5.8	670
I-129	Aerial	9454	0.0025	24
	Liquid	690	0.48	330
Kr-85	Aerial	0.004	100,000	400
H-3	Aerial	0.002	622	1.2
	Liquid	0.00004	1,800	0.07
TOTAL				**1,800**

Source: Schneider, Fairlie et al (2001)

Table 6. Global Collective doses from 1999 discharges - La Hague

Nuclide		Global Dose Coefficient Person Sv/TBq	La Hague Releases -TBq	Global Dose Person Sv
C-14	Aerial	115	19	2180
	Liquid	115	10	1150

I-129	Aerial	9454	0.0074	70
	Liquid	690	1.83	1260
Kr-85	Aerial	0.004	300000	1200
H-3	Aerial	0.002	80	0.16
	Liquid	0.00004	12,900	0.52
TOTAL				**5,900**

Source: Schneider, Fairlie et al (2001)

Table 7. Anticipated Global Collective doses from estimated annual discharges – Rokkasho

Nuclide		Global Dose Coefficient Person Sv/TBq	Estimated Rokkasho Releases -TBq	Global Dose - Person Sv
C-14	Aerial	115	52	5,980
	Liquid	115	-	0
I-129	Aerial	9454	0.011	104
	Liquid	690	0.043	30
Kr-85	Aerial	0.004	330,000	1,320
H-3	Aerial	0.002	1,900	3.8
	Liquid	0.00004	18,000	0.72
TOTAL				**7,400**

Source: Reprocessing Plant - Application for the Permit to Modify the Reprocessing Installation" submitted by JNFL to Japan's Prime Minister in 2001.
Global dose conversion factors from Simmonds et al (1996) and Mayall et al (1993).

21. It can be seen from these tables that the anticipated annual collective dose from Rokkasho discharges is about 4 times greater than from Sellafield and about 1/3rd greater than from La Hague. Table 8 below compares the three reprocessing plants on a fuel tonnage basis and indicates that, based on the Rokkasho discharge estimates made in 2001, the collective dose per tonne fuel throughput at Rokkasho will be about 7 times greater than from Sellafield, and about 2.5 times greater than from La Hague in 1999.

22. The reason for Rokkasho's greater nuclide discharges is that Rokkasho will be reprocessing fuels with higher burnups than the fuels reprocessed at the other two plants in 1999. Sellafield reprocesses mostly UK (Magnox and AGR) fuels which have low burnups (~6 and ~20 GWdays/tonne respectively). In France, most of the older 900 MW French PWR reactors are restricted to low (<30 GWdays/tonne) burnups for safety reasons[3]. By contrast, Rokkasho will be expected to reprocess fuels currently used by Japanese utilities with higher burnups of 45 to 55 GWdays/tonne or more.

Table 8 Collective doses per tonne of fuel reprocessed

Reprocessing Plant	Fuel Reprocessed Tonnes per year	Global Collective Dose (Person Sv)	Collective Dose per tonne reprocessed Person Sv per tonne
Sellafield	1,380 (in 1999)	1,800	1.3
La Hague	1,560 (in 1999)	5,900	3.8
Rokkasho	800 (estimated)	7,400	9.2

[3] mainly because these reactors need to be ramped up and down to follow diurnal electricity loads.

Commentary on Collective Doses from Rokkasho

23. Using the figure 7,400 person Sv, if one were to apply the ICRP fatal cancer risk coefficient of 5% per Sv to Rokkasho releases, the result would be about 370 fatal cancers each year throughout the world from Rokkasho releases each year. If Rokkasho were to operate at maximum capacity for 40 years, its officially predicted life, the cumulative collective dose over this period would be 296,000 person Sv (see table 8 below) resulting in about 15,000 future cancer deaths throughout the world.

24. The collective doses from tritium and krypton would be mainly delivered in the first 50 years after discharge, but the doses from iodine and carbon would continue thereafter over many millennia to future generations throughout the world. Many uncertainties exist over whether these doses will actually occur in the absolute sense. Nevertheless, given the information available to us today and given currently accepted models of radiation effects[4], these are the best estimates available to us of future radiological detriment from reprocessing releases at the two sites.

25. Because of these uncertainties, collective dose estimates are usually used in a relative rather than absolute way; that is, they are used in comparisons with collective doses from other processes. For example, the collective doses from Rokkasho can be compared with the 5 person Sv reference level for annual releases from 1 GW Swedish nuclear reactors. They are seen to be extremely large when compared in this way.

26. Other comparisons are made in table 9 which ranks collective doses from other nuclear processes/accidents with predicted collective doses from 40 years' releases from Rokkasho, its predicted lifetime. It can be seen that the cumulative collective dose from predicted Rokkasho releases are very large indeed.

Table 9 Global Collective Doses from Anthropogenic Radiation Sources

Source of Exposure	Global collective dose (Person Sv)
Chernobyl Accident 1986	600,000
World Nuclear Power Production to 1989	400,000
Rokkasho Reprocessing 2008 to 2047 (estimate assuming 7,400 person Sv per year)	**296,000**
La Hague Reprocessing between 1990 and 2019 (estimate assuming 5,900 person Sv per annum)	180,000
World Radioisotope Production and Use to 1989	80,000
World Nuclear Weapons Fabrication to 1989	60,000
Sellafield Reprocessing for 30 years (1980-2009) (estimate assuming 1,800 person Sv per annum)	54,000
Kyshtym Accident USSR 1957	2,500
Windscale Incident UK 1973	2,000

[4] In particular, use of the linear no threshold theory (LNT) of radiation's effects which continues to be used by all radiation authorities throughout the world

World Underground Nuclear Testing to 1989	200
Three Mile Island Accident US 1979	40

Sources: Bennett, 1995; UNSCEAR, 1993; author estimates

Use of Collective Dose in Cost-Benefit Studies

27. To compare costs and benefits of options which have no common comparator (eg to compare the **financial costs** of new abatement facilities with the **discharges** they may save), collective doses from discharges may be converted to fatalities using the conventional ICRP radiation risk factor of 5% per person Sv, and fatalities may be converted to £, $ or € values using monetary values per statistical life. The two costs may then be compared as occurs in conventional cost benefit analyses. Conventionally, this procedure is shortened to adopting a £, $, yen or € value per person Sv saved.

28. The range of values used for a person Sv is wide and has been reported as extending from £20,000 to £100,000 per person Sv. In 1995, the European Commission used a value of $3 million in its statistical valuation of a life for the external costs of fuel cycles, equivalent to a person Sv value of £100,000 (CEC, 1995). This is close to the value of £100,000 used by BNFL to reflect "statistical risk and corporate profile" (Robb, 1990). A reasonable figure for comparison purposes is £100,000 (or 20 million yen) per person Sv. When applied to untruncated global doses from 30 years' of Sellafield and La Hague releases, the values in Table 10 are obtained.

Table 10. Maximum Values for Optimisation Measures (exchange rate £1= 200 yen)

	Total Global Dose Person Sv	Billion Yen Per Person Sv	Maximum Value: Billion Yen
Sellafield (30 years)	54,000	0.02	1,080
La Hague (30 years)	177,000	0.02	3,600
Rokkasho (40 years)	296,000	0.02	5,900

29. In optimisation studies (i.e. studies to reduce doses as low as reasonably achievable), expenditure up to the amounts in the final column should be considered for remedial or abatement measures to reduce doses. These are extremely large sums: the amounts that could be spent on abatement measures comfortably exceeded annual operating profits at La Hague and Sellafield, and would most likely do so at Rokkasho.

Conclusions

30. This report has examined official Japanese estimates for nuclide emission rates at Rokkasho. It has estimated normalised emission rates for H-3, C-14, Kr-85 and I-129 for Rokkasho, and has revealed inconsistencies when compared with French operating data for reprocessing discharges from La Hague and with estimated nuclide evolution data for Swiss nuclear fuel. It is recommended that information should be provided by Japanese authorities on how they calculated their estimates of nuclide emissions from Rokkasho, in particular what fuel burnups and fuel cooling times were used to derive their estimates. Also what process (if any) is to be used to reduce iodine-129 discharges, and what is the respective efficiency of this process.

31. This report has also derived global untruncated collective doses from annual reprocessing releases at Sellafield and La Hague, and from expected maximum releases at Rokkasho. In order to assess the significance of these doses, this report has compared these doses with collective doses from other nuclear-related activities and has valued them using conventional cost benefit analyses.

35. Global untruncated collective doses from expected reprocessing releases at Rokkasho and actual releases at other reprocessing facilities are very large relative to other nuclear operations. When valued in monetary terms, the collective dose valuations are also very large and easily exceed operating profits of the plants concerned. From these considerations, the operation of these plants does not fulfill conventional expectations of commercial or radiological justification one of the ICRP's principles of radiological protection (ICRP, 2007). From these very large estimates of its collective doses, the proposed operation of the Rokkasho facility appears to be highly questionable. In practice, the real level of collective doses from Rokkasho will be crucially dependent on the correct operation and 100% efficiency of its iodine-129 separation processes.

Appendix A

Carbon-14

i. Carbon-14 is a radioactive isotope of carbon with a half-life of 5,730 years: it emits beta particles of maximum energy 156 keV. Carbon-14 arises in irradiated nuclear fuel from neutron activation of nitrogen (as impurity or additive) and oxygen (as UO_2). Carbon-14 is retained in spent fuel until reprocessing when it is released in gaseous and liquid discharges. Carbon-14 is produced naturally in the upper atmosphere as a result of the capture of cosmic ray neutrons by nitrogen-14. Because carbon-14 behaves in the same way as stable carbon, it is rapidly distributed among environmental compartments — stratosphere, troposphere, biosphere and surface ocean waters. Transfers between atmosphere, biosphere and surface ocean waters take place within a few years; transfer to deep ocean proceeds more slowly.

ii. Carbon is a major constituent of all life forms. All carbon-14, whether anthropogenic or naturally-occurring enters the carbon pool in biota. Because the half-life of carbon-14 is long, doses from carbon-14 introduced into the environment will be delivered to local, regional and global populations for thousands of generations. C-14 is the main contributor to collective doses from reprocessing discharges. C-14 collective doses are similar whether the C-14 is released to atmosphere or sea.

iii. Although carbon-14 discharged from reprocessing is distributed globally, there are significant local increases in concentration. For example, Begg et al (1991) have reported that carbon-14 discharged from Sellafield has resulted in an approximate doubling of current ambient concentrations in the Irish Sea. In 1998 and 1999, C-14 levels in botanic plants near La Hague as measured by OPRI and Cogema were 500 to 2,000 Bq/kg (cf natural background levels of 260 Bq/kg) Guillemette (2000) states that a level of 2000 Bq/kg in humans near La Hague corresponds to an annual dose of about 130 µSv, of which 115 µSv (90%) would be due to La Hague C-14 discharges. This is an appreciable fraction of the 0.3 mSv per year dose constraint usually applied to critical group doses.

iv. Reprocessing plants have different approaches to carbon-14 management. For example, the Rokkasho plant is designed to remove most carbon-14 to sea. However no details are available of how the Rokkasho operators intend to achieve this aim. This is an important matter, because C-14 is a potent contributor to collective dose. At Sellafield, about 27% of sea discharges of carbon-14 is removed in a caustic soda washing column and then precipitated as a solid (barium carbonate) which is then encapsulated in cement and stored in drums (BNFL,1993). Cogema currently releases all C-14 arisings from reprocessing. It has stated that C-14 abatement was not cost effective in their view (Cogema, 1999).

Krypton-85

v. Kr-85 is a fission product retained in reactor fuel until released during reprocessing. Kr-85 is a strong beta-gamma emitter with a half-life of 10.7 years. The mean energy of the beta particle is 251 keV and that of the less frequent gamma ray is 514 keV. Krypton-85 exposes people to external beta irradiation of the skin, and to uniform whole body gamma irradiation. Although the dose from a single decay of Kr-85 is small, the amounts of Kr-85 discharged are very large – indeed, the largest of the various nuclides emitted from reprocessing. Accordingly, doses from Kr-85 are appreciable. Krypton-85 distributes uniformly throughout the earth's atmosphere within a few years after release, therefore its collective doses are important.

vi. Being immersed in a radioactive gas inevitably results in radiation doses. Most of the dose from krypton comes from external exposures to its beta and gamma radiation. The dose from beta particles is a skin dose, and the gamma dose is to the whole body. In equivalent dose terms, the beta and gamma doses are about the same.

vii. As for internal doses, Krypton is an inert gas and is currently not thought to enter life processes nor be incorporated in biota, unlike I-129, C-14 and H-3. Although it is breathed in, it is presumed to be breathed out relatively quickly with a half time of about 30 seconds (Diethorn and Stockho, 1972). However it should be borne in mind that this model for krypton-85 uptake is based on intuition rather than experimental data. Some people could raise the example of radon-222 which is also an inert radioactive gas but which does result in substantial internal doses. The reason for these high doses is the extremely radioactive daughters of radon-222 which result from its decay inside the lung. On the other hand, Kr-85 decay results instead in stable rubidium. (Rubidium is an element in the alkali metals group of the periodic table, similar to potassium and caesium.)

viii. In 1989, the global inventory of krypton-85 was estimated to be about 3,300 PBq (1 PBq = 10^{15} Bq) nearly all of which was from reprocessing plants (Kollert and Butzin, 1989). Since 1989, this will have decayed to about a quarter of this figure, but a further 2,000 PBq was discharged from La Hague and 1,000 PBq from Sellafield between 1980 and 2001. Kr-85 is a significant contributor to collective dose. For example, for Sellafield's Kr-85 releases, Jackson et al (1998) have calculated that the (500 yr truncated) global collective dose from the annual doses of 0.1 to 4 person Sv over the past approx 50 years was 594 person Sv.

Separation and Storage of Kr-85

ix. Since the 1970s, many observers have expressed clear views on the need for the separation and storage of Kr-85 from reprocessing plants. For example, in 1972, Diethorn and Stockho (1972) from the US Nuclear Engineering Department of Pennsylvania State University concluded that

> "…although the dose from krypton-85 is small, …there seems to be little justification for continuing the present world-wide practice of dumping it into the atmosphere......the only solution for krypton-85 is permanent storage."

In 1975, the US NCRP (1975) concluded

> "…prudence would seem to dictate that fuel reprocessing plants be equipped with krypton-85 removal systems as soon as the technology is practicable"

At an IAEA Conference in 1977, J Couture (1977) then director of COGEMA stated as regards the then proposed UP3 reprocessing plant at La Hague in France

> "The UP3 plant will be accompanied by new equipment for the solidification and vitrification of all the liquid effluent. Moreover, studies will continue within the French Commissariat a l'Energie Atomique on the trapping of gases and, if necessary, krypton trapping units can be installed in 1985. Studies on the trapping of tritium are making good progress and studies on carbon-14 form part of the programme."

During the 1977 Windscale Inquiry on the proposed Thermal Oxide Reprocessing Plant (THORP) at Sellafield, UK, Mr Justice Parker (1978) in recommendation 3 required that

> "BNFL should devote efforts to the development of plant for the safe removal and retention of krypton 85 and, if development proves successful, should incorporate it in the proposed plant."

In 1983, US regulations limited krypton-85 releases to a maximum of 1.85 PBq per 1000 MW electricity produced, a ten-fold reduction from previous practice (NCRP, 1980).

x. However by the end of the 1990s, no such technologies had been implemented at any reprocessing facility in the world. Speaking on the experience at BNFL, Roger Coates – a BNFL director stated in 1999 (CEPN, 2000)

> "Krypton was rather different. You can do the cost-benefit analysis, dispersion modeling, and for the lifetime of Thorp, you can come up with detriment valuations of a few tens of millions of pounds. If we could have got viable Krypton removal and storage technology, then on the information that was available, it was clearly going to cost a few hundreds of millions of pounds. That makes it not viable, economically. The critical group doses were about 2 or 3 µSv, so on that basis, it was easy to make the decision, but it was even easier to make the decision, because there's no viable technology. People have operated troublesome pilot plants on small scales, on an intermittent basis, some using CFCs. If we had have installed that, the most promising technology, there would have been major discharge of CFC greenhouse gases. So it was a reasonably simple decision to make no Krypton removal."

xi. However in 2002 Dr Coates' comments were contradicted by a comprehensive analysis of noble gas removal and storage carried out for the UK Environment Agency (2002). This study investigated the feasibility of re-routing reprocessing off-gases to a cryogenic plant in order to separate and recover xenon (a stable inert gas also released during reprocessing) as a by-product, in parallel with krypton-85 abatement. The study reviewed UK and international developments in cryogenic and gas separation technology, and carried out a literature review of the industrial applications for xenon and the extent of commercial markets available worldwide. The study showed that the quantity of xenon recoverable from THORP off-gas was a significant proportion (15%) of current world production. The report concluded that cryogenic separation of xenon from reprocessing plant off-gas was technically possible as part of a Kr-85 abatement process, and appeared to be commercially feasible. Its market survey indicated an expanding market for xenon, with growth driven by research in high technology industries.

xii. To date, neither BNFL nor La Hague has indicated any intention to retrofit krypton removal and storage. It is not proposed to construct krypton removal and storage at Rokkasho.

Iodine-129

xiii. Iodine-129 is a medium energy beta emitter (maximum energy 193 keV) with a very long half-life of 16 million years. It is produced in the fission of uranium with a yield of 1% and is released during reprocessing in relatively large quantities. Its long half-life means it will accumulate in the environment, become part of the iodine pool, and deliver a thyroid dose to the general population. Iodine is mobile in the environment, and rapidly incorporated in foodstuffs ingested by individuals. The highest environmental concentrations of iodine occur in seawater. Considerable uncertainty surrounds the transfer of iodine-129 to deep oceans and the sedimentation processes that may remove activity from biological chains (UNSCEAR, 1988). The estimated residence of iodine in the ocean is ~ 100,000 years (Raisbeck, 1995).

xiv. Iodine-129 discharges from La Hague between 1975-1992 are estimated to be about 632 kg, and from Sellafield between 1967-1992 about 608 kg. The total from the two plants up to 1992 is therefore about 1.2 tonnes. This is 10 times larger than the total iodine-129 present in the oceans before the nuclear era, approximately 25 times the input from nuclear weapons testing, and several hundred times greater that released by Chernobyl. In the period from 1993-1998, a further 1.4 tonnes of iodine-129 were discharged from La Hague and 0.36 tonnes from Sellafield, i.e. discharges in the 6 years after 1992 were more than in the previous 25 years. Annual I-129 discharges from the two sites have been steadily increasing, and have risen 10 fold in the past decade.

xv. The amount of iodine-129 discharged from La Hague and Sellafield in 1999 was eight times the total iodine-129 from all weapons test fallout. Iodine-129 from reprocessing therefore dominates all other iodine-129 sources in the world's oceans. In comparison, annual reprocessing emissions of caesium-137 and strontium-90 were never larger than 1% of cumulative nuclear weapons fallout (Raisbeck et al, 1995).

Tritium

xvi. Tritium (^3H) is the radioactive isotope of hydrogen. It is a low-range beta-emitter with a maximum decay energy of 18 MeV, and a half-life of 12.3 years. Tritium is formed naturally through cosmic ray interaction with H in the upper atmosphere. However anthropogenic tritium emissions considerably exceed natural production. Tritium commonly occurs as tritiated water, i.e. ^3HOH, and as elemental tritium gas, ^3HH. Tritium is created in nuclear fuel by activation of ^1H and ^2H, and as a tertiary fission product. Some tritium is released at reactors but the majority is released from reprocessing plants at the fuel dissolution stage.

xvii. In many respects, tritium is an unusual radionuclide. Tritium's high mobility and cycling in the biosphere, its multiple pathways to man, its relatively high RBE[5] value, its ability to bind with cell constituents to form organically-bound tritium (OBT), and the heterogeneous distribution of bound tritium in humans mark it out as a hazardous radionuclide (Fairlie, 1992). Unfortunately, these characteristics are not recognised in tritium's exposure limits: these are based solely on its extremely low dose per unit intake[6]. Ingested tritiated water has a biological half-life of 10 days, but ingested tritiated foodstuffs (ie OBT) have much longer half-lives which are poorly defined and may extend to many years in low turnover tissues. In sum, tritium is a very efficient distributor of radioactivity in the environment, including humans: its dose coefficient may be considerably greater than the current low value. The UK Government's CERRIE Report (2004) put forward a number of scientific arguments for increasing tritium's dose coefficient by a factor of ten or more. In 2006, the US EPA (2006) recommended that tritium's radiation weighting factor and therefore its dose coefficient) should be increased by a factor of 2.5 The following year, the UK Government's AGIR Committee (2007) recommended that tritium's radiation weighting factor (and dose coefficient) should be doubled.

[5] RBE – the relative biological effectiveness of tritium's beta particle compared with gamma rays
[6] tritium's dose coefficient is by far the lowest of all common radionuclides, and this is a cause for serious concern

Appendix B

Approximate age of fuels to be used during commissioning at Rokkasho reprocessing plant.

Fuel Type and Configuration	Quantity (tonnes U)	Burnup (GWd/ tU)	Years of cooling
PWR 17x17	90	12-47	8 to 20 years
PWR 15x15	110	34-47	6 to 15 years
PWR 14x14	10	32-36	9 to 17 years
BWR 8x8	220	18-40	8 to 20 years

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