

## C-14 ACCUMULATED IN ION EXCHANGE RESINS IN SWEDISH NUCLEAR POWER PLANTS Part 1: Results from Analyses and Calculation of Accumulation Factors based on Samplings 2008-2015

Per-Olof Aronsson Energy Solutions AB

Christina Lillfors-Pintér Vattenfall AB (Earlier: Vattenfall Research and Development AB and Ringhals AB)

> Åsa Henning Ringhals AB

## Abstract

This document reports the accumulation factors of C-14 in certain fractions of ion exchange resins in the Swedish Nuclear Power Plants, viz. Condensate Clean-up (CCU) in BWRs and Reactor Water Clean-up (RWCU) in PWRs.

The accumulation in wet CCU resins may be summarized by two correlations to the amount of CCU anion equivalents used, divided by the number of Equivalent Full Power Hours (EFPH). The first correlation is for units where the resins do not have an efficient air bubbling. The second correlation is for units with efficient air bubbling of the spent resins. These correlations enable the estimates of historic and future accumulations in wet CCU resins for all units.

The major part of the C-14 in the CCU resins is inorganic, i.e. members of the carbonate system. The organic fraction is usually in the range 1-5 % of the total accumulation.

The accumulation in the RWCU of the Ringhals PWR units R2, R3 and R4 is of the same magnitude as observed in the BWR CCU resins. However, the organic fraction is higher, 30 %.

The study reported here is a response to the SSM requirement to characterize the fractions of ion exchange resins responsible for the main accumulation of C-14. It is also a follow-up to an earlier study sponsored by SKB and performed by Åsa Magnusson (now Henning) in her doctoral thesis and related reports.

## **Executive Summary**

#### Introduction

This document reports the accumulation factors of C-14 in certain fractions of ion exchange resins in the Swedish Nuclear Power Plants; see *Summary Figure 1*. It includes data from samplings from 2008 up to and including 2015 (for one unit). The study reported here is a follow-up to an earlier study sponsored by SKB and performed by Åsa Magnusson (now Henning) in her doctoral thesis and related reports.





Summary Figure 1. Accumulation factors for total C-14 expressed as Bq/MWh<sub>th</sub> for the Swedish NPPs. The inset is a zoom-in for dried resins from the Forsmark units. The vertical bars indicate ±1 standard deviation. O1+O2 have a standard deviation of 2.4 Bq/MWh<sub>th</sub>. (A-2, sheet D Sum acc tot).

#### Background

The purpose of the original study by Magnusson et. al. was to establish data to calculate the inventory of C-14 in spent ion exchange resins in SFR1, the Final Repository for Short-lived Radioactive Waste. According to earlier safety analyses for SFR1, organic C-14 would be the dominating source for doses to the public in the remote future.

Following Magnusson's reports, The Swedish Radiation Safety Authority ordered a continuation in sampling and analyses of the resin fractions accumulating the largest amounts of C-14. These fractions are Reactor Water Clean-up systems (334) in the PWR units and condensate Clean-up systems (332) in the BWR units.

#### **Summary of Results**

The accumulation factors are shown in *Summary Table 1* and *Summary Figures 1 and 2*, together with relative standard deviations. The accumulation is shown as Bq/MWh<sub>th</sub> for total, inorganic and organic C-14. The inorganic C-14 consists of species in the carbonate system, mainly hydrogen carbonate, based on the chemical conditions in the systems. The organic species are most likely simple organic acids, since they have been identified in at least PWR systems during operation.





Summary Figure 2. Accumulation factors for organic C-14 expressed as  $Bq/MWh_{th}$  for the Swedish NPPs. The inset is a zoom-in for the units with low accumulations. The vertical bars indicate  $\pm 1$  Standard Deviation. (A-2, sheet D Sum acc org).

The organic fraction of the total accumulation is also given in the table. The standard deviations include the standard deviations in the sample analysis<sup>1</sup> and a 10 % standard deviation in the resin amounts used in the calculations<sup>2</sup>.

Unit	Accum	ulation, Bq,	/MWh <sub>th</sub>	Organic	Relative s	standard de	viation, %
	Total	Inorganic	Organic	fraction	Total	Inorganic	Organic
F1 + F2	2.30E+02	2.26E+02	3.97E+00	1.7%	13%	13%	14%
F3	6.12E+02	6.01E+02	1.06E+01	1.7%	4.9%	4.9%	7.2%
F1 + F2	1.67E+00	1.53E-01	1.52E+00	91%	22%	110%	24%
F3	7.45E+00	5.25E-01	6.92E+00	93%	15%	43%	17%
01+02	1.03E+02	9.74E+01	5.29E+00	5.2%	2.3%	2.3%	9.5%
03	4.15E+02	4.09E+02	5.95E+00	1.4%	2.9%	2.9%	8.1%
(B1+) B2	8.54E+02	8.36E+02	1.76E+01	2.1%	13%	16%	23%
R1	8.64E+02	8.40E+02	2.43E+01	2.8%	6.5%	5.2%	27%
PWR	4.95E+02	3.50E+02	1.46E+02	30%	8.4%	8.9%	7.7%

Summary Table 1. Accumulation factors for C-14 in Condensate Clean-up (BWR) and Reactor Water Clean-up (PWR) as Bq/MWh<sub>th</sub>, with relative standard deviations. The yellowmarked values for F1+F2 and F3 are for dried resins, prior to solidification. All other values are for wet resins. (A-2, sheet SUMTAB).

<sup>&</sup>lt;sup>1</sup> The standard deviation used is the maximum of the values obtained in the activity analysis (dominated by the standard deviation in counting) and the scatter between the replicates analyzed for each sample.

<sup>&</sup>lt;sup>2</sup> The PWR sampling has been assigned an additional standard deviation of 30 % due to the sampling procedure.

Since there are some uncertainties in the calculated production rates of C-14, the accumulation has been expressed as  $Bq/MWh_{th}$ , where all input data have been measured. To allow a comparison with earlier results, *Summary Table 2* shows the accumulation expressed as a percentage of the calculated production.

Unit	Accumula	tion, % of p	roduction	Organic	Relative s	standard de	viation, %
	Total	Inorganic	Organic	fraction	Total	Inorganic	Organic
F1 + F2	0.93%	0.92%	0.016%	1.7%	13%	13%	14%
F3	2.6%	2.5%	0.044%	1.7%	4.9%	4.9%	7.2%
F1 + F2	0.0068%	0.00062%	0.0062%	91%	22%	110%	24%
F3	0.031%	0.0022%	0.029%	93%	15%	43%	17%
01+02	0.49%	0.47%	0.026%	5.2%	2.3%	2.3%	9.5%
03	2.3%	2.2%	0.032%	1.4%	2.9%	2.9%	8.1%
(B1+) B2	4.3%	4.2%	0.088%	2.1%	13%	16%	23%
R1	3.8%	3.7%	0.11%	2.8%	6.5%	5.2%	27%
PWR	3.3%	2.3%	1.0%	30%	8.4%	8.9%	7.7%

Summary Table 2. Accumulation factors for C-14 in Condensate Clean-up (BWR) and Reactor Water Clean-up (PWR) as % of production with relative standard deviations. The yellowmarked values for F1+F2 and F3 are for dried resins, prior to solidification. All other values are for wet resins. (A-2, sheet SUMTAB).

#### **Results for Units**

**Forsmark 1 and 2 wet resins** have accumulations around 1 % of the calculated production. The Best Estimate has been chosen as 0.93 % total accumulation with 1.7 % organic fraction. Several calculation models were applied and the Mass Balance Model was chosen as the Best Estimate. The complete results are displayed in *Table 6* in the main body of the report.

**Forsmark 1 and 2 dried resins** contain only 0.0068 % with an organic fraction of 91 %. These values should be representative for the resins being sent to SFR1. However, the values for dried resins are based on only two dried samples, which are reflected by the high standard deviations<sup>3</sup>.

**Forsmark 3 wet resins** have an accumulation of 2.6 % with an organic fraction of 1.7 %. The complete results are displayed in *Table 7* in the main body of the report.

**Forsmark 3 dried resins** contain only 0.031 % with an organic fraction of 93 %. These values should be representative for the resins being sent to SFR1. However, the values for dried resins are based on only four dried samples, which is reflected by the high standard deviations.

**Oskarshamn 1 and 2** show an average accumulation of 0.49 % with an organic fraction of 5.2 %. The results are displayed in *Table 8* in the main body of the report.

 $<sup>^{3}</sup>$  The release of C-14 in the drying process has been measured in the exhaust air from the dryer. The release seems to be nearly 100 % for inorganic C-14, which the analyses of dried samples show. The sampling equipment was not designed to measure release of organic C-14.



**Oskarshamn 3** shows an average accumulation of 2.3 % with an organic fraction of 1.4 %. The result is somewhat lower than the values for its twin plant Forsmark 3. The results are displayed in *Table 9* in the main body of the report.

**Barsebäck 1 and 2** have not been analyzed in this sampling campaign, since the plant was permanently shut-down in 2005. The results from Magnusson et. al. have been recalculated using a different method for calculating average values of the subsamples. The accumulation has also been corrected for body-feed. The recalculated values are 0.87 % with an organic fraction of 8.7 %. The resulting standard deviation for the organic fraction is 51 %, due to the large scatter in the values. These values were judged too low, so a correlation model using input data from other units has been used, yielding an accumulation factor of 4.3 % with an organic fraction of 2.1 %. However, the accumulation factor for the organic C-14 is almost identical for both calculation methods. The results are displayed in *Table 10* in the main body of the report.

**Ringhals 1:** The present results for five separate cycles yield an average accumulation of 3.8 % with an organic fraction of 2.8 %. The first short cycle in 2009 had an accumulation of 7.3 %. The variation can be explained by the ratio of ion exchange resins used in relation to the water volume cleaned. This phenomenon is described in the next paragraph. The results are displayed in *Table 11* in the main body of the report.

**Ringhals 2, 3 and 4:** A combination of the present results and the corrected previous results for the PWRs yields a total accumulation of 3.3 % of the C-14 produced. The organic fraction is 30 %. The results are displayed in *Table 13* and *Figure 3* in the main body of the report.

**Samples collected and analyzed:** The results obtained in this study comprise many individual samples (5-173) per unit or group of units collected over some 5-7 years. The large number of samples and the long time periods covered are factors favoring the results of the present study; see *Summary Table 3* for details.

		Sam	ples	Sampling		
Unit	EFPY	Collected*	Analyzed*			
F1+F2	8.4	27	21	Collection tank		
F3	3.6	28	15	Collection tank		
01+02	3.8	56	56	Collection tanks		
03	2.9	15	15	Concrete tanks or collection tank		
R1	3.7	173	35	Collection tank		
R2	5.2	6	6	R2-R4: RWCU beds. Each bed		
R3	5.3	7	7	yields 10-20 subsamples		
R4	4.1	5	5	from separate transport vessels		
* Two or n	nore samp	les may com	nbined into a	a batch before analysis		
Batching applies only to units with Mass Balance Calculations; F1+F2, F3 and R1						
* In BWR CCU: One sample represents many back-flushings of resin from filters						
Three re	plicates an	alyzed for e	ach BWR ba	tched sample. Five replicates for PWR samples		

Summary Table 3. Overview of samples collected and batches analyzed, together with the energy production during the sampling time. (A-3, sheet Sampling Matrix 2014 rev 1).

## Correlation of the Total Accumulation in Condensate Clean-up Resins to the Parameter $x_a$

The results for the accumulation in BWR Condensate Clean-up resins show some results diverging within the units and between the units. Some of these variations can be explained



by the ratio between the anion equivalents used and the mass of cleaned water. See *Summary Figure 3*, where the accumulation of total C-14 (expressed as Bq/MWh<sub>th</sub>) is plotted vs. the parameter  $x_a$ = number of anion equivalents used/EFPH<sup>4</sup>. EFPH is proportional to the production of C-14 and the volume of condensate cleaned.

The variations in accumulation factors can be explained by two main sets of regression parameters:

One set for resins without efficient air bubbling; O3, F3, R1 and B1+B2 One set for resins with efficient air bubbling; O1+O2 and F1+F2

The upper regression line in *Summary Figure 3* has been fitted to the data from O3, F3 and R1. The reason for choosing these units was that they all lack an efficient air bubbling of the spent CCU resins. The lower regression line has been fitted to data from O1+O2 and F1+F2 with an efficient air bubbling of spent resins. All input data and the regressions lines are shown in *Summary Figure 3*.



Summary Figure 3. BWR CCU: Summary of regression input data and the two main regression lines based on O3, F3 & R1 and O1+O2 & F1+F2, respectively. (X-1, sheet D 2 tot).

The good fit of the regression lines may be interpreted as a saturation effect of C-14 in the CCU resins. This effect is discussed in *Appendix 22* for BWR and in *Appendix 5* for PWR. This regression model may be used as an alternative model to estimate historic and future accumulations of C-14, based on the value of the parameter  $x_a$ . The correlation has been performed for total, inorganic and organic C-14, respectively. The results are found in *Table 14* for BWR and *Table 15* for PWR.

The reason why the regression line for O1+O2 and F1+F2 fall below the regression line for the other units is probably due to the air bubbling used to homogenize spent resin slurry stored in tanks. The carbon dioxide in the air lowers the pH-value in the weakly buffered water. The pH-value is sufficiently low to allow carbonate on the resins to be converted to

<sup>&</sup>lt;sup>4</sup> EFPH = Equivalent Full Power Hours.

carbon dioxide, which is released to the water and transported away with the air. However, tests at Ringhals 1 did not show any significant release of C-14 when the Condensate Cleanup resins were air bubbled. The reason for this is unclear. The release of activity from resins in the storage tanks are supported by measurements of airborne releases of C-14 from O1 and  $R1^5$  when these units have had long shut-downs; see *Appendix 18*.

It has also been observed in several cases that samples with low total concentration often have a higher organic *fraction*. This is probably due to the fact that simple organic acids, such as formic acid and acetic acid with pK<sub>a</sub>-values lower than "carbonic acid", will remain in their anionic forms on the resin during air bubbling.

#### **Variations between Fuel Cycles**

*Summary Table 4* shows the variation in accumulation for different fuel cycles together with the standard deviations for the calculated accumulation factors. The variation is expressed as (maximum value)/(mean value). The number of cycles included in the study is also given. For the units F1+F2 and F3 it is not possible to attribute the accumulation to separate cycles. The reason for this is the handling of resins (see *Appendix 1, 2* and *14*). As a substitute, the ratios between the maximum and the mean concentration have been calculated. For the PWRs, a grand mean including all the units has also been calculated.

			Accun	nulation	Conce	ncentration Accumulation		ulation	
		Num-			Conc.	Conc.			
		ber	Total	Organic	Total	Organic	Total	Organic	
		of cy-	Max/	Max/	Max/	Max/	Standard	Standard	
Unit	Period	cles*	mean	mean	mean	mean	deviation	deviation	Comment
F1 + F2	2009-2013	10			3.0	3.0	13%	14%	Not possible to keep resins from
F3	2009-2014	3.5			2.5	1.8	4.9%	7.2%	different cycles separated
01+02	2008-2013	8	1.3	3.4			2.3%	9.5%	
03	2009-2014	5	2.3	3.6			2.9%	8.1%	
R1	2009-2015	7	1.9	1.4			6.5%	27%	
R2	See	7	1.9	3.1			19%	17%	2003 (2 cy); 2008; 2010; 2012-2014
R3	Comment	7	2.0	2.0			14%	13%	2003; 2008; 2009; 2011-2014
R4	See	5	1.9	1.7			17%	16%	2009; 2011-2014
PWR	Comment	19	2.6	3.0			8.4%	7.7%	One sample per cycle
* Cycles in	* Cycles included in accumulation calculations; for F1 + F2 some resins were produced before the sampling campaign								
No data available to show the variations for B1+B2									

Summary Table 4. Variations between cycles: "Accumulation" refers to accumulation in a limited amount of resin, which can be related to a certain operational period. This calculation was not possible to perform for F1+F2 and F3, since the collection tanks never were emptied during the sampling period. As a substitute, the variations in concentrations have been used. The standard deviations in the accumulations are also shown. (A-2, sheet MEANMAXSUM).

The ratios for the maximum accumulation to the mean accumulation are between 1.3 and 2.6 for the total accumulation. The corresponding concentration ratios for F1+F2 and F3 are 3.0 and 2.5. For the organic fractions the ratios are higher: 1.4-3.7. For F1+F2 and F3 they are 3.0 and 1.8, respectively.

For the PWRs, the ratios do not vary much for each unit; the exception is R2 for the very short cycle in 2012, which exhibits an unusually high organic accumulation. This is most

<sup>&</sup>lt;sup>5</sup> RWCU resins from the PWR units R2, R3 and R4 are also stored in the Waste Handling Building of R1 and contributing to the measured releases.



likely due to the (non)-saturation effect of the resins and is discussed in *Appendix 5* and in Chapter 13.4, "PWR Units".

### **Operational Periods Covered by this Study**

Despite the variations between individual fuel cycles, the most important feature for the SFR1 repository is the accumulated activity over time. For the units where the variations between the cycles are easy to follow (O1+O2, O3, R1, R2, R3 and R4), there is a tendency that the average stabilizes over the time period studied, according to "the law of large numbers".

*Summary Table 5* shows the number of Equivalent Full Power Years (EFPY) studied for each unit or unit pair. All units (except O1) have been followed for more than 2 EFPY each. All the other units have between 2.6 for O2 up to 4.6 EFPY for F1. BWRs have been followed for 22.4 EFPY and PWRs for 14.7 EFPY; altogether 37.1 EFPY. In the table the numbers of cycles and EFPY have been summed for the unit pairs which have a common handling of the resins.

		Numeron					
		Number					
	Sampling	of		Std.dev.	Std.dev.		
Unit	period	cycles*	EFPY	Total	Inorg.	Comment	
F1 + F2	2009-2013	10	8.38	13%	13%	Wet resin; ***	
F1 + F2	2009-2013	10	8.38	22%	110%	Dried resin; ***	
F3	2009-2014	3.5	3.63	4.9%	4.9%	Wet resin; ***	
F3	2009-2014	3.5	3.63	15%	43%	Dried resin; ***	
01+02	2008-2013	8	3.79	2.3%	2.3%		
03	2009-2014	5	2.90	2.9%	2.9%		
B1+B2	**	**	**	13%	15%		
R1	2009-2015	7	3.68	6.5%	5.2%		
BWR		33.5	22.4				
R2	See	7	5.22	19%	22%	2003 (2 cy); 2008; 2010; 2012-2014	
R3	Comment	7	5.33	14%	14%	2003; 2008; 2009; 2011-2014	
R4		5	4.12	17%	17%	2009; 2011-2014	
PWR		19	14.7	8.4%	8.9%	One sample per cycle	
BWR + PW	/R	52.5	37.1				
* Cycles included in accumulation calculations; for F1 + F2 some resins were produced before the sampling campaign							
** Not possible to relate the samples to a specific operating period, due to post-operational sampling							
*** Not possible to keep resins from different cycles separated							

Summary Table 5. The number of fuels cycles sampled and the corresponding number of Equivalent Full Power Years (EFPY) together with the standard deviation in the accumulations of C-14. For F1+F2 and O1+O2 the numbers of cycles and EFPH are summed for the unit pairs. (A-2, sheet STDDEV).

#### Are there Safety Margins in the Results?

The wet resin samples analyzed have been collected as late as possible in the waste handling process. However, for the Forsmark BWRs and the Ringhals PWRs there are important steps after the sampling, where C-14 may be driven off before solidification.

For the Ringhals PWR units R2-R4 there is air bubbling after the sampling and before the resins solidification. Measurements of airborne release from the Waste Handling Building indicate that the solidified amount of C-14 from the Ringhals PWR resins most likely is some 20-30 % lower than the values shown in *Summary Tables 1* and 2.

For the Forsmark BWRs, the values for the dried samples are significantly lower than for the wet resins. Measurements of C-14 in the exhaust air from the resin dryer supports the release



of C-14 from the resins, but cannot be used to quantify the residual in the resins. The values from the dried samples have already been included in the *Summary Tables 1* and 2.

#### Comparison with Previous Results by Magnusson et. al.

*Summary Table 6* shows a comparison between the results from this study and those from Magnusson et. al. The previous results have been corrected for dry substance contents and body-feed of inert materials, corrosion products (and for PWR) loading with lithium and boric acid. Some values from this study are higher than from Magnusson et. al., while others are lower. This pattern can be explained by

- 1 All units were not sampled in the previous study. E.g. unit B2 was used as reference plant for O1, O2 and R1. F3 was used as reference plant for O3
- 2 This study has analyzed substantially more samples covering longer operating periods covering start-up, operation and shut-down
- 3 The accumulation in BWR condensate clean-up resins depends on the parameter  $x_a$  described above
- 4 The organic fractions found in this study are usually lower than previously found. A possible explanation for the scatter in the BWR values may be the low concentrations. The organic fractions for the PWR resins show less scatter than for the BWR resins.
- 5 The samples from F1 were collected early in the handling chain, which overestimated the concentration, compared to this study, where the samples have been collected prior to drying and solidification.

		This	study		Ref. 4, corrected for dry substance and body-feed				
Unit	Accumulation, % of production			Organic	Accumulation, % of production			Organic	Sample
	Total	Inorganic	Organic	fraction	Total	Inorganic	Organic	fraction	from
F1 + F2	0.93%	0.92%	0.016%	1.7%	5.1%	4.3%	0.82%	16%	F1*
F3	2.6%	2.5%	0.044%	1.7%	1.7%	1.3%	0.40%	24%	F3
F1+F2 dried	0.0068%	0.00062%	0.0062%	91%					
F3 dried	0.031%	0.0022%	0.029%	93%					
01+02	0.49%	0.47%	0.026%	5.2%	0.50%	0.42%	0.080%	16%	B2
03	2.3%	2.2%	0.032%	1.4%	1.7%	1.3%	0.40%	24%	F3
(B1+) B2	4.3%	4.2%	0.088%	2.1%	0.50%	0.42%	0.080%	16%	B2
R1	3.8%	3.7%	0.11%	2.8%	0.50%	0.42%	0.081%	16%	B2
PWR	3.3%	2.3%	1.0%	30%	5.4%	3.9%	1.5%	29%	R2+R3
* The samples analyzed in Ref. 4 were collected early in the handling chain which overestimated									
the conce	ntration, co	ompared to	this study	/					

Summary Table 6. Comparison of the accumulation values in wet resins from Condensate Clean-up (BWR) and Reactor Water Clean-up (PWR) as % of production from this study and corrected values from Ref. 4. (A-2, sheet SUMTAB).

## **Doses Obtained in Sampling**

The doses obtained in sampling and analyses are about 0.3 mmanSv for one PWR cycle, despite high dose rates on some samples. The doses for sampling and analyses of condensate clean-up resins are estimated to be less than 0.1 mmanSv for one BWR cycle.





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## Versions

Version	Date	Changes
1	2011-09-28	First version
2	2012-01-25	Added samples fromO1+O2, O3, R3
		Included improved mass balance for CCU resins from F1+F2
		Calculation of C-14 inventory in SFR1
3	2012-04-11	Included correction for dry substance determination for all samples from the present report
		and Ref. 4
		One sample from F1+F2 (F12-09B3) corrected to lower ds, and consequently, to higher
		concentration
		Included all samples from 2011
		Added Appendix 17 on combustion correction
		Added density correction for BWR bead resins
4a	2013-08-15	New analyses data from 2011 and 2012 (some units also for 2013)
		BFF correction for PWR resins
		Minor corrections of body-feed data for O2
		Added Appendix 18 on losses of C-14 from resins in storage tanks (O1, R1-R4) and in
		drying process for resins (F1-F3)
4b	2013-10-14	Included comments from FKA, OKG, RAB and SKB
		Added:
		Appendix 19 Barsebäck
		Appendix 20 Chemical species of C-14
		Chapters in the main text related to Appendix 19 and 21
		Revised Executive Summary
5	2014-09-23	Added:
		Appendix 21: Drying of resin for F1+F2 and F3
		Appendix 22: correlation of accumulations in CCU resins
		Appendix 23: Analyses of some resin samples with respect to formic acid
		Chapters in the main text related to Appendix 21, 23 and 24
		Data from PWR 2013; O1+O2 (period 4) and O3 up to 2013 and R1 up to 2014
		Corrected mass balance calculation for F3 (one sample had wrong concentration values)
	2015 11 02	Revised Executive Summary
0	2015-11-02	Divided the document in Part 1 and Part 2.
		Added Chapters 17, 10: Appendices 22, 24: economilations in resins
		Added Chapters 17-19, Appendices 25-54, accumulations in resins
		<i>Executive Summary</i> with respect to above revisions
7	2016-01-06	Appendix 22 moved to Part 1
/	2010-01-00	Appendix 22 moved to Fart 1.
		Introduced Calculation Model 2 as <i>Best Estimate</i>
		Rewritten <i>Chapter 16</i> and <i>Appendices</i> 23-33 to reflect this
		Augmented Executive Summary, <i>Chapters</i> 17 and 18.
8	2016-01-14	Revised R1 accumulations by using recorded resin use 1992-98
Ũ	2010 01 11	Revised R2 accumulations by using extra anion resin data. Removed correction factor for
		extra anion resin.
		All tables, diagrams and text affected by this have been revised.
8	2016-01-16	Rev. 2: Included dried resin accumulations for FKA units.
8	2016-06-13	Revision 7 Anion resin mass replaced by anion equivalents in x -regression One
	2010 00 15	sample from $\Omega_3$ revised Appendices 24.33 recalculated Tables and diagrams
		sample from 05 fevrsed. Appendices 24-55 feediculated. Tables and diagrams
		revised. Appendix 34 added. Executive summary, chapters 16, 17 18 revised to
		reflect the revised results in Appendices. Files added and file names corrected in
		chapt. 21, Attachment files.



## Abbreviations

B1, B2	Shut-down reactor units (BWR-HWC) at the Barsebäck nuclear power plant
BKAB	Barsebäck Kraft AB
BFF	Body-Feed Factor
BM	Batch Model
BTRS	Boron Thermal Regeneration System; ion exchangers in R3 and R4
BWR	Boiling Water Reactor
CCU	Condensate Clean-up System (system number 332)
CS ds	Chemistry and Volume Control System (=RWCU in PWRs; syst. number 334) dry solids (dry substance); fraction of water-free substance in wet ion exchange resin ds=dw/ww
dw	dry weight
EFPH	Equivalent Full Power Hours
EFPY	Equivalent Full Power Years
F1, F2, F3	Reactor units (BWR-NWC) at the Forsmark nuclear power plant
FKA	Forsmarks Kraftgrupp AB
FP	Forward pumping of hot drains from the turbine system
(F)SAR	(Final) Safety Analysis Report
HWC	Hydrogen Water Chemistry
IX	Ion Exchange
MB	Mixed Bed
MBM	Mass Balance Model
MCM	Mean Concentration Model
NPP	Nuclear Power Plant
NWC	Neutral Water Chemistry
01	Reactor unit (BWR-NWC) at the Oskarshamn nuclear power plant
02	Reactor unit (BWR-HWC) at the Oskarshamn nuclear power plant
03	Reactor unit (BWR-NWC) at the Oskarshamn nuclear power plant
OKG	Oskarshamn nuclear power plant (OKG AB)
PWR	Pressurized Water Reactor
R1	Reactor unit (BWR-HWC) at the Ringhals nuclear power plant
R2, R3, R4	Reactor units (PWRs) at the Ringhals nuclear power plant
RAB	Ringhals nuclear power plant (Ringhals AB)
RC	Reactor Coolant: reactor water before clean-up
RWCU	Reactor Water Clean-Un System (system number 331 in BWR 334 in PWR)
SFP	Spent Fuel Pool Clean-up System (system number 324)
SFR(1)	Final Repository for Short-lived Radioactive Waste
SG	Steam Generator
SKB	Svensk Kärnbränslehantering AB (Swedish Nuclear Fuel and Waste Management Co.)
SRST	Spent Resin Storage Tank (system 342 in PWR)
122	Statens Strålskyddsinstitut (Swedish Radiation Protection Institute)
SSM	Strålsäkerhetsmyndigheten (Swedish Radiation Safety Authority)
WHR	Waste Handling Building
WD(CID	Waste Processing System (Clean Un): system number 242
WF(CU)	waster rocessing system (Clean-Op), system number 542
WW	wet weight





## 1. Introduction to and Scope of the Present Study

This report summarizes the analyses of the C-14 accumulated in some fractions of ion exchange resins in Swedish NPPs in the years 2008-2014 (some samples were also taken in 2015). The study has been performed according to a requirement from the SSM, The Swedish Radiation Safety Authority (*Ref. 1*).

Earlier work on this topic has been performed by Åsa Henning (née Magnusson), and has been summarized in her dissertation and some further reports (*Ref. 2, 3, 4*). Henning's study covered all fractions of ion exchange resins produced in Swedish NPPs. Based on those reports, the Swedish Radiation Protection Institute, SSI (now Swedish Radiation Safety Authority, SSM) decided that further analyses should be done on the fractions accumulating the largest amounts of C-14, viz.

- Reactor Water Clean-Up (RWCU) in PWRs
- Condensate Clean-Up (CCU) in BWRs

## 2. Reporting Format

In *Ref. 4* the resin accumulation of C-14 was expressed as a percentage of the production in the reactor system. This requires knowledge of the production rate for C-14 in the reactor coolant. The FSARs of the various reactors have values for the calculated production rates. The completely dominating production pathway is the reaction

O-17(n,α)C-14

in the reactor coolant. The calculated production rate can be used to prepare a material balance including the releases to air and water, which was done in *Ref. 2*.

However, expressing the accumulation of C-14 as  $Bq/MWh_{th}$  is a better way, since it only includes measured quantities and does not require the calculation of the production of C-14<sup>6</sup>. The evaluation of reliable accumulation rates will allow the estimation of the activity of C-14 in the resins by correlation to the thermal energy produced.

To allow comparison with earlier studies, this report will use both formats for reporting the accumulation. The percentage will also be used in the discussions.

In the text and tables, decimal separator is point (.) Results are usually given with two significant digits. Tables and diagrams show all results and important input data for the calculations. The appendices have a "Stand Alone" status, i.e. they may be read separately, since they contain comprehensive texts, diagrams and tables describing the input data and the results of the calculations. The complete calculations and all input data can be found in *Attachment Files*. All tables, diagrams and figures have a reference to the *Attachment File* and the particular sheet. Text, tables and figures from the appendices may be repeated in the main body of this report to give a full context.

<sup>&</sup>lt;sup>6</sup> Table 2 and its footnote illustrate this.



## 3. Operational Strategy for RWCU in Swedish PWRs

The operational strategies for the RWCU in the Ringhals PWR units are virtually the same in all three units. At the beginning of a fuel cycle, during start up, a fresh RWCU mixed-bed (cation in  $\text{Li}^+$ -form; anion in borate-form) is loaded and used for the whole cycle<sup>7</sup>. The resin is taken out of operation before being exposed to acidic and oxidizing conditions during shut-down. During shut-down, a mixed bed (cation in H<sup>+</sup>-form, anion in borate-form) is used<sup>8</sup>.

According to *Ref.* 2, very little C-14 remains in the reactor system after the refueling shutdown. The amount of resin added differs between the units, depending on the level of activated corrosion products in the unit.

The spent resin is back-flushed to a storage tank (SRST), from which it is transported to the BWR Waste Handling Building (WHB) in a shielded transport vessel. For the sampling campaigns covered by this report, the SRST has been emptied before the RWCU resin has been back-flushed<sup>9</sup>.

## 4. Operational Strategy for CCU in Swedish BWRs

The design and operational strategy of the CCU systems differ between the plants. Twoturbine plants (R1, F1 and F2) have one set of filters for each turbine. The amount of resin loaded differs, as well as the operational time. In addition, some plants also add some inert material in connection with loading (F1, F2)<sup>10</sup>, or continuously during operation as body-feed (F3, O2). The cation part is in H<sup>+</sup>-form and the anion part is in OH<sup>-</sup>-form. The operating temperature also varies between the plants. As of 2015, most all BWR plants except F1 and O2<sup>11</sup>, operate with Forward Pumping (FP) of the hot drains from the turbine; these drains are not cleaned by the CCU. See Table 1 for details<sup>12</sup>.

In addition, the CCU also accumulates metal oxides during operation. Inert masses and metal oxides are included in the determination of the dry weight of the resin samples. To obtain the net dry weight of the resin, a correction for the uptake of masses and metal oxides a correction is done; see *Chapter 7*.

The storage tanks for spent resin also receive additional resins in some plants. In F3 it receives resins from the Condensation Pool Clean-up System (SFP, 324) and from the Waste Processing System (WP, 342). The contribution of C-14 from these systems are estimated to 0.06 % (value from O3) and 0.04 % (value from F2) respectively, according to *Ref. 4*. These two sources will not contribute significantly, compared to the accumulation in CCU.

<sup>&</sup>lt;sup>7</sup> Well defined volumes of wet cation and anion resins are loaded. From the analyses certificates, the cation and anion capacities can be calculated in equivalents.

<sup>&</sup>lt;sup>8</sup> Li is removed from the reactor coolant in the beginning of the shut-down procedure by connecting the shutdown RWCU mixed bed and disconnecting the operational RWCU mixed bed. Boric acid is then added to the reactor coolant to establish subcritical conditions for reactor shut-down and refueling.

<sup>&</sup>lt;sup>9</sup> Except Ringhals 2 in 2003.

<sup>&</sup>lt;sup>10</sup> This practice stopped in the fall 2012. However, it will not affect the resins analyzed in this report. Annelie Jansson, FKA, personal communication, November 2015.

<sup>&</sup>lt;sup>11</sup> Status in October 2015: O2 will not operate any more.

<sup>&</sup>lt;sup>12</sup> After a power up-rate in 2013, F2 is forward pumping a part of the hot drains corresponding to approximately 15 % of the total feed water flow.



In O1 and O2, the storage tanks 342T48, T49 receive resin from CCU and WP. Sometimes fresh resin is added into those tanks to get more cleaning capacity for floor drainage which also is treated directly in those tanks<sup>13</sup>. R1, F1, F2 and O3<sup>14</sup> do not mix any other resins with the CCU resin before sampling and solidification.

						Body				Earlier	Current	
			Ion		Inert	feed of	Other	Virgin	Forward	ope-	ope-	
	Num-	Opera-	exch.		mass,	inert	resins	resins	pump-	rating	rating	
	ber of	tion	resin,	Cation:	kg	mass,	in sto-	to sto-	ing of	tempe-	tempe-	
	filters	time,	kg dw/	Anion	dw/	kg/d/	rage	rage	heater	rature,	rature,	Refe-
Unit	in CCU	weeks	filter	ratio ££	filter	filter	tank	tank	drains	deg. C	deg. C	rence
F1+F2	10	6-16	80	1.65:1	10¤¤				\$, \$\$	65-72	65-72	*
F3	7	6-16	70	3:1		0.43	324, 342		Yes	-	51-57	*
01	4	4-15	70	3:1			342	Yes	Yes	100?	67¤	**
02	6	4-15	60	3:1		0.35	342	Yes	No	-	56¤	****
03	7	5-20	90	3:1		0.40#			Yes	-	46¤	**
R1	8£	6+	90	2:1					&	81-83	35-55	***
* Jan-0	Ola Helr	nersson	, FKA			** Karl-I	Erik Inger	nansson	, Paul Arv	idsson O	KG	
*** Ri	kard He	llström,	Ringhal	s		**** Bo	Arnberg,	OKG				
# Only	in the p	period F	ebruary	2011-Ma	y 2011							
+ From	n June 2	013 the	operatir	ng time is	deter	mined by	the pres	sure dro	op , 1.5 bai	-		
\$ F1 N	lo forwa	rd pum	ping			\$\$ F2 Fo	rward pu	mping fi	rom June 2	2012		
£ R1I	n the cy	cle 2013	8-2014 oi	nly 6 filte	rs wer	e in oper	ation sim	ultaneo	usly			
& R1 F	P one t	urbine t	rain fror	n Septen	nber 20	)12. Both	turbine t	rains fro	om June 20	013		
¤ Nor	ninal te	mperati	ures; the	e actual te	empera	atures de	epend on	the cool	ing water	temper	ature	
¤¤ Pra	ctice sto	opped in	n autum	n 2012								
? The	earlier	operatir	ng temp	erature v	vas "hi	gh". No c	definite v	alue ava	ilable.			
${\tt ff}$ During the sampling period O1, O2 , O3 & F3 used resins where the ratio was based on cation and anion												
capa	capacities. All other units had resins with weight ratios. Appendix 34 describes the problem and											
calc	ulates t	he actua	al anion	capacitie	s used	to calcul	ate the a	ccumula	tions in P	art 2 of t	this repo	t

*Table 1. Summary of operational strategy for CCU in Swedish BWRs. All weights are given as dry weight (dw)*<sup>15</sup>*. (A-2, sheet CCU new rev4).* 

F1+F2 use carboxylate cation resin, while all other plants use sulfonate cation resins. The anion resin is of the strong quaternary type in all units. However, the fraction of anion capacity varies between some of the units; see *Table 1*. Since there are no cationic C-14 species (*Chapter 3, Appendix 20*) all C-14 is expected to be adsorbed on the anion resin in the CCU. Thus, the anion fraction in the resin is the governing factor for the accumulation of C-14.

## 5. Analytical Procedures

The analytical procedures closely follow those described in *Ref. 2, 3* and *4*. See *Appendix 9* for Analytical Procedures and *Appendix 10* for Calculation Formulas used to obtain the concentration values from liquid scintillation data. The analytical results, including calculations, are found in the *Attachment files*. *Appendix 12* shows two tables with analytical results as an example.

<sup>14</sup> O3 only during the sampling campaign. Sofie Englund, OKG, personal communication, January 2016.

<sup>&</sup>lt;sup>13</sup> Karl-Erik Ingemansson, OKG, personal communication, Sept. 2011.

<sup>&</sup>lt;sup>15</sup> One important factor for the accumulation of C-14: The number of anion equivalents per kg condensate cleaned; see *Chapter 12.1* and *Appendix 22* and *Appendix 34* in *Part 2*.



When the detected values are less than half the detection limit, one quarter of the detection limit replaces the detected value in the calculations<sup>16</sup>.

In Version 3, a further correction for the determination of the dry solids contents (ds) was introduced. This correction increased the dry weight concentrations and the accumulated amounts of C-14, compared to earlier data. The correction is 4 -16 %, depending on type of resin.

# 6. Calculation Models for the Accumulated Activity of C-14 in Sampled Resins

Depending on the operating and sampling possibilities, there are several calculation models available<sup>17</sup>:

**Batch Model (BM)**. In this model, a well defined amount of spent resin is operated for a well defined time period. When the resin is discarded, it is sampled in a representative way. The concentration obtained is multiplied by the amount of resin. To obtain consistent results, all weights are converted to dry weight (dw). The BM is used for RWCU in the PWR units, for the collection tanks for CCU resin at O1+O2 and for the concrete tanks at O3, respectively.

**Mass Balance Model (MBM)**. This model is applied to collection tanks obtaining a continuous flow (although at discrete times) of spent resin (see *Figure 1*). The contents of the tank is mixed by air bubbling, pump circulation and/or stirring. Furthermore, resin is also removed from the collection tank to solidification. Over the time period studied, several samples are withdrawn; mostly in connection with solidifications of resin.



Figure 1. Schematic mass balance over tank TD42 in F3. (A-5, pict. 12).

Careful record keeping of the additions and removals of resin allows a calculation of how much activity has been removed to solidification. The tank needs not to be empty at the start of the period studied, but then an initial sample is required to correct for activity not generated during the period studied.

The tank needs not to be empty at the end of the period; the residual content of activity may be determined from analysis of a sample and the residual resin amount. The MBM is used for R1, F1+F2 and F3. For F1+F2 the MBM is applicable only to a limited extent; see *Appendix 1* for details.

<sup>&</sup>lt;sup>16</sup> See last paragraph in *Appendix 10* and *Ref. 9* for a more detailed explanation.

<sup>&</sup>lt;sup>17</sup> These calculation models are used only to evaluate the activity accumulated in the sampled resins. They have not been used to calculate historical or future accumulations.



**Mean Concentration Model (MCM)**. If there is a less detailed record keeping of additions and removals, the Mass Balance Model may not be applicable. Instead, the total resin amount used during a long period (e.g. one year or longer) may be multiplied by the average concentration of activity in the resin, obtained from multiple samples during the period studied. The MCM is used as a supplementary method for F1+F2. The MCM might also be applied to calculations with the maximum and minimum concentration.

## 7. Resin Weights Used in the Calculations

Ion exchange resins contain varying amounts of water. To obtain consistent weights, the dry weight (dw) is used. All concentration values from the analyses are reported as Bq/kg dw. The procedure for determining the dw is briefly described in *Appendix 6*.

All powder resins used in CCU, SFP and WP have the dw given on the packages. Accordingly, it is easy to keep records on resin loadings and transfers based on dw. Complicating factors are body-feed; a continuous addition of inert material and accumulation of corrosion products, mainly iron oxides. For each unit, these factors are considered by calculating the Body-Feed Factor (BFF). See *Tables A1-1, A2-1, A3-4, A4-1, A13-3, A28-1*.

The bead resins used in RWCU in the PWR units are loaded on a volume basis. Each package has a well defined volume of wet resin. The manufacturer also states the dry substance (ds) fraction on a weight basis. To obtain the dw, the gross density of the wet resin has to be determined by weighing the plastic bags with a defined volume. The ds is typically 0.45 and the gross density 0.70 kg/liter. These resins are in H<sup>+</sup> and OH<sup>-</sup> form. Before operation, they are converted to lithium and borate form. The calculation of BFF is shown in *Table A6-2*.

		Produc-	
	Thermal	tion rate	
	power,	C-14,	
Unit	$\mathrm{MW}_{\mathrm{th}}$	Bq/s	C-14, Bq/MWh <sub>th</sub>
F1	2928	2.00E+04	2.46E+04
F2	2928	2.00E+04	2.46E+04
F2 new *	3253	2.20E+04	2.43E+04
F3	3300	2.20E+04	2.40E+04
01	1375	9.00E+03	2.36E+04
02	1800	1.00E+04	2.00E+04
03	3900	2.00E+04	1.85E+04
R1	2540	1.70E+04	2.41E+04
R2	2652	1.10E+04	1.49E+04
R3	3135	1.40E+04	1.61E+04
R4	2775	1.10E+04	1.43E+04
R4 new **	3300	1.47E+04	1.61E+04
* Test opera	ation start	ing in early	2013
** Test ope	ration star	ting in early	y 2015

## 8. Calculation of C-14 Production

Table 2. Thermal power and production rates of C-14 in the Swedish reactor units obtainedfrom their respective FSAR. The values apply to the conditions in the sampling period<sup>18</sup>.(A-3, sheet Prodrates).

<sup>&</sup>lt;sup>18</sup> The calculated production rate in O3 at 3900  $MW_{th}$  is lower than in F3 at 3300  $MW_{th}$ . This is due to the specification for the equilibrium cores used in the calculations. The most probable value for O3 is 2.6E4 Bq/s. It



In the FSAR of each unit there is a value for the production rate of C-14 at full power. By means of the thermal power, the production rate is converted to  $Bq/MWh_{th}$ . The thermal power production during the period is then used to calculate the total production of C-14. Table 2 summarizes the production rates used in the calculations.

## 9. Chemical Species of C-14 in the Reactor Systems and in the Resins

In *Appendix 20* there is a literature review and a discussion of C-14 species stable at the different conditions in the reactor systems and ion exchange resins during operation and shutdown. Table 3 summarizes the stable species as given in the *Ref. 11-15* at 300 °C and 25 °C. These temperatures approximate the temperatures in the reactor system during operation and in the RWCU and CCU, respectively. See *Appendix 20* for actual temperatures in the various systems.

Reactor type		BW	/R			PWR				
Operational			Shut-				Shut-			
state	Power o	peration	down	All	Power o	peration	down	All		
Thermody-										
namically		25 °C	25 °C	Airbor-			25 °C	Airbor-		
stable	300 °C	RWCU,	RC, SFP,	ne re-	300 °C	25 °C	RC, SFP,	ne re-		
species	RC	CCU	RWCU	leases	RC	RWCU	RWCU	leases		
С						x				
H <sub>2</sub> CO <sub>3</sub> , CO <sub>2</sub>	х		x	*			x	*		
HCO <sub>3</sub> <sup>-</sup>	х	x	x			x				
HCOO	(x)				х	**				
СН₃ОН					х					
CH₄				***		х		***		
* Measured	as "oxidize	ed species'	' in stack n	nonitorin	g					
** Analytically identified in Ringhals PWRs together with acetate and oxalate										
*** Measure	d as "redu	ced specie	s" in stack	monitor	ing					
(): With HW	C in parts of	of the syste	em where	the redo	x-potentia	l is below	- 0.5 V			

Table 3. Summary of thermodynamically stable species in reactor systems at various operating conditions (x) and species detected in analyses (\*, \*\*, \*\*\*). (A-2, sheet Species).

From a stability point of view, only the carbonate system should be stable in the RWCU and CCU resins at or near room temperature<sup>19</sup>. However, in the Ringhals PWRs, formic acid, acetic acid and oxalic acid have been identified in samples from the reactor coolant (See *Appendix 20* and *Attachment file R234-3*). The samples were analyzed after cooling to room temperature in the sampling line. Furthermore, organic compounds in the resins may not be stable over time; radiation induced reactions may convert the acids into other compounds by decarboxylation or oxidation by the radicals formed by irradiation of water (*Ref. 11*).

is obtained by scaling the F3 production rate by the thermal power ratio for O3/F3. However, the value from the FSAR has been used in the calculations. Klas Lundgren, ALARA Engineering, personal communication, 2013-08-14.

<sup>&</sup>lt;sup>19</sup> Elementary carbon is also stable in PWR RWCU during operation.



*Appendix 15* summarizes some experiments where formic acid has been determined. Three samples from BWR CCU and one sample from PWR BTRS were analyzed. The CCU samples showed that 50-90 % of the organic fraction consisted of formic acid, while it was only 1.5 % of the organic in the PWR sample<sup>20</sup>. Even if the analytical method is not fully qualified, the results give an indication that formic acid very likely is a major constituent in BWR CCU resins.

From Table 3 and the previous chapter, it seems reasonable that only anionic species are collected in CCU and RWCU resins. Therefore, the anion part of the resins is of special interest. See Chapter 12 and *Appendix 22* for further discussion.

## 10. Sampling

## 10.1. Samples collected

An overview of the handling and sampling of the spent resins is given in *Appendix 11*. Table 4 gives an overview of the sampling campaigns included in this study. F1+F2 and O1+O2 are sampled in pairs, named F12 and O12, respectively. In the appendices dealing with the details in the calculations, the separate samples and batches are indicated.

Year		20	800			20	09			20	10			20	11			20	)12			20	)13			20	14		-15	Sum				Ş	Sam	pliı	ng		
Unit	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1										
F1+F2*+					1	4	2			1	3	2						4	4	2	1		3							27	Col	lec	tio	n ta	nk				
F3**++							1	1	6	1	3		3	1					1	2			5			2	1	1		28	Col	lec	tio	n ta	nk				
01+02				2	4	3						5	7	2	1	5	6	5	4	2	3	6	1							56	Col	lec	tio	n ta	inks				
03													1		2		1		1	1	2	1	1		2	1				13	Cor	ncre	ete	tan	ıks d	or c	ollect	ion tar	۱k
R1					11				6	18	16	1		19	21	8	12	6	8	8	8	2	3	5	10	1	4	2	4	173	Col	lec	tio	n ta	nk				
R2 ***		1												1					1				1				1			6	R2-	R4:	RV	VCL	Jbe	ds.	Each	bed	
R3 ***			1				1									1		1					1			1				7	yie	lds	10-	20	sub	san	nples		
R4		1												1					1			1					1			5	fro	m s	ера	arat	te tr	ans	sport	vessels	5
* 20100	22:	Sar	mpl	e to	oo s	mal	ll to	o an	aly	ze																		Ο,	Sum	315									
** 20120	<b>)</b> 3:	Sar	nple	e to	o sr	mal	l to	ana	alyz	ze																													
*** One	*** One additional sample from 2003 for each unit																																						
+ incl. 1	+ incl. 1 wet reference sample 2013 Q3; 2 additional dried samples																																						
++ incl. 2	Lw	et r	efe	ren	ce s	am	ple	20	13 (	Q4;	4 ad	ldit	ion	al c	Irie	d sa	am	ples	5																				

Table 4. Overview of sampling for C-14 analyses. (A-3, sheet Sampling matrix 2014 rev 1).

For the PWR units R2, R3 and R4, one resin bed/fuel cycle is sampled; however, the sample from each bed is composed of several subsamples from the transport vessel.

For O1+O2 each collection tank is sampled prior to transfer to concrete tanks.

At O3, the sampling is done in connection with transfer to the concrete tanks.

At R1, the storage tank T41, receiving spent resin batches and delivering resin to waste solidification, is sampled.

At F3, the corresponding tank TD42 can be sampled only when resin is brought to drying before solidification.

For F1+F2, the corresponding tank, TC56, may be sampled when the tank contents is recirculated and when resin is brought to drying before solidification. In 2012 the tank TC58 was used for temporary storage of resins. It was also sampled.

<sup>&</sup>lt;sup>20</sup> According to *Ref. 16*, iron oxides are necessary to decompose formic acid to carbon monoxide; the BTRS resins are expected to contain virtually no iron oxides, due to their position in the RWCU. See also *Appendix 22*.



The sampling is deemed adequate for R2, R3, R4, R1, F1+F2, O1+O2 and O3 based on the number of samples collected. For F3, the sampling should have been more frequent<sup>21</sup>.

## 10.2. Doses from Sampling and Analyses

The doses from sampling are very low for BWR CCU samples. For PWR RWCU samples the sampling of one resin bed yields  $0.2-0.3 \text{ mmanSv}^{22}$ . Further handling and analyses of the resin samples yield only low doses; the chemists who analyzed the samples obtained 0.5 mmanSv.

These doses have been extracted from the electronic work dosimetry system. Since the TLD dosimetry system has a reporting threshold of 0.1 mSv, it will underestimate the doses. Table 5 summarizes the doses obtained in the period January 2010-September 2011.

The collective dose for a 21 month period amounts to about 2.2 mmanSv as an average with an upper and lower bound of 2.6 and 1.8 mmanSv, respectively. The main dose source is the sampling of six PWR resin beds. One PWR cycle yields about 0.3 mmanSv and one BWR cycle is estimated to yield about 0.1 mmanSv.

	Collecti	ive dose, n	nmanSv	
Type of work	Min	Max	Mean	Note
Analysis	0.5	0.5	0.5	Electronic work dose
Sampling PWR, 6 resin beds	1.2	1.8	1.5	Estimate from Electronic work dose
Sampling BWR, 3 sites	0.2	0.3	0.2	Estimate from Ringhals conditions
Sum	1.8	2.6	2.2	

Table 5. Summary of collective doses obtained in sampling and analyses in January 2010-September 2011. (A-3, sheet Person doses).

## 11. Results

The accumulation of C-14 in the resins is calculated in the *Appendix 1-5* and *13*. *In Appendix 19*, the accumulations for Barsebäck (*Ref. 3* and *4*) have been recalculated. In addition, a recalculation of the results reported in *Ref. 4* for the PWR units has been done, since a wrong density for the ion exchange resin had been used; see *Appendix 6-7*.

## 11.1. Uncertainty in Reported Values

Standard deviations given in the concentrations are the maximum value of the scatter between the subsamples analyzed in each batch, and the uncertainty in activity counting and analytical steps. With few exceptions, the scatter between the subsamples is larger than the combined uncertainties from counting, background subtraction, pipetting, weighing etc in the analytical procedures. The standard deviation in the values for the organic fraction is usually larger than for the inorganic and the total fraction; mainly due to concentrations close to the background level.

In addition, standard deviations in the accumulated activities and accumulation factors (% of production and  $Bq/MWh_{th}$ ) include a 10 % standard deviation for the amounts of resin. The PWR samples were assigned an additional 30 % uncertainty due to the sampling procedure.

<sup>&</sup>lt;sup>21</sup> The waste handling at F3 had given priority to solidification of other resins than CCU, due to storage space problems. Since the F3 CCU storage tank can be sampled only in connection with drying and solidification, few samples were taken.

<sup>&</sup>lt;sup>22</sup> Anders Höglund, Ringhals R1M, personal communication, Sept. 2011.



## **11.2. BWR F1 and F2 (Appendix 1 and Appendix 21)**

The results from F1 and F2 comprise two different types of resins:

- 1 Wet resins from the storage tanks prior to drying and solidification (*Appendix 1*)
- 2 Dried resins, ready for solidification (*Appendix 21*)

#### 11.2.1. Wet Resins (Appendix 1)

The record keeping for the resins handled during the period has not been comprehensive with respect to additions from the WHB in F1 and removals for solidification. There are not many details on the additions of resin to TC56, which may be the major uncertainty factor in the calculations. To establish the foundation for Mass Balance calculations, records for the resin handling in the tanks T42 and T43 in the F1 WHB have been used as supplementary information (*Appendix 14*) to recalculate the inventory of the storage tank TC56. Personal interviews by phone and e-mail and the records for the activity analysis of waste containers produced have been used as supplementary sources of information (*Attachment File F12-1, sheet TC56*).

Calculation Model	0	1	3	4	5	6	7	8
							Ref. 4 +	
	Recalc. to	Best		Inter-	Тоо		BFF + ds-	Compari-
Classification	dried resins	estimate	High	mediate	Low***	Too high	corr.	son
	Dried Mass				Minimum			
	balance	Mass	Maximum	Mean	conc. +			Back-
	TC56+	balance	conc. +	conc. +	annual	Ref. 4	Ref. 4 +	flushing
	T43+T42	TC56+	annual	annual	resin	BFF=1 no	BFF + ds-	tank (one
	****	T43+T42	resin use*	resin use	use**	TS-corr.	corr.	filter)
Equivalent Full Power Hours, F1	40057	40057						218
Equivalent Full Power Hours, F2	33356	33356						
Equivalent Full Power Years, F1	4.57	4.57						0.025
Equivalent Full Power Years, F2	3.81	3.81						
Body-Feed Factor, Average (BFF)	1.00	1.00	1.33	1.33	1.33	1.00	1.33	1
Dry Solids Correction Factor in Sample	2						1.04	
Organic fraction	91%	1.7%	1.7%	1.7%	58%	16%	16%	0.70%
Total accumulation in CCU	0.0068%	0.93%	3.6%	1.2%	0.0057%	3.7%	5.1%	7.7%
Organic accumulation in CCU	0.0062%	0.016%	0.063%	0.021%	0.0033%	0.59%	0.82%	0.054%
Total accumulation, Bq/MWh <sub>th</sub>	1.67E+00	2.30E+02	8.80E+02	2.95E+02	1.40E+00	9.10E+02	1.26E+03	1.90E+03
Inorganic accumulation, Bq/MWh <sub>th</sub>	1.53E-01	2.26E+02	8.73E+02	2.90E+02	4.39E-01	7.64E+02	1.06E+03	1.89E+03
Organic accumulation, Bq/MWh <sub>th</sub>	1.52E+00	3.97E+00	1.54E+01	5.15E+00	8.05E-01	1.46E+02	2.02E+02	1.34E+01
Total Std. Dev.	22%	13%	10%	104%	26%			11%
Inorganic Std. Dev.	110%	13%	10%	105%	51%			11%
Organic Std. Dev.	24%	14%	10%	78%	27%			26%
*Max. value is maximum for each : to	t, inorg, org							
**Min. value is minimum for each : to	ot, inorg, org							
*** Close to dried resins								
**** Based on two samples								

Table 6. Comparison of accumulation of C-14 in CCU resins at F1 and F2 from this study and Ref. 4. The column with Calculation model 0 is based on dried resins. All other columns are based on wet resins. (F12-1, sheet Summary dried).

From the discussion in *Appendix 1*, it is obvious that it is not possible to perform a correct Mass Balance calculation. The Mass Balance Model requires that the whole inventory of resin is completely mixed. In this case, this requirement is not fulfilled, since three separate tanks are involved, and only the last one in the handling chain is sampled.



To compensate for this problem, five calculation models are possible: The models use:

- 1. Mass Balance over T42, T43 and TC56, using all samples from TC56 (in 2012 also TC58)
- 2. Mass Balance over T42, T43 and TC56, using samples with the highest concentrations<sup>23</sup>
- 3. Annual use of CCU resins for the years 2008-2012, using Maximum Concentration
- 4. Annual use of CCU resins for the years 2008-2012, using Mean Concentration
- 5. Annual use of CCU resins for the years 2008-2012, using Minimum Concentration

Table 6 gives a summary of the accumulation of C-14 in the CCU resins based on the different calculation models used.

The Following additional calculation models are also included in Table 6:

- 0. Recalculates the mass balance from Model 1 to the concentration of dried resin samples, using the mean concentration from the mass balance.
- 7. Calculations from Ref. 4 corrected for BFF and ds

8. One single sample collected from a back-flushing tank, representing one filter; see *Appendix 16*.

Model 1 is the best estimate for the accumulation in wet resins; 0.93 % of the production with an organic fraction of 1.7 %. Model 4, using the average concentration of the samples and the annual resin consumption, yields a value of 1.2 % with an organic fraction of 1.7 %. However, the standard deviation is much higher for the Mean Concentration model, due to the large variations in the concentration values.

The value previously reported by Magnusson et. al. Model 7 (corrected), are considered too high, since the sample had been collected in the beginning of the handling chain. This conclusion is supported by the comparison sample from the Back-flushing tank, Model 8, which has an accumulation of 7.7 % and an organic fraction of 0.70 %. For the full discussion of the results from the various Calculation Models, see *Appendix 1*, Discussion.

The organic fraction found in wet resins in this study is lower than previously reported in *Ref.* 4. There is no obvious explanation for the discrepancy in the organic fraction. However, the present value is in the range 1-5 %, where most of the BWR samples in this study are found.

There are some peculiarities in the concentration values from this study; see figures and tables in *Appendix 1*. There is a decrease in the total concentration of C-14 from the samples collected in 2009 to those collected in 2010. Unfortunately, there is a period in between these two sampling periods, where no samples were taken. The reason for this was necessary campaigns for solidification of other resins.

A comparison with the inventory curve (*Figure A1-2B*) of tank TC56 implies that the lower concentration was present in the resins when they arrived in TC56; i.e. the losses have occurred already in F1 342T42+T43 or in the transfer of the resins. Another possible explanation for the variations may be that the sampling bottles have been left in the sampling position for varying times; since there is a slight underpressure, C-14 compounds may have been released<sup>24</sup>. The concentration of the organic fraction does not decrease as much as the

<sup>&</sup>lt;sup>23</sup> Used for preliminary calculations only.

<sup>&</sup>lt;sup>24</sup> Annelie Jansson, FKA, personal communication August 2013 and *Ref.* 8.



total (or actually, the inorganic fraction). This phenomenon is discussed in Chapter "Release of C-14 from Ion Exchange Resins in Storage Tanks" later on.

#### 11.2.2. Dried Resins (Appendix 21)

Calculation Model 0 is a recalculation of the Mass Balance according to Model 1, assuming that the concentrations from two dried samples are representative for all resins<sup>25</sup>. The remaining accumulation is only 0.0051 % of the calculated production of C-14. The organic fraction in the dried resins is 91 %, but it corresponds to an absolute accumulation of 0.0047 %. The standard deviation is 22 % resp. 24 % for the total and. the organic fraction.

#### 11.3. BWR F3 (Appendix 2 and Appendix 21)

The results from F3 comprise two different types of resins:

- 1 Wet resins from the storage tanks prior to drying and solidification (*Appendix 2*)
- 2 Dried resins, ready for solidification (*Appendix 21*)

11.3.1. Wet Resins (Appendix 2)

	Dried resins*			
	Mass	Mass		
	balance	balance		
	TD42	TD42	Ref. 4 +	O3 Sum
	Sept 2009-	Sept 2009-	BFF + ds-	090913-
	Dec 2012	Dec 2012	corr.	140829
Equivalent Full Power Hours (EFPH)	31800	31800		25380
Equivalent Full Power Years (EFPY)	3.63	3.63		2.90
Body-Feed Factor, Average (BFF)	2.08	2.08	2.08	1.12
Dry Solids Correction Factor in Sample			1.16	
Residual after drying:				
Organic fraction	93%	1.7%	24%	1.4%
Total accumulation in CCU	0.031%	2.6%	1.7%	2.3%
Organic accumulation in CCU	0.029%	0.044%	0.40%	0.032%
Resin, kg dw		22555		
Total accumulation, Bq/MWh <sub>th</sub>	7.45E+00	6.12E+02	4.05E+02	4.15E+02
Inorganic accumulation, Bq/MWh <sub>th</sub>	5.25E-01	6.01E+02	3.08E+02	4.09E+02
Organic accumulation, Bq/MWh <sub>th</sub>	6.92E+00	1.06E+01	9.72E+01	5.95E+00
Total Std. Dev.	15%	4.9%		2.9%
Inorganic Std. Dev.	43%	4.9%		2.9%
Organic Std. Dev.	17%	7.2%		8.1%
* Based on four samples				

Table 7. Comparison of accumulation of C-14 in CCU resins at F3 from this study, Ref. 4 and<br/>from O3. The leftmost column with results is based on dried resins.(F3-1, sheet Summary dried).

The iron oxides in the resins will be around 10 % of the total dw. For details, see *Appendix 2*, *Table A2-1*. The Body-Feed Factor, (BFF), also including inert material, is probably somewhat low, since some inventory values in the storage tank TD42 are slightly negative in September 2010 and April 2011.

The record keeping for the resins handled during the period seems to be fairly good with respect to additions and removals for solidification. Personal interviews by phone and e-mail

<sup>&</sup>lt;sup>25</sup> The reference concentration is the average concentration calculated from the Mass Balance model.



and the records for the activity analysis of waste containers produced have been used as supplementary sources of information (*Attachment File F3-1, sheet TD42*). The major uncertainty factor seems to be the Body-Feed of inert material. With a nominal addition rate of inert material, it will be almost as much inert material as resin.

The fraction accumulated, 2.6 %, is higher than 1.7 % obtained when correcting the results from *Ref. 4* for BFF and ds. On the other hand, the organic fraction found in this study, 1.7 %, is lower than 24 % in the previous one. There is no obvious explanation for the discrepancy in the organic fraction. The present value is in the range 1-5 %, which is obtained for most BWR samples.

The results from O3 have also been included as a comparison, since the units are considered as twin plants<sup>26</sup>. The best way to compare the units is to use the accumulation expressed as Bq/MWh<sub>th</sub>. The F3 accumulation is higher for the inorganic C-14, as well as for the organic.

There are some peculiarities in the concentration values from this study; *Appendix 2*, *Figure A2-4* and *Table A2-2* show that there is a decrease in the total concentration of C-14 in the samples collected in the latter part of 2010 and in early 2011, compared with the earlier samples. The concentration of the organic fraction does not decrease as much as the total (or actually, the inorganic fraction). The sampling procedure, discussed under F1+F2 above, may be one possible explanation for this.

#### 11.3.2. Dried Resins (Appendix 21)

A recalculation of the Mass Balance for wet resins to dried resins has been done, assuming that the results from four dried samples are representative for all resins. The remaining accumulation is only 0.019 % of the calculated production of C-14. The organic fraction in the dried resins is 93 %, but it corresponds to an absolute accumulation of 0.018 %. The standard deviation is 15 % resp. 17 % for the total and the organic fraction.

#### 11.4. BWR 01 and 02 (Appendix 3)

The record keeping for O1 and O2 is adequate. From *Tables A3-3, -22, -32* and *-42* in *Appendix 3* it can be seen that the accumulation of C-14 in individual tank fillings ranges between 0.01 % and 2.7 % of the calculated production. *Table 8* summarizes the results from O1 and O2. The mean value is 0.49 %, including a Body-Feed Factor, BFF=1.09. The organic fraction is 5.2 %.

The organic concentration varies strongly between the batches; usually samples with low total concentrations have the lowest *organic concentration* but the highest *organic fraction*. The largest amounts of organic C-14 come from the tanks with the highest concentrations, so the average value of 5.2 % for the organic fraction is not influenced by the use of ¼ of detection limit values in samples with low concentrations. In Part 4, two tanks had unusually high organic fractions and contributed almost 80 % of the organic accumulation. The largest contribution came from a tank with resins collected in connection with shut-down of O2 in June 2013; cf. *Tables A3-3, A3-22, -32 and -42*. There is a ratio of about 100 between the

 $<sup>^{26}</sup>$  However, O3 is operating at higher power, but has a lower calculated production rate of C-14; see Table 2 and its footnote.

highest and the lowest organic concentrations. The results from Part 3 and Part 4 are almost exclusively from O2, since O1 was shut down for most of the sampling time<sup>27</sup>.

	Part 1	Part 2*	Part 3*	Part 4*	Part 1.4
	Sep2008-	Sep 2010-	Jul 2011-	Jan2013-	Sep 2008-
01+02	Mar 2009	May 2011	Dec 2012	Jun 2013	Jun 2013
Equivalent Full Power Hours (EFPH), O1	5338	2399	2691	66	10493
Equivalent Full Power Hours (EFPH), O2	6333	3673	9454	3259	22719
Equivalent Full Power Years (EFPY), O1	0.61	0.27	0.31	0.01	1.20
Equivalent Full Power Years (EFPY), O2	0.72	0.42	1.08	0.37	2.59
Equivalent Full Power Years (EFPY), O1 + O2	1.33	0.69	1.39	0.38	3.79
Body-Feed Factor, Average (BFF)	1.06	1.17	1.10	1.11	1.11
Dry Solids Correction Factor in Sample					
Organic fraction	3.0%	4.2%	4.3%	14%	5.2%
Total accumulation in CCU	0.30%	0.63%	0.57%	0.63%	0.49%
Organic accumulation in CCU	0.0090%	0.026%	0.024%	0.087%	0.026%
Total Bq/MWh <sub>th</sub>	6.51E+01	1.33E+02	1.17E+02	1.26E+02	1.03E+02
Inorganic Bq/MWh <sub>th</sub>	6.32E+01	1.27E+02	1.12E+02	1.09E+02	9.74E+01
Organic Bq/MWh <sub>th</sub>	1.93E+00	5.58E+00	5.06E+00	1.75E+01	5.29E+00
Total Std. Dev.	5.8%	4.1%	4.8%	4.0%	2.3%
Inorganic Std. Dev.	5.9%	4.1%	4.9%	4.3%	2.3%
Organic Std. Dev.	7.5%	7.6%	17%	15%	9.5%
* Inert masses for O2 calculated from operation	onal time a	nd dosage	rate		

Table 8. Summary of accumulation of C-14 in CCU resins for O1 and O2.(O12-5, sheet Summary).

## 11.5. BWR 03 (Appendix 13)

The record keeping for O3 is adequate. *Table 9* summarizes the results from O3. It also includes a comparison with results from F3 in *Appendix 2* and the corrected results from *Ref. 4*. The mean value is 2.3 %, including a Body-Feed Factor, BFF=1.12. The results from O3 are lower than those for F3, on a percentage basis, as well as Bq/MWh<sub>th</sub>. See the discussion for BWR F3. The organic fraction of 1.4 % is within the "normal" BWR range 1-5 %.

<sup>&</sup>lt;sup>27</sup> Some tanks contents with resins solely from WPCU had low concentrations of C-14. It can be assumed that WPCU does not contribute any significant amounts of C-14.



	O3 Sum		Ref. 4 +
	090913-	F3, Mass	BFF + ds-
	140829	Balance	corr.*
Equivalent Full Power Hours (EFPH)	25380	31800	
Equivalent Full Power Years (EFPY)	2.90	3.63	
Body-Feed Factor (BFF)	1.12	2.08	2.08
Dry Solids Correction Factor in San		1.16	
Organic Fraction	1.4%	1.7%	24%
Total accumulation in CCU	2.3%	2.6%	1.7%
Organic accumulation in CCU	0.032%	0.044%	0.40%
Resin kg dw	9820		
Total Bq/MWhth	4.15E+02	6.12E+02	3.11E+02
Inorganic Bq/MWhth	4.09E+02	6.01E+02	2.37E+02
Organic Bq/MWhth	5.95E+00	1.06E+01	7.43E+01
Tot. std. dev.	2.9%	4.9%	
Inorg. std. dev.	2.9%	4.9%	
Org. std. dev.	8.1%	7.2%	

Table 9. Summary of accumulation of C-14 in CCU resins at O3. F3 was used as model plant for O3 in Ref. 4. (O3-2 sheet Summary).

## 11.6. BWR B1 and B2 (Appendix 19)

The data from B1 and B2 analyses (*Ref. 3* and 4) have been recalculated in this report. The reasons for recalculating the accumulation of C-14 in CCU are the following:

- Calculating a grand mean from all samples available
- Including standard deviations from the samples and the resin amount
- Including Body-Feed Factor (1.07)<sup>28</sup> and dry substance correction factor (ds, 1.16, *Appendix 6*) in the calculations.
- Including a calculation based on the  $x_a$ -correlation

	B1+B2 analyses	B1+B2 x <sub>a</sub> -corre- lation	B2, Ref. 4 + BFF + ds-corr.	01+02	B2, Ref. 4
x <sub>a</sub> -value (average B1+B2 1992-2005)		0.20			
Organic fraction	8.7%	2.1%	16%	5.2%	16%
Total accumulation in CCU	0.87%	4.3%	0.50%	0.49%	0.40%
Inorganic accumulation in CCU	0.79%	4.2%	0.42%	0.47%	0.34%
Organic accumulation in CCU	0.075%	0.088%	0.080%	0.026%	0.064%
Total Bq/MWh <sub>th</sub>	1.73E+02	8.54E+02	1.06E+02	1.03E+02	8.53E+01
Inorganic Bq/MWh <sub>th</sub>	1.58E+02	8.36E+02	8.93E+01	9.74E+01	7.16E+01
Organic Bq/MWh <sub>th</sub>	1.50E+01	1.76E+01	1.70E+01	5.29E+00	1.36E+01
Total Std. Dev.	20%	13%		2.3%	
Inorganic Std. Dev.	22%	16%		2.3%	
Organic Std. Dev.	51%	23%		9.5%	

Table 10. Summary of the accumulation of C-14 in B1 and B2.(B-2 sheet Summary).

The samples were taken from the top layers of dewatered resin in concrete tanks. However, it was not possible to correlate the sampled resin to a certain operating period, so the

<sup>&</sup>lt;sup>28</sup> Same as originally calculated for O1+O2, part 1.

calculations were done according to the Batch Model, using the annual consumption of resin. The annual accumulation was then calculated, using the average annual production of energy. The details are shown in *Appendix 19*.

The calculated accumulation is 0.87 % of the calculated production, some 50 % higher than for O1+O2. However, there is a possibility that part of the C-14 in the sampled resin has been volatilized. To estimate a maximum value for the accumulation, the  $x_a$ -correlation described in chapter 12, Eq. (12-1) has been used. This estimate yields an accumulation of 4.3 % with an organic fraction of 2.1 %. However, the organic accumulation factor is almost identical for both calculation models; 0.088 % for the  $x_a$ -correlation and 0.0075 % from the analyses. This indicates that the organic C-14 is less volatile than the inorganic.

*Table 10* shows the calculated accumulation, compared with the values for O1+O2 and the corrected values from *Ref. 4*. The values for B1+B2 are higher than for O1+O2, which is reasonable, since B1+B2 did not use air bubbling in the collection tanks. The uncertainties for the values based on the analyses are rather high, which is a consequence of the scatter in the analysis values and the few samples (four) analyzed. The value obtained from the  $x_a$ -correlation, 4.0 %, has a lower standard deviation and may be used as an upper limit.

#### 11.7. BWR R1 (Appendix 4)

The record keeping for the resins handled during the period is adequate in almost all aspects. The only uncertainty is the Body-Feed Factor, BFF, which accounts for accumulation of iron oxides. It has been calculated in *Appendix 4* and has the value 1.09, which has been included in the calculations.

*Table 11* gives a summary of the accumulation of C-14 in the CCU resins for the seven periods covered. The first short cycle in 2009 had an accumulation of 7.3 %, while the years 2010-2013 were in the range 4.0-4.6 %. The cycle 2013-2014 had only 1.7 % accumulation. In the latest cycle, 2014-2015, the accumulation is 3.5 %. The organic fraction was highest in the cycle 2013-2014; 7.8 %. The lower accumulation in 2013-2014 is discussed in *Appendix 22* and in *Chapter 12*," Factors Governing the Accumulation of C-14 on Resins". The variation is mainly in the inorganic part. The average accumulation over the seven periods is 3.8 % with an organic fraction of 2.8 %.

The organic fraction is in the range 1-5 %, where most of the BWR samples are found.



								Mean
				2011- Aug	Aug 2012-	Apr 2013-	Jul 2014-	2009-Apr
	2009	2010	2011	2012	Apr 2013	Mar-2014	Apr 2015	2015
Equivalent Full Power Hours	1497	4125	4510	4267	5235	6366	6281	32280
Equivalent Full Power Years	0.17	0.47	0.51	0.49	0.60	0.73	0.72	3.68
Body-Feed Factor, Average (BFF)	1.09	1.09	1.09	1.09	1.09	1.09	1.09	1.09
Dry Solids Correction Factor in Sa	mple							
Organic fraction	1.3%	2.1%	1.2%	3.7%	1.7%	7.8%	3.9%	2.8%
Total accumulation in CCU	7.3%	3.9%	4.6%	4.0%	4.6%	1.7%	3.5%	3.8%
Organic accumulation in CCU	0.095%	0.080%	0.056%	0.15%	0.077%	0.13%	0.14%	0.11%
kg resin used	1530	4680	4230	5447	4746	3573	4485	28691
Total Bq/MWhth	1.72E+03	9.13E+02	1.08E+03	9.48E+02	1.09E+03	4.01E+02	8.35E+02	8.64E+02
Inorganic Bq/MWhth	1.70E+03	8.94E+02	1.06E+03	9.13E+02	1.08E+03	3.70E+02	8.03E+02	8.40E+02
Organic Bq/MWhth	2.23E+01	1.89E+01	1.31E+01	3.53E+01	1.82E+01	3.14E+01	3.22E+01	2.43E+01
Total Std. Dev.	7.4%	4.9%	9.0%	13%	22%	23%	17%	6.5%
Inorganic Std. Dev.	7.4%	3.8%	8.9%	14%	12%	24%	16%	5.2%
Organic Std. Dev.	21%	23%	50%	40%	67%	13%	94%	27%

Table 11. Summary of accumulation of C-14 in CCU resins at R1. (R1-4, sheet Summary).

#### 11.8. PWR R2, R3 and R4 (Appendix 5)

The results for the PWR units are reported in *Appendix 5*. In addition, a recalculation of the results reported in *Ref. 4* for the PWR units has been done in *Appendix 7*, since a wrong density for the ion exchange bead resins had been used. The correction is based on a determination of the resin gross density and a recalculation of the resin dry weight in *Appendix 6*. This correction also includes the ds-correction in analyses. One additional correction factor, BFF=1.15, has also been introduced to compensate for the resin loading by lithium and borate. See *Appendix 6* for details. *Table 12* shows the results of the recalculation; it reduces the accumulation of C-14 in the resins to some 80 % of the previously reported values (*Ref. 4*).

Unit	Resin use	Old	Corrected	Operation	Org. fraction
R3	RWCU operation	7.3%	5.9%	2002-2003	29%
	RWCU 2 y operation				
R2	(incl. some other resins)	6.0%	4.9%	2001-2003	28%
R4	RWCU shut down, SFP, WP	1.6%	1.3%	2002-2003	35%
R3	RWCU shut down, SFP, WP	2.5%	2.0%	2002-2003	30%
Mean	RWCU operation	6.7%	5.4%		29%
Mean	RWCU shut down, SFP, WP	2.1%	1.7%		33%
Mean	All	8.7%	7.1%		31%

Table 12. Summary of corrected accumulation of C-14 in PWR RWCU resins<sup>29</sup>. "Corrected" means that the accumulation has been corrected for the wrong resin density and dsdetermination used in Ref. 4. In addition, a BFF-value of 1.15 has been introduced to account for the lithium and borate loading of the resin. Note that the values for accumulations are in "Absolute %", i.e. percent of the production. The "Organic fraction" is given in the rightmost column. (R234-1, sheet Old).

<sup>&</sup>lt;sup>29</sup> The corrected accumulations differ from the values given in *Table 13*. The reason for this is that the values in *Table 12* have been recalculated from the original concentration values in *Ref.4*, but with a lower amount of resin.



Samp- ling year	Unit	Cycle	EFPY	Accum. total	Accum. Inorg.	Accum. Org.	Org. frac- tion	Total Bq/ MWhth	Inorganic Bq/ MWhth	Organic Bq/ MWhth	Std. Dev. Total	Std. Dev. Inorg.	Std. Dev. Org.
2003	R2	2001-2003	1.75	4.1%	3.0%	1.2%	27%	6.12E+02	4.47E+02	1.75E+02	39%	42%	35%
2008	R2	2007-2008	0.64	5.8%	4.9%	0.91%	16%	8.72E+02	7.37E+02	1.35E+02	32%	32%	32%
2011	R2	2010-2011	0.99	1.7%	1.1%	0.62%	36%	2.57E+02	1.64E+02	9.33E+01	32%	32%	32%
2012	R2	2012-2012	0.27	4.1%	1.1%	2.9%	72%	6.06E+02	1.69E+02	4.37E+02	33%	33%	33%
2013	R2	2012-2013	0.79	1.7%	1.1%	0.54%	32%	2.49E+02	1.68E+02	8.02E+01	32%	32%	32%
2014	R2	2013-2014	0.79	1.7%	1.1%	0.53%	32%	2.47E+02	1.68E+02	7.98E+01	32%	32%	32%
2003	R3	2002-2003	0.68	5.4%	3.9%	1.6%	28%	8.58E+02	6.18E+02	2.47E+02	35%	36%	34%
2008	R3	2007-2008	0.81	2.9%	1.9%	0.92%	32%	4.53E+02	3.07E+02	1.46E+02	32%	32%	32%
2009	R3	2008-2009	0.93	2.2%	1.6%	0.56%	26%	3.43E+02	2.55E+02	8.83E+01	32%	32%	32%
2011	R3	2010-2011	0.90	1.4%	1.1%	0.30%	22%	2.19E+02	1.71E+02	4.82E+01	32%	33%	32%
2012	R3	2011-2012	0.43	3.8%	2.7%	1.1%	28%	6.07E+02	4.37E+02	1.81E+02	32%	32%	32%
2013	R3	2012-2013	1.04	2.1%	1.4%	0.67%	32%	3.33E+02	2.26E+02	1.07E+02	32%	32%	32%
2014	R3	2013-2014	0.53	2.2%	1.4%	0.83%	37%	3.56E+02	2.24E+02	1.32E+02	32%	32%	32%
2009	R4	2008-2009	0.82	6.9%	4.7%	2.1%	31%	9.78E+02	6.72E+02	3.05E+02	32%	32%	32%
2011	R4	2010-2011	0.85	8.5%	6.3%	2.2%	26%	1.22E+03	9.01E+02	3.16E+02	32%	32%	32%
2012	R4	2011-2012	0.70	2.9%	2.0%	0.85%	29%	4.13E+02	2.91E+02	1.22E+02	32%	32%	32%
2013	R4	2012-2013	0.62	2.7%	1.8%	0.89%	32%	3.89E+02	2.63E+02	1.26E+02	32%	32%	32%
2014	R4	2013-2014	1.13	1.5%	1.0%	0.49%	33%	2.13E+02	1.44E+02	6.92E+01	32%	32%	32%
	R2-R4	Mean	14.68	3.3%	2.3%	1.0%	30%	4.95E+02	3.50E+02	1.46E+02	8.4%	8.9%	7.7%
	R2	Mean	5.22	3.1%	2.2%	0.9%	30%	4.66E+02	3.30E+02	1.39E+02	19%	22%	17%
	R3	Mean	5.33	2.7%	1.9%	0.79%	30%	4.25E+02	3.01E+02	1.26E+02	14%	14%	13%
	R4	Mean	4.12	4.4%	3.1%	1.3%	29%	6.33E+02	4.48E+02	1.85E+02	17%	17%	16%
	Number of cycles		19										

Table 13. Summary of accumulation of C-14 in PWR RWCU resins, including corrected results from Ref. 4 (years 2001-2003). The sampling in 2003 for R2 was for resins used for two cycles. (R234-1, sheet New Summary).

*Table 13* shows a summary of the results obtained in this study for RWCU, including the corrected results from *Ref. 4* (years 2001-2003). The average accumulation for RWCU is 3.3 %, with an organic fraction of 30 %. The maximum accumulation, 8.5 %, was found in R4 in 2011 and the lowest, 1.4 %, in R3 in 2011.

The organic fraction is fairly stable, with two exceptions: R2 in 2008 and 2012. The exceptionally high value in 2012 is discussed in *Appendix 5* and *Appendix 20*. Most likely, it is an effect of a non-saturation of the RWCU ion exchange resin early in the fuel cycle. The correction of the old results brings them down to a level in the upper range of those found in this study. However, among the units, R4 has a higher average value than R2 and R3. There are also variations between R2 and R3. The reason for this variation is not clear, but there are several chemical and operational factors that may contribute. The factors are listed in the chapter" Discussion" and are discussed more thoroughly in *Appendix 5*.

## **12. Factors Governing the Accumulation of C-14 on Resins**

#### 12.1. BWR CCU Resins (Appendix 22)

For several of the BWR units there seems to be a fairly stable total concentration of C-14 on the CCU resins. This concentration is in the range 1-4E6 Bq/kg dw for O3, F3 and R1; see *Figures A13-2, A2-4 and A4-52*. This may be a hint that there is a saturation effect for the


According to the literature, no cationic species of C-14 are present; see chapter 9 and *Appendix 20*, where the speciation of C-14 is discussed. Therefore, it is reasonable to assume that it is only the anion part of the resin that absorbs C-14. Accordingly, only the anion part of the resin is considered<sup>30</sup>.



Figure 2. Summary of regression input data and the regression line based on R1data. (X-1, sheet D R1 tot).

The introduction of FP lowered the amount of condensate to be cleaned to some 70 %. In connection with this, the use of CCU resin also decreased. Therefore, an effort was made to correlate the accumulation against the amount of CCU anion equivalents used in the cycles. Since many cycles had very different lengths, the consumption was normalized to the production of power and C-14, expressed as EFPH.

*Figure 2* shows the total accumulation rate vs. this normalized consumption of anion equivalents for R1. Despite the visible scatter of the points, the regression has a good fit  $(r^2=0.91)$ . A similar good fit was obtained combining data from O3, F3 and R1; see *Figures 3<sup>31</sup>* and 6.

The correlation for the accumulation factor is  $^{32}$  (see *Appendix 22*)

$$f = ax_a \qquad \qquad Eq. (A12-1)$$

The accumulation of C-14 from the energy production may be written as

<sup>&</sup>lt;sup>30</sup> For the anion capacities of CCU resins, see *Appendix* 34 in *Part* 2 of this report.

<sup>&</sup>lt;sup>31</sup> The regression in *Figure 3* is vs. the parameter anion equivalents/condensate amount cleaned.

<sup>&</sup>lt;sup>32</sup> The justification for choosing Eq. (A12-1) is the following: If you use no CCU resin at all ( $x_a$ =0) there will be no accumulation of C-14.



where

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$$Q = Eax_a \qquad \qquad Eq. (A12-2)$$

which may be simplified to

$$Q = an_a P \qquad \qquad Eq. (A12-3)$$

f	accumulation factor of C-14, Bq/MWh <sub>th</sub>
Q	accumulation of C-14, Bq
E	energy production, MWh <sub>th</sub>
$n_a$	number of anion equivalents used
Р	thermal power of the unit, MW <sub>th</sub>
Ν	<i>E/P</i> , number of EFPY
$x_a = n_a/N$	anion equivalents/h
a	coefficient (slope) for $x_a$ in the regression,
	(Bq/anion equivalents)/ MW <sub>th</sub>
а	anion

*Eq.* (A12-3) can be interpreted in the following way: The accumulation is proportional to the product of the anion equivalents used and the thermal power (= the production rate of C-14) of the reactor. The factor a may be interpreted as a modification factor accounting for conditions in the process system and in the resin handling.

Following this, a systematic test of regressions for all data sets available was performed in *Appendix 22. Table 14* shows a summary for all regression analyses made on accumulation factor data.

The regression is good for O3  $(1)^{33}$ , R1 (2), O3, F3 and R1 (3), O1+O2 (4)<sup>34</sup> and O1+O2 together with F1+F2 (8). For the last one, the organic fraction was too high for resins from F1+F2. In the application, a reduced organic fraction of 1.7 %, obtained in sample analyses, will be used for F1+F2. These results are used to estimate the historical accumulations of C-14. The procedure is outlined in *Appendix 23*.

	Regression from	Coefficient	a, (Bq/anion	equiv.)/MW <sub>th</sub>	Org.	Standard deviation			Used for
Nr	Unit	Total	Inorganic	Organic	fract.	Total	Inorganic	Organic	
1	03	8.84E+02	8.71E+02	1.30E+01	1.5%	8.8%	8.8%	18%	O3, F3
2	R1	7.89E+02	7.71E+02	1.83E+01	2.3%	13%	13%	18%	R1
3	O3 F3 R1	8.23E+02	8.06E+02	1.69E+01	2.1%	7.2%	7.4%	11%	B1+B2; O1+O2 before 2001
4	01+02	2.94E+02	2.72E+02	2.23E+01	7.6%	16%	17%	33%	O1+O2 from 2001 onwards
8	01+02 F1+F2	3.17E+02	2.72E+02	5.47E+00	1.7%	10%	11%	41%	F1+F2 with adj. org. fract.

Table 14. Summary of regression of accumulation factors vs. x<sub>a</sub>-parameter for BWR CCU.Regressions 5-7 are not used. See Appendix 22 for details. (X-1, sheet Table).

The normalization to EFPH can be interpreted in two ways: Either a normalization to the production of C-14 or to the amount of condensate cleaned<sup>35</sup>.

*Figure 3* shows the plot of the accumulation factor vs. the amount of anion equivalents<sup>36</sup> normalized to the condensate amount cleaned, parameter  $x_c$ . The regression function and the

<sup>&</sup>lt;sup>33</sup> Numbers in parenthesis refer to the regressions in *Table 14*.

<sup>&</sup>lt;sup>34</sup> Batch means; for individual samples the regression is poor (7).

<sup>&</sup>lt;sup>35</sup> For a given plant configuration, with respect to FP or no FP, the amount of condensate cleaned is proportional to EFPH.



regression coefficient are the same as for the  $x_a$ -regression. The calculation details can be found in *Appendix 22*.



Figure 3. Regression of the accumulation of C-14 in BWR CCU resins as a function of the parameter  $x_c$ , anion equivalents/kg condensate cleaned. (A-2, sheet D3 f).

These two regression models may be used to estimate the historic and future accumulation of C-14 in CCU resins when the use of CCU resins is known, as an alternative to the average factors expressed as Bq/MWh<sub>th</sub>. Se *Appendix 23* for calculation details. In Part 2 of this report, the normalization to EFPH will be used, since this information is easily available for the estimation of historical accumulations.

The regression also implies that the less CCU anion equivalents are used, the less C-14 will be accumulated. However, the use of CCU resin is governed by the chemistry specifications for the reactor to keep feed water and reactor water sufficiently clean and not to minimize the accumulation of C-14. Hopefully, a compromise between these two purposes may be possible.

#### 12.2. PWR RWCU Resins (Appendix 5)

The regression analysis described for BWR CCU resins has also been applied to PWR RWCU resins; see *Appendix 5, Chapter A5.4.10* for full details. The regression coefficients are lower than for BWR CCU, but still good; see *Table 15. Figure 4* shows the regression for the organic fraction from all samples from all PWR units.

				In-		Organic	Std. Dev.	Std. Dev.	Std. Dev.
Reactor	RWCU	Unit	Total	organic	Organic	fraction	Total	Inorg.	Org.
R2-R4	Regression	(Bq/anion equivalents)/MWh <sub>th</sub>	5.79E+03	3.82E+03	2.00E+03	34%	14%	18%	10%
	1-param	r <sup>2</sup>	0.74	0.64	0.85				

Table 15. Summary of regression of accumulation factors vs. the parameter  $x_a$  for PWR RWCU. (R234-1, sheet Accrat).

<sup>&</sup>lt;sup>36</sup> All C-14 species are assumed to be bound to the anion resin.





Figure 4. Regression of accumulation of organic C-14 vs. the parameter  $x_a$ : anion equivalents normalized to the energy production in EFPH. (R234-1, sheet D regr x org).

This fit indicates that there is a saturation effect with respect to organic C-14. A saturation effect is also supported by the concentration data in RCS discussed in *Appendix 5*.

The amount of inorganic C-14 shows the largest variations; probably due to the chemical conditions in the end of the fuel cycle and during the shut-down procedure. The main purpose of the correlation is to corroborate that there seems to be a saturation effect with respect to organic C-14 in the RWCU resins.

#### 13. Discussion

#### 13.1 General

The results obtained in this study comprise many individual samples (5-173) per unit or pair of units) collected over some 5-7 years; cf. *Table 4* and *Summary Table 3*. The larger number of samples and the longer time period covered are factors favoring the results of the present study, compared to *Ref. 4*. However, there are still differences between samples collected at different times, both for PWRs and BWRs.

#### 13.2. Comparison with Previous Results by Magnusson et. al. (Ref. 4)

*Table 16* shows a comparison between the results from this study and those from Magnusson et. al. The previous results have been corrected for dry substance contents and body-feed of inert materials, corrosion products (and for PWR) loading with lithium and boric acid.



		This	study		Ref. 4, c	orrected fo	r dry substa	nce and boo	dy-feed
Unit	Accumula	tion, % of p	production	Organic	Accumula	ition, % of j	production	Organic	Sample
	Total	Inorganic	Organic	fraction	Total	Inorganic	Organic	fraction	from
F1 + F2	0.93%	0.92%	0.016%	1.7%	5.1%	4.3%	0.82%	16%	F1*
F3	2.6%	2.5%	0.044%	1.7%	1.7%	1.3%	0.40%	24%	F3
F1+F2 dried	0.0068%	0.00062%	0.0062%	91%					
F3 dried	0.031%	0.0022%	0.029%	93%					
01+02	0.49%	0.47%	0.026%	5.2%	0.50%	0.42%	0.080%	16%	B2
03	2.3%	2.2%	0.032%	1.4%	1.7%	1.3%	0.40%	24%	F3
(B1+) B2	4.3%	4.2%	0.088%	2.1%	0.50%	0.42%	0.080%	16%	B2
R1	3.8%	3.7%	0.11%	2.8%	0.50%	0.42%	0.081%	16%	B2
PWR	3.3%	2.3%	1.0%	30%	5.4%	3.9%	1.5%	29%	R2+R3
* The sample	es analyze	d in Ref. 4	were colle	cted early i	in the hand	dling chain	which over	estimated	
the concer	ntration, co	ompared to	o this study	,					

Table 16. Comparison of the accumulation values in wet resins from Condensate Clean-up(BWR) and Reactor Water Clean-up (PWR) as % of production from this study and correctedvalues from Ref. 4. (A-2, sheet SUMTAB).

Some values from this study are higher than from Magnusson et. al., while others are lower. This pattern can be explained by

- 1 All units were not sampled in the previous study. E.g. unit B2 was used as reference plant for O1, O2 and R1. F3 was used as reference plant for O3
- 2 This study has analyzed substantially more samples covering longer operating periods covering start-up, operation and shut-down
- 3 The accumulation in BWR condensate clean-up resins depends on the parameter  $x_a$ , as described above
- 4 The organic fractions found in this study are usually lower than previously found; however, the PWR units show almost the same values. A possible explanation for the scatter in the BWR values may be the low concentrations in the resins. The organic fractions for the PWR resins show less scatter than for the BWR resins.
- 5 The samples from F1 were collected early in the handling chain, which overestimated the concentration, compared to this study, where the samples have been collected prior to drying and solidification

#### 13.3. CCU Resins from BWR units

Figure 5 summarizes the results for CCU resins from the BWR-units.

**F1 and F2**: The values obtained for <u>wet resins</u> are appreciably lower than those reported in *Ref.* 4. The concentrations of C-14 (mainly the inorganic fraction) in the resin samples vary over the period sampled. The organic concentration shows less variation. There is no obvious explanation for this, but it might be an indication on variations in air bubbling or sampling. The best estimate accumulation is 0.93 %. *Table* 6 gives a numerical overview of the various calculation models used, together with standard deviations. The variations in concentration may be found in *Figure A1-4*. The <u>dried resins</u> have appreciably lower concentrations, and



contain some 90 % organic C-14. The values for dried resins are most likely representative for the resins going to SFR1.



Figure 5. Summary of accumulation of C-14 in wet CCU resins (blue bars, left axis) and dried resins (black bars, right axis) from the present report. (A-2, sheet D BWR tot% all).

**F3**: The <u>wet resins</u> from F3 show less scatter in the concentration values than those from F1 and F2; *Figure A2-4. Table 7* gives a numerical overview of results, together with standard deviations. The accumulation of C-14 amounts to 2.6 %, and is somewhat higher than the value obtained for its sister plant O3. The <u>dried resins</u> have appreciably lower concentrations, but contain some 90 % organic C-14. The values for dried resins are most likely representative for the resins going to SFR1.

**O1 and O2**: The accumulation values obtained for O1 and O2 are at the low end of the range. The values for individual tanks scatter between almost 0 and 2.7 % with an average around 0.49 %. *Table 8* gives a numerical overview of the four sampling campaigns together with standard deviations.

**O3**: The results from O3 are based on resins used in 2009 to 2014. The accumulation is 2.3 %; somewhat lower than the value for its sister plant F3. *Table 9* gives a numerical overview of the values for the tanks sampled, together with standard deviations.

**B1 and B2**: The accumulation values obtained for B1 and B2 are recalculated from *Ref. 3* and 4. The accumulation is 0.87 %, about 50 % higher than for O1+O2. The value may be too low, since the samples were collected close to the surface of a concrete tank, where some of the C-14 may have been volatilized. An upper bound of 4.3 % is obtained, using the  $x_a$ -correlation. The organic accumulation factor is almost identical for the two calculation models.

**R1**: The sampling for R1 also includes sampling of initial and final amounts of resin in the storage tank, enabling the calculation of mass balances for separate fuel cycles. The results for



R1 are in the high end of the BWR range. The results for 2009 and 2013-2014 differ almost a factor of five from the 2009 results, peaking at 7.3 %. The average value is 3.8 %. *Table 11* gives a numerical overview of the values for the individual cycles, together with standard deviations. *Figure A4-32* shows that the *total* and *inorganic concentrations* are fairly stable over time. However, the *organic concentrations* show more variation.

Actually, the points used for the regression in *Figure 2* indicate that the  $x_a$ -value is the most important factor governing the accumulation.

**Remaining organic C-14 on the resins after extraction:** For the BWR, the organic fraction is in the range 1-5 %, which is lower than most of the results reported in *Ref. 4*. The reason for this difference is unclear. Several resin samples, representing all units and resin types have been combusted to release any C-14 remaining in the resin after the wet extraction. All samples combusted have had <5 % of the organic fraction left. See *Appendix 17* for details.

#### Explanation to (some of) the variations in C-14 accumulation

The variations in accumulation may be explained by the use of different amounts of CCU anion resin to clean the condensate, and the parameter  $x_a$ , as described in the previous chapter. Actually, the variations in accumulation factors can be explained by two main sets of regression parameters:

One set for resins without efficient air bubbling; O3, F3 and R1 (also to be used for B1+B2)



One set for resins with efficient air bubbling; O1+O2 and F1+F2

Figure 6. Summary of regression input data and the two main regression lines based on O3, F3 & R1 and O1+O2 and F1+F2, respectively. (X-1, sheet D 2 tot).

*Figure 6* shows all input data points and the two main regression lines. The sample from the back-flushing tank in F1, prior to air bubbling, with an appreciably higher accumulation factor than the resins in the F3 WHB, indicates the efficiency of a vigorous air bubbling.



These regression models may be used as an alternative model to estimate historic and future accumulations of C-14, based on the value of the parameter  $x_a$ . The correlation has been performed for total, inorganic and organic C-14, respectively.

The reason why O1+O2 and F1+F2 fall below the regression line is probably due to the air bubbling used to homogenize spent resin slurry stored in tanks. The carbon dioxide in the air lowers the pH-value in the weakly buffered water. The pH-value is sufficiently low to allow carbonate on the resins to be converted to carbon dioxide, which is released to the water and blown away with the air.

However, tests at Ringhals 1 did not show any significant release of C-14 when the Condensate Clean-up resins were air bubbled. The reason for this is most likely that the ratio of air to resin mass is lower than for F1+F2 and O1+O2 or a higher pH-value. O3 used air bubbling only intermittently. F3 has no provisions for air bubbling in its storage tank.

The large variations between samples for O1+O2 make it likely that air bubbling and the pH-value in the storage tank water are factors contributing to a low accumulation of C-14.

The releases of C-14 to air during shut-down strongly indicates that there is a substantial release from the resins in the WHB. See *Appendix 18* for the full text. *Table 17* in the following chapter shows data supporting this explanation.

#### 13.4. PWR units

One important result is the recalculation of the results from *Ref. 4*, based on a too high resin density. This recalculation, also including a correction for dry weight determination in the samples, reduces the accumulation to some 80 % of the previously reported values.

The values obtained in this study show a scatter between the units; R4 has the highest values and R2 and R3 the lowest. This may be explained as an effect of differences in operational and chemical parameters, mainly pH, in the reactor coolant and the RWCU. After the SG replacement in 2011, R4 has operated at the same pH-value as R2 and R3. *Appendix 20* discusses the stability of various C-14 compounds by means of Pourbaix diagrams. In *Appendix 5* the following possible sources for the variations between units and cycles are discussed:

1) The sampling procedure. The sampling procedure may be one factor; the "fishing" with a liquid scintillation vial in a transport vessel without stirring may yield varying proportions between cation and anion resins. The anion resin has a slightly lower density than the cation resin. There is a possibility that the anion resin has been enriched in the top part of the transport vessel prior to the sampling, leading to an overrepresentation of anion resin, which will overestimate the accumulation of  $C-14^{37}$ .

On the other hand: If the majority of the anion resin is in one or a few transport vessels, it may lead to an underrepresentation of the anion resin, leading to un underestimation of the accumulation. To account for these possibilities, the PWR samples have been assigned a sampling uncertainty of 30 %, which dominates the uncertainty in the accumulation for single years. However, summing all the accumulations reduces the uncertainty to some 10 %.

<sup>&</sup>lt;sup>37</sup> C-14 is assumed to be bound to anion resin.



**3)** Low concentration of inorganic C-14 (carbonate system) in the RWCU in the first few months of a fuel cycle. It may apply to R2 in 2012 with only four months of operation. See *Appendix 20* for a more thorough discussion.

4 (Non)-saturation of the resin with organic (acids)<sup>38</sup> C-14. It may also apply to R2 in 2012 with only four months of operation. This concept is supported by the correlation for the organic accumulation shown in *Figure 5*.

**5) Cold shut-downs followed by extensive boron dilution during restart.** It may apply to R3 in 2011.

6) The amount of resin in the RWCU. This behavior seems to be of minor importance. However, cf. point 10) below.

7) High pH in the RWCU system at the end of a fuel cycle. It depends on the extent of coast-down operation and the final boron concentration in the RC.

**8**) **Exposure of the RWCU resin to acidic and oxidizing conditions at shut-down.** It depends on when the operational RWCU resin is taken out of operation and replaced by the shut-down RWCU resin. It may differ between the units and from cycle to cycle.

**9)** Possible formation of inorganic C-14 from organic acids accumulated on the ion exchanger by decarboxylation or oxidation by the radicals formed by irradiation of water. This is a speculative explanation, but possible, since there is a strong radiation field within the spent ion exchange resin during operation. Yim and Caron (*Ref. 11*), hold this as a possible alternative.

<sup>&</sup>lt;sup>38</sup> In R3 and R4 formate, acetate and oxalate have been detected in the concentration range 1-10 ppb during operation and > 100 ppb during shut-down and start-up transients. See *Appendix 20* and *Attachment File R234-3* with data from the Ringhals Chemistry Data Base.





Figure 7. Summary of the accumulation of C-14 in PWR RWCU resins from the present study and corrected values from Ref. 4 (2001-2003). (R234-1, sheet D New Summary).

#### 10) The amount of anion equivalents normalized to EFPH (energy production);

**parameter**  $x_a$ . *Chapter 12.2* shows a fairly good correlation for the organic accumulation to the parameter  $x_a$ . Effects of the high pH-value in the RWCU at the end of a cycle with coast-down or the exposure to acidic conditions during shut-down may be the factors reducing the accumulation.

*Figure 7* shows a summary of the results for the PWR units, where the variations also may be seen. *Table 6* gives the numerical values including the standard deviations. The accumulation for all three units is 3.3 % with an organic fraction of 30 %, which is in good agreement with the previous results. Taking credit for the release of C-14 at shutdown from the R1 WHB (see next chapter), the accumulation may be some 20 % lower, or some 3.0 % of the production.

#### 13.5 Release of C-14 from Ion Exchange Resins in Storage Tanks

**General.** For some units there are large variations in the concentrations of inorganic C-14 (carbonate system); see *Appendix 1* for F1+F2 and *Appendix 3* for O1+O2. The reason for this is not quite clear, but air bubbling of the contents in the storage tanks is a plausible explanation. The addition of powder resin from the WPCU system may also effect a dilution, which may explain part of the variations.

*Appendix* 8 discusses this topic in detail; it also shows that the simple organic acids will remain on the resins during air bubbling, which also has been found in most samples, even those with a low carbonate concentration.

#### 13.5.1.01+02.

*Figure* 8 shows pH-values measured in the storage tanks at  $O1+O2^{39}$ . The first pK<sub>a</sub>-value<sup>40</sup> (6.37) for the carbonate system is also indicated. As many as 18 values out of 23 are below

<sup>&</sup>lt;sup>39</sup> Measurement of pH-values in the range 5 to 8 is very difficult in the weakly buffered waters in the storage tanks.



the  $pK_a$ -value, which supports the explanation given above. Of these values, eleven are within 0.2 pH-units from the theoretical value for water in equilibrium with carbon dioxide in air, 5.5.

On the other hand, two values are above 7.1, where non-volatile hydrogen carbonate is the dominating species. Such variations in the pH-value may explain the scatter in measured concentrations of C-14. These higher pH-values may be caused by floor drain water, containing detergents. See first footnote in *Appendix 3* for details.

The "bubbling effect" is supported by the release rate of C-14 to the air when the reactor is shut down. *Appendix 18* analyzes the C-14 releases from O1 and R1 during long shut-downs, where the only source for releases of C-14 comes from ion exchange resins in storage tanks<sup>41</sup>. *Table 17* shows a summary of releases from O1 in 2011-2013. For O1 the release rate is about nine times higher than the measured accumulation rate in CCU resins. The only reasonable source for this release of C-14 must be the tanks holding the CCU resins from O2 in the WHB of O1. There is probably a fairly large uncertainty in the factor nine, since there are uncertainties in the measured releases as well as in the calculated production rate. However, *Table 17* gives a strong indication that there is a release from spent resins stored in the WHB collection tanks.



*Figure 8. pH-value in storage tanks for powder resin at O1+O2, August-September 2011 (A-7, sheet D pH O12).* 

The accumulation results given in *Table 8* are not affected by these releases, since the sampling is done when the resins are transferred to the concrete tanks for SFR1.

<sup>&</sup>lt;sup>40</sup> For "carbonic acid": when the pH-value is below the  $pK_a$ -value, it will almost completely decompose into gaseous carbon dioxide and water; see *Appendix 8* for details.

<sup>&</sup>lt;sup>41</sup> The WHB in O1 handles all resins from O1 and O2. The ventilation air from the storage tanks is released via the O1 main stack. P. Arvidsson, OKG, Pers. Comm. November 2013.



		С	-14 Releas	e	C-14 ca	lc. Prod	No-op. release,		
		Inorganic	Organic	Total			Fraction of O2		
Start	End	Bq	Bq	Bq	01, Bq	02, Bq	production		
2011-11-01	2013-04-15	9.93E+09	1.23E+10	2.23E+10				Incl. O1 re	actor op.
2011-11-01	2013-04-02	8.77E+09	1.20E+10	2.08E+10				O1 no ope	eration
2011-11-01	2013-04-15	1.17E+09	3.40E+08	1.51E+09	2.53E+09	4.41E+11		From read	tor operat.
2011-11-01	2013-04-15			1.98E+10			4.5%	Release - o	alc. O1 prod.
Accumulati	on in CCU a	fter releas	e to air, fro	m Table 6			0.49%		
Fraction rel	eased of ori	ginally acc	umulated	C-14			90%		

Table 17. Summary of C-14 releases from O1 during shut-down November 2011-April 2013.(O12-6, sheet Summary).

The information in *Figure 8* and *Table 17* supports that the variations in the C-14 concentrations in the CCU resins in O1+O2 is due to the air bubbling and the pH value in the water. Furthermore, the CCU resins are also mixed with resins from the WPCU and, in some cases, fresh powder resins to reduce the activity of fission and/or corrosion products in the water.

#### 13.5.2. R1-R4.

The samples of PWR resins are taken when they arrive in the WHB of R1. In the WHB the resins are stored in a tank which is regularly air bubbled. A study of the release rates from R1during shut-down (*Table 18*), similar to that for O1 above, has yielded the results in *Table 18*. It includes all years from 2002 to 2013. The long shut-down in 2009-2010 is of special interest, since there were no spent CCU resins from R1 in storage during that period (see *Appendix 18* for details). Since R1 has a simple delay system for the off-gases without charcoal columns<sup>42</sup>, there are no other sources for releases of C-14 than the resins in storage. The release rate in 2009-2010 has been compared with the average production rate in the PWR units in the period 2002-2012. It amounts to 0.83 %. It means that there most likely is a reduction in the accumulation values for the PWR units in *Table 13* from 3.3 % to some 2.5 %-3.0 %, i.e. some 25 % lower.

The accumulation in the solidified CCU resins from R1 is not expected to have any appreciable reduction from the values in *Table 11*, based on the two last rows in *Table 18*.

Unit	Release rate during shut- down	Inorg. C-14, Bq/s	Organic C-14, Bq/s	Total C-14, Bq/s	Orga- nic fraction
	Mean 2002-2013	3.81E+02	6.69E+01	4.48E+02	15%
	2009-2010	2.03E+02	3.28E+01	2.36E+02	14%
		Fraction	of produc	tion rate	
		Inorg.	Org.	Total	
R1-R4	Mean 2002-2010	0.95%	0.17%	1.1%	15%
R2-R4	2009-2010**	0.72%	0.12%	0.83%	14%
** No (	CU resins in tan	ks			

Table 18. Summary of C-14 releases from R1 during shut-down periods.(R1-5, sheet Rel rate).

 $<sup>^{42}</sup>$  *Ref. 10* describes situations in F1 and F2 where carbon dioxide is significantly accumulated in the off-gas delay system with charcoal columns.



# 14. Variations between Fuel Cycles and Uncertainty in Calculated Values

			To	otal accu	imulatio	n	Or	ganic acc	umulatio	n			
				% of pro	duction			% of pro	duction		Concer	ntration	
						Accus				Accu-			
						mula-				tion		Conc.	
		Num-				tion				Orga-	Conc.	Orga-	
		ber				Total				nic	Total	nic	
		of cy-				Max/				Max/	Max/	Max/	
Unit	Period	cles*	Mean	Max	Min	Mean	Mean	Max	Min	Mean	Mean	Mean	Comment
F1 + F2	2009-2013	10									3.0	3.0	Not possible to keep resins from
F3	2009-2014	3.5									2.5	1.8	different cycles separated
01+02	2008-2013	8	0.49%	0.63%	0.30%	1.3	0.026%	0.087%	0.0090%	3.4			
03	2009-2014	5	2.3%	5.3%	0.72%	2.3	0.032%	0.12%	0.011%	3.6			
R1	2009-2015	7	3.8%	7.3%	1.7%	1.9	0.11%	0.15%	0.056%	1.4			
R2	See	7	3.1%	5.8%	1.7%	1.9	0.93%	2.9%	0.53%	3.1			2003 (2 cy); 2008; 2010; 2012-2014
R3	Comment	7	2.7%	5.4%	1.4%	2.0	0.79%	1.6%	0.30%	2.0			2003; 2008; 2009; 2011-2014
R4	See	5	4.4%	8.5%	1.5%	1.9	1.3%	2.2%	0.49%	1.7			2009; 2011-2014
PWR	Comment	19	3.3%	8.5%	1.4%	2.6	1.0%	2.9%	0.30%	3.0			One sample per cycle
* Cvcles	included in	n accum	nulation	calculati	ions: for	F1 + F2	some resi	ns were i	produced	before	the sam	pling ca	mpaign

Table 19. Summary of Mean, maximum and minimum values for total and organic accumulation, together with the ratios Max/Mean in BWR-CCU and PWR-RWCU.
"Accumulation" refers to accumulation in a limited amount of resin, which can be related to a certain operational period. This calculation was not possible to perform for F1+F2 and F3, since the collection tanks never were emptied during the sampling period. As a substitute, the variations in concentrations have been used. For further information, please see the text. (A-2, sheet MEANMAX).

The variations between the fuel cycles have already been discussed in the previous paragraph. *Table 19* shows the mean, maximum and minimum values for the accumulation over the fuel cycles studied. The number of cycles in the study is also included. For the units F1+F2 and F3 it is not possible to attribute the accumulation to separate cycles. The reason for this is the handling of resin; the storage tanks for the resin have not been emptied between the cycles (see *Appendix 1, 2* and *14*). As a substitute, the ratios between the maximum and the mean concentration have been calculated. For the PWRs, a grand mean including all the units has also been calculated.

In general, the ratios Max/Mean for the accumulations are between 1 and 2. The concentration ratios are larger for F1+F2 and F3 than the accumulation ratios for the other units. It is more difficult to explain, but a possible influence from the sampling has been discussed previously.

For the PWRs, the largest variation is in the organic accumulation, where R2 has the highest value in 2012. The reason for this is discussed in *Appendix 5* and in the chapter "Discussion".

*Table 20* also shows the standard deviations for the accumulation factors given in *Table 19*. The organic factors usually have higher standard deviations than the inorganic. This is mainly due to the lower activity in the organic samples and the resulting larger standard deviation in the activity counting. In some cases, the scatter between the subsamples determines the standard deviation, rather than the compounded standard deviation in the counting of the



subsamples. The standard deviations also include a 10 % standard deviation in the amount of resins.

The standard deviations are in the range 3-13 % for the inorganic factors and 3-20 % for the organic factors, except for B1+B2 and dried resins from FKA. *Table 20* also shows that the BWR units have been sampled for altogether 33.5 fuel cycles, corresponding to 22.4 Equivalent full Power Years (EFPY). The PWR units have been sampled for 19 cycles corresponding to 14.7 EFPY.

Despite the variations between individual fuel cycles, the most important feature for the SFR1 repository is the accumulated activity over time. For the units where the variations between the cycles are easy to follow (O1, O2, O3, R1, R2, R3 and R4), there is a tendency that the average stabilizes over the time period studied, according to "the law of large numbers".

In general, the ratios Max/Mean for the accumulations are between 1 and 2. The concentration ratios are larger for F1+F2 and F3 than the accumulation ratios for the other units. It is more difficult to explain, but a possible influence from the sampling has been discussed previously.

For the PWRs, the largest variation is in the organic accumulation, where R2 has the highest value in 2012. The reason for this is discussed in *Appendix 5* and in the chapter "Discussion".

		Number									
	Sampling	of		Std.dev.	Std.dev.	Std.dev.					
Unit	period	cycles*	EFPY	Total	Inorg.	Org	Comment				
F1 + F2	2009-2013	10	8.38	13%	13%	14%	Wet resin; ***				
F1 + F2	2009-2013	10	8.38	22%	110%	24%	Dried resin; ***				
F3	2009-2014	3.5	3.63	4.9%	4.9%	7.2%	Wet resin; ***				
F3	2009-2014	3.5	3.63	15%	43%	17%	Dried resin; ***				
01+02	2008-2013	8	3.79	2.3%	2.3%	9.5%					
03	2009-2014	5	2.90	2.9%	2.9%	8.1%					
B1+B2	**	**	**	13%	16%	23%					
R1	2009-2015	7	3.68	6.5%	5.2%	27%					
BWR		33.5	22.4								
R2	See	7	5.22	19%	22%	17%	2003 (2 cy); 2008; 2010; 2012-2014				
R3	Comment	7	5.33	14%	14%	13%	2003; 2008; 2009; 2011-2014				
R4		5	4.12	17%	17%	16%	2009; 2011-2014				
PWR		19	14.7	8.4%	8.9%	7.7%	One sample per cycle				
BWR + PW	/R	52.5	37.1								
* Cycles in	* Cycles included in accumulation calculations; for F1 + F2 some resins were produced before the sampling campaigr										
** Not possible to relate the samples to a specific operating period, due to post-operational sampling											
*** Not p	ossible to k	eep resins	from diff	erent cycle	es separate	ed					

Table 20. Summary of Equivalent Full Power Years (EFPY) and standard deviations in the calculated accumulation factors. (A-2, sheet STDEV).

*Table 20* also shows the standard deviations for the accumulation factors given in *Table 19*. The organic factors usually have higher standard deviations than the inorganic. This is mainly due to the lower activity in the organic samples and the resulting larger standard deviation in the activity counting. In some cases, the scatter between the subsamples determines the standard deviation, rather than the compounded standard deviation in the counting of the subsamples. The standard deviations also include a 10 % standard deviation in the amount of resins.



The standard deviations are in the range 3-13 % for the inorganic factors and 3-20 % for the organic factors, except for B1+B2 and dried resins from FKA.

*Table 20* also shows that the BWR units have been sampled for altogether 33.5 fuel cycles, corresponding to 22.4 Equivalent full Power Years (EFPY). The PWR units have been sampled for 19 cycles corresponding to 14.7 EFPY.

Despite the variations between individual fuel cycles, the most important feature for the SFR1 repository is the accumulated activity over time. For the units where the variations between the cycles are easy to follow (O1, O2, O3, R1, R2, R3 and R4), there is a tendency that the average stabilizes over the time period studied, according to "the law of large numbers".

### **15. Accumulation Factors**

*Table 21* summarizes the accumulation factors obtained in this study. For PWR, the corrected results from *Ref. 4* are included in the mean values in *Table 21*. The factors are expressed as a percentage of the production and as Bq/MWh<sub>th</sub>. The latter approach is the best one, since there are some uncertainties associated with the calculated production rates (see footnote to *Table 2*). The standard deviations in the values are shown in *Table 20*. The factors are also separated for total, inorganic and organic C-14.

		Accumu	lation, % of	production	Accumulation, Bq/MWhth				
Unit(s)	System	Total	Inorganic	Organic	Total	Inorganic	Organic		
F1+F2	332	0.93%	0.92%	0.016%	2.30E+02	2.26E+02	3.97E+00		
F3	332	2.6%	2.5%	0.044%	6.12E+02	6.01E+02	1.06E+01		
F1+F2	332	0.0068%	0.00062%	0.0062%	1.67E+00	1.53E-01	1.52E+00		
F3	332	0.031%	0.0022%	0.029%	7.45E+00	5.25E-01	6.92E+00		
01+02	332	0.49%	0.47%	0.026%	1.03E+02	9.74E+01	5.29E+00		
03	332	2.3%	2.2%	0.032%	4.15E+02	4.09E+02	5.95E+00		
B1+B2	332	4.3%	4.2%	0.088%	8.54E+02	8.36E+02	1.76E+01		
R1	332	3.8%	3.7%	0.11%	8.64E+02	8.40E+02	2.43E+01		
R2+R3+R4	334	3.3%	2.3%	1.0%	4.95F+02	3.50F+02	1.46F+02		

Table 21. Summary of accumulation factors for C-14 in CCU resins for BWR and in RWCUand all resins for PWR. The yellow-marked values are for dried resins in F1+F2 and F3.(A-2, sheet FactorsSFR).

In Appendix 33, a summary is given of the accumulation of C-14 in the resins analyzed.

#### **19. Acknowledgements**<sup>43</sup>

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<sup>&</sup>lt;sup>43</sup> Chapters 16, 17 and 18 are in Part 2.



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2	<b>Magnusson</b> $\mathbf{\dot{A}}$ : <sup>14</sup> C produced by nuclear power reactors –	ISBN: 978-91-628-7248-9
	generation and characterization of gaseous, liquid and	
	solid waste. Lund University, Doctoral thesis (2006)	
3	Magnusson Å, Stenström K: <sup>14</sup> C produced in Swedish	SKB R-05-78
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	exchange resins, various process water systems and ejector	
	off-gas. Swedish nuclear fuel and waste management	
	company (SKB) (2005)	
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### **21. Attachment Files**

Att. File	Title	File type
A-1	Sammanställning C14-Projekt TSkorr	Excel 97-2003
A-2	Summary C-14 rev 3	Excel 2007
A-3	Provhistorik maj 2013	Excel 97-2003
A-4	Energiproduktion	Excel 97-2003
A-5	Sampling rev 15	Power Point 2007
A-6	Composition of standard deviation rev 1	Excel 2007
A-7	pH-studie	Excel 97-2003
A-8	carbonate system pH-concdiagram	Excel 97-2003
A-9	TS jämförelse omg3	Excel 97-2003
A-10	Combustion	Excel 2007
A-11	Myrsyra test	Excel 2007
B-1	NOT USED	
B-2	BKAB rev 2	Excel 2007
F12-1	F12 C-14 ack	Excel 97-2003
F12-2	Logg FKA backspolning	Excel 97-2003
F12-3	F12332massor rev 2	Excel 97-2003
F12-4	Studiebesök på Forsmark 2011	Word 2007
F12-5	Forsmark filters väg till prov	Power Point 2007
F12-6	Tavelbild från Forsmark 20111130	ipg
F12-7	RAV2012.xlsm	Excel 97-2003
F12-8	Kopia av Loggböcker F3 TD42 TC56 TC58 2012	Excel 97-2003
F12-9	FKA drving rev2.	Excel 2007
F3-1	F3 C-14 ack	Excel 97-2003
F3-2	C-14 i avfallsskorsten	Excel 2007
F3-3	KOKILLER med c-14mätning i frånluft	Excel 2007
FKA-1	Fortsatt Provtagningsprogram för Forsmark	Word 97-2003
FKA-2	Nya loggböcker för Forsmark	Word 97-2003
FKA-3	Logbok T42-T43	Excel 97-2003
FKA-4	Loggbok F3 TC56	Excel 97-2003
FKA-5	Loggbok F3 TD42	Excel 97-2003
012-1	Provtagning O1 O2	Excel 97-2003
012-2	O12 C-14 ack 1	Excel 97-2003
012-3	O12 del 2	Excel 97-2003
012-4	O12 C-14 ack 2	Excel 97-2003
012-5	O12 C-14 ack3	Excel 2007
012-6	C-14 utsläpp från O1 och O2 arbex3	Excel 2007
012-7	O12 C-14 ack 4	Excel 2007
03-1	O3 original	Excel 2007
03-2	O3 C-14 ack	Excel 97-2003
03-3	O3-332original	Excel 2007
03-4	O3 KRA PO Aronsson	Excel 2007
R1-1	R1 T41-Provtagningslogg C14	Excel 97-2003
R1-2	R1 C-14 ack 1	Excel 97-2003
R1-3	R1 C-14 ack 2	Excel 2007
R1-4	R1 C-14 ack 3	Excel 2007
R1-5	R1 C-14 luft arbetsex	Excel 2007
R1-6	RAB thermal energy	Excel 2007
R234-1	PWR C-14 rev 7	Excel 97-2003
R234-2	pH Summary may 2004 PWR	Excel 97-2003
R234-3	ORGANISKA SYROR RC PWR	Excel 97-2003
		2.1001 > 1 2002



X-2	Appendix 33	Excel 2007
X-3	Concentrations	Excel 2007
X-4	Accplot	Excel 2007
H-1	PWR historik rev 9	Excel 2007
H-2	R1 historik rev 5	Excel 2007
H-3	O3 historik rev 5	Excel 2007
H-4	O1 historik rev 5	Excel 2007
H-5	O2 historik rev 3	Excel 2007
H-6	F1 historik rev 2	Excel 2007
H-7	F2 historik rev 1	Excel 2007
H-8	F3 historik rev 2	Excel 2007
Y-1	JBmassor kapacitet	Excel 2007
Y-2	Rapportformulär rev 6	Excel 2007
Y-3	S KB databas rev 1	Excel 2007



## APPENDIX 1 Results from F1 and F2: Accumulation in Wet CCU Resins

## A1.1. Resin Handling

All calculations for F1 and F2 are included in an excel-book. It can be found as *Attachment File F12-1* to this report. Reference to the various sheets will be made at tables, figures etc.



*Figure A1-1. Overview of handling of spent powder resin at F1 and F2.* <sup>44</sup> (A-5, pict. 8).

In the Condensate Clean-up system (CCU) of F1 and F2 (10 filters per unit), 80 kg powder resin (carboxylic resin in cation part) + 10 kg of inert material is loaded and operated for periods usually ranging from 6 to 16 weeks.

When the resin is exhausted, it is flushed via a temporary storage tank in the unit (back-flushing tank) to either of two tanks in the unit F1 WHB, 342T42 and T43. These tanks are mixed by air bubbling. From these tanks, the resin is transferred in a shielded transport vessel (T80 with a stirrer) via tank 343T2 (with a stirrer) to tank TC56<sup>45</sup> (with a stirrer) in the WHB at F3. A schematic overview is given in *Figure A1-1*. The resins are stored in 342T42 and -T43 for 1-2 years before transport to the F3 WHB.

From the storage tank TC56 (continuously stirred) resin is brought to a dryer/evaporator, where the concentration of dry solids is increased to a proper value before solidification in bitumen. The dryer/evaporator also receives concentrate from the evaporation of waste waters, i. a. floor drains. This concentrate may also contain some C-14; however, this activity is not covered by this study. In connection with the transfer from TC56 to the dryer, a sample is withdrawn. It is also possible to take a sample without a transfer for solidification. (This text is also found in *Appendix 14*).

<sup>&</sup>lt;sup>44</sup> Christina Lillfors-Pintér: Report from visit to FKA 2011-11-30. See Attachment file F12-4, -5 and -6.

<sup>&</sup>lt;sup>45</sup> Tank TC56 is not the normal storage tank for resins from F1+F2. Normally these resins are mixed with the corresponding resins from F3. However, TC56 was used during the sampling campaign to keep the F1+F2 resins separated from the F3 resins. Annelie Jansson, FKA, personal communication, November 2015.



## A1.2. Mass Balance for Spent Resins

The record describing the additions and removals of resin in TC56 was delivered as pdfcopies of a handwritten log book until mid-2011. The data in the log have been transferred to an *excel-sheet TC56* in *Attachment File F12-1*. From mid-2011 data have been delivered in excel-sheets. The record for resin handling in T42 and T43 are described in *Appendix 14*.

All resin masses added to T42, T43 and TC56 are given as dry weight (dw), including inert filter material and accumulated corrosion products. On the other hand, calculations based on annual consumptions of CCU resins have to be corrected for added inert filter material and accumulated corrosion products. *Table A1-1* shows a summary of such an accumulation, which yields a single correction factor, here called the Body-Feed Factor (BFF), which is used to multiply the mass of powder resin to obtain the total dry weight, including inert filter material and corrosion products. *(Sheet BodyfeedF12)*.

The additions to the storage tanks T42 and T43 in unit F1 are based on an estimate of the total dry substance in the back flushed resins, while the removals to F3 WHB are based on measured total dry substance, i.e. including corrosion products<sup>46</sup>. This procedure may sometimes lead to inconsistencies in the mass balances.

				Resin +
				inert+
				corr.
		Fresh	resins	Prod.
		Fe3O4	Fe2O3	
Fe	atomic mass	56	56	
0	atomic mass	16	16	
Iron oxide	mole mass	232	160	
Iron oxide/Fe		1.381	1.429	
Fe in condensate	ppb	10	10	
ionic fraction		0.1	0.1	
Volume filtered	m3	1.20E+06	1.20E+06	
Iron oxide	kg	14.9	15.4	
Ionic Fe	kg	1.2	1.2	
Iron oxide	kg	1.49E+01	1.54E+01	
Ionic Fe	kg	1.20E+00	1.20E+00	
Powder resin	kg dw/ filter	80	80	
Inert material	kg dw/ filter	10	10	
Powder res.+Inert. Mater.	kg dw/ filter	90	90	
Total	kg dw/ filter	1.06E+02	1.07E+02	
Fraction (iron oxide+ionic	Fe)	1.52E-01	1.56E-01	
Fraction powder resin		7.54E-01	7.50E-01	
Total Body-Feed Factor (B	FF)	1.33	1.33	1.00

Table A1-1. Calculation of the correction for iron oxide accumulation (BFF).(F12-1, sheet BodyfeedF12).

*Figure A1-2A* shows the additions, removals and inventories in the CCU resin storage tanks T42 and T43 at F1+F2. A more comprehensive description of the data underlying *Figure A1-2A* is given in *Appendix 14*. The data may be found in *Attachment file F12-4*. From these data it can be seen that both tanks T42 and T43 were simultaneously empty on 2007-05-07. Further, T43 also was empty on 2009-04-02. Accordingly, the "power history" will start on 2007-05-05, since all the resins in T42, T43 and TC56 have been added after 2007-05-07. It means that the production of C-14 has to be calculated from this date. Since

<sup>&</sup>lt;sup>46</sup> Pers. Comm., Lena Eriksson, FKA§ May 2013.



the filter operational time is at least one month, the production of C-14 in the resins analyzed, is assumed to start on 2007-04-01.



*Figure A1-2A. Additions, removals and inventory in the CCU resin storage tanks at F1+F2. (F12-3, sheet D A14-5).* 



Figure A1-2B. The log data for resin storage tank TC56 in graphics. The calculated inventory in early September 2009 is slightly negative, due to uncertainties in the added amounts. The strong negative values from April 2010 are due to missing additions in the logs. The peaks in the curve "Corrected inventory" in April 2009 and September 2010 are corrections introduced to bring the additions to TC56 in agreement with the removals from T42+T43. The automatically recorded level in TC56 is in good qualitative agreement with the "Corrected inventory". (F12-1, sheet D TC56; data are in Sheet TC56).

*Figure A1-2B* shows the additions, removals and the inventory in TC56 for the period 2009-2012. In addition, the samples withdrawn for analyses are also indicated. 13 samples were



collected; however, one was too small to analyze. The inventory is given as total dw, which is used throughout in the calculations below. Due to insufficient record keeping, this diagram has not been updated after November 2011.

The inventory of TC56 based on the logs from the WHB turns slightly negative in April 2010. The negative inventory increases through 2010. Since solidifications continue, the inventory must be under-estimated. The most likely reason is that additions have not been logged. A full discussion of this is done in *Appendix 14*, where appropriate corrections also are introduced.

The corrected inventory shown in *Figure A1-2B*, is based on the record for T42+T43 at F1+F2. Additional information, supporting this correction, is found in the automatically recorded level in TC56, which shows a good qualitative agreement with the corrected inventory. The record keeping is further discussed in section "Discussion" below.

## A1.3. Calculation Models

From *Figure A1-3* below, it is obvious that it is not possible to perform a correct Mass Balance calculation since it requires that the whole inventory of resin is completely mixed. This requirement is not fulfilled, since three separate tanks (T42, T42 TC56) are involved, and it is only the last one in the handling chain, TC56, which is sampled.

To compensate for this, five calculation models may be used:

- 1) Mass Balance over T42, T43 and TC56, using all samples from TC56
- 2) Mass Balance over T42, T43 using TC56, using the initial samples from TC56 with the highest concentration (batches F12-09B1, -09B2 and -09B3) (NOT USED)<sup>47</sup>.
- 3) Annual use of CCU resins for the years 2008-April 2012, using Maximum Concentration
- 4) Annual use of CCU resins for the years 2008- April 2012, using Mean Concentration
- 5) Annual use of CCU resins for the years 2008- April 2012, using Minimum Concentration



Figure A1-3. Transport pathways and storage tanks for CCU resins from F1 and F2. All transports are intermittent. The storage tank TC56 has continuous mixing by stirring. In 2012 TC58 was used as a temporary storage volume in order to maintain storage capacity in T42 and T43 (A-5, pict. 13).

In early January 2009 the tank TC56 was assumed to be empty. Then resin was added from the units F1 and F2 and subsequently removed for solidification. In connection with the solidifications (but not all of them) samples were withdrawn for analysis. Since the tank

<sup>&</sup>lt;sup>47</sup> This calculation model was used only for preliminary calculations.



content is mixed by stirring (but not air bubbling), it is assumed that the content is fairly homogeneous.

*Table A1-2* and *Figure A1-4* show the concentrations obtained in the analyses of the batched samples. These concentrations are used to calculate the quantities of C-14 brought to solidification.



Figure A1-4. Concentrations of C-14 used for calculation of the quantities of C-14 in the powder resins. Some of the samples are from tank TC58; see Table A1-2 below. (F12-1, sheet D F12conc).

#### A1.3.1. Model 1: Calculation using all the samples from TC56

There is a large variation in the concentrations of C-14 in the resin samples. It is not clear whether the variations take place in T42 and T43 or just in TC56. Since the samples were taken only from TC56, these concentrations are not necessarily those prevailing in T42 and T43.

Nevertheless, a mass balance calculation is attempted for the period starting 2007-04-01 to 2012-04-30. Even if this estimate is wrong by a few months, it will give only a marginal effect on the calculated accumulation of C-14, expressed as a percentage of the production or as  $Bq/MWh_{th}$ .



Analysis data								
				Org,			SD	
		Total,	Inorg,	Bq/kg	0	SD	inorg,	SD
Batch	End date	Bq/kg aw	Bq/kg aw	aw	Org, %	τοτ, %	%	org, %
F12-09B1	2009-03-28	1.08E+06	1.07E+06	1.11E+04	1.1%	17%	17%	25%
F12-09B2	2009-05-15	9.45E+05	9.33E+05	1.26E+04	1.3%	1.6%	1.6%	25%
F12-09B3	2009-08-05	1.78E+06	1.76E+06	1.86E+04	1.0%	3.2%	3.1%	28%
F12-10B2	2010-07-10	3.26E+04	3.29E+03	2.93E+04	90%	5.2%	27%	5.7%
F12-10B3	2010-08-31	7.12E+03	3.10E+03	4.02E+03	58%	20%	37%	25%
F12-10B4	2010-10-18	6.98E+03	2.19E+03	4.79E+03	71%	24%	50%	19%
F12-2	2012-05-05	2.58E+06	2.55E+06	3.02E+04	1.2%	1.1%	1.1%	3.7%
F12-6 TC58	2012-05-23	4.40E+06	4.36E+06	3.93E+04	0.89%	1.1%	1.1%	23%
F12-3	2012-06-13	3.01E+06	2.96E+06	4.82E+04	1.7%	1.1%	1.2%	2.6%
F12-5	2012-06-24	3.12E+06	3.04E+06	7.68E+04	2.5%	1.1%	1.1%	2.2%
F12-4	2012-07-12	1.76E+05	1.62E+05	1.39E+04	7.9%	1.4%	1.4%	6.1%
F12-11	2012-08-02	2.08E+04	7.95E+03	1.28E+04	62%	13%	20%	9.9%
F12-7 TC58	2012-08-27	3.73E+04	2.62E+04	1.12E+04	31%	22%	29%	9.4%
F12-1	2012-08-29	6.15E+05	6.00E+05	1.44E+04	2.3%	1.2%	1.2%	7.1%
F12-8	2012-10-29	5.91E+04	4.43E+04	1.47E+04	25%	2.8%	2.9%	7.1%
F12-9	2012-12-10	3.30E+06	3.26E+06	3.42E+04	1.0%	1.1%	1.1%	3.6%
F12-10	2013-01-20	3.90E+06	3.84E+06	6.07E+04	1.6%	3.3%	3.5%	12%
Mean		1.47E+06	1.45E+06	2.57E+04	1.7%	103%	104%	78%
Geom. mean		3.52E+05	2.46E+05	1.90E+04	5.4%			
Max. conc		4.40E+06	4.36E+06	7.68E+04	1.7%	1.1%	1.1%	2.2%
Min conc.		6.98E+03	2.19E+03	4.02E+03	58%	24%	50%	25%
Median conc.		9.45E+05	9.33E+05	1.47E+04	1.6%			
Number of points	17	17	17	17				
Mean conc from kokil	ler/tank	1.44E+06	1.42E+06	2.49E+04	1.7%	13%	13%	14%

Table A1-2. Concentration data used in the calculations.  $(F12-1, sheet F12acc model 1)^{48}$ .

The production of C-14 in F1 and F2 has been calculated from the production rate given in the FSAR and the production of thermal energy obtained from the operational reports. The energy production is summarized in sheets *F1energy* and *F2energy* and in the lower sections of *Table A1-3*, which also summarizes the results for this calculation model. The model accounts for the resins being solidified from TC56 and remaining in T43+T42, respectively. The resin amount solidified corresponds to the average production in some 3 years. The remaining inventory in T42+T43 corresponds to the average production for some 2 years.

<sup>&</sup>lt;sup>48</sup> Samples withdrawn for other purposes indicate that the concentration of the resins being back-flushed is around 1E7 Bq/kg dw with an organic fraction of about 1 %. (*A-1, sheet FKAX+SKB*).



Calculated amo	ounts of C-14						Resin	uncertainty	10%
ourounded unit		-					Realit	uncertainty	1070
Kokill/tank	Batch	Date		kg dw	% of total	Total Bq	Inorg Bq	Org Bq	
F3091702	F12-09B1	2009-03-11		842	3.6%	9.12E+08	9.02E+08	9.32E+06	
F3091703	F12-09B1	2009-03-18		890	3.8%	9.63E+08	9.54E+08	9.86E+06	
F3091704	F12-09B1	2009-03-28		883	3.8%	9.56E+08	9.46E+08	9.78E+06	
F3091705	F12-09B1	2009-04-13		592	2.5%	6.41E+08	6.34E+08	6.56E+06	
F3091706	F12-09B1	2009-04-20		444	1.9%	4.81E+08	4.76E+08	4.92E+06	
F3091707	F12-09B2	2009-05-03		592	2.2%	5.60E+08	5.52E+08	7.44E+06	
F3091708	F12-09B2	2009-05-15		473	1.8%	4.47E+08	4.41E+08	5.95E+06	
F3091709	F12-09B3	2009-08-05		566	4.0%	1.01E+09	9.96E+08	1.05E+07	
F3091710	F12-09B3	2009-09-02		566	4.0%	1.01E+09	9.96E+08	1.05E+07	
F3101708	F12-10B2	2010-05-23		610	0.079%	1.99E+07	2.01E+06	1.78E+07	
F3101709	F12-10B2	2010-07-10		627	0.081%	2.04E+07	2.07E+06	1.83E+07	
F3101710	F12-10B2	2010-07-13		620	0.080%	2.02E+07	2.04E+06	1.81E+07	
F3101713	F12-10B3	2010-08-31		631	0.018%	4.49E+06	1.95E+06	2.54E+06	
F3101715	F12-10B4	2010-09-22		686	0.019%	4.79E+06	1.50E+06	3.28E+06	
F3101716	F12-10B4	2010-10-18		640	0.018%	4.47E+06	1.40E+06	3.06E+06	
F3101717	F12-10B4	2010-11-16		735	0.020%	5.13E+06	1.61E+06	3.52E+06	
F3121702	F12-4	2012-07-18		571	0.399%	1.01E+08	9.26E+07	7.95E+06	
F3121703	F12-1	2012-08-06		690	1.7%	4.24E+08	4.14F+08	9.91E+06	
F3121704	F12-1	2012-08-30		651	1.6%	4.00E+08	3.91E+08	9.35E+06	
E3121706	F12-8	2012-11-01		647	0.15%	3.82E+07	2 87E+07	9.54E+06	
F3121708	F12-9	2012-12-10		616	8.1%	2.03E+09	2.01E+09	2 11E+07	
TC56 remainder	F12-9	2012-12-10		3887	60%	1.52E+10	1 49F+10	2.36E+08	
Sum		2012 12 10		17459	100.0%	2 52E+10	2 48F+10	4 36F+08	
Inventory in F1-	-342T42+T43	at end of pe	riod	16710	100.070	2.022110	2.402110	4.002100	11%
Correction facto	or for invent	orv in F1-342	T42+T43	1.96		4 94F+10	4.85E+10	8.53E+08	1170
% of production	2007-04-01	2012-04-30	1421140	1.00		0.93%	0.92%	0.016%	
Ba/MWhth	2007-04-01	2012-04-50				2 30F+02	2 26F+02	3 97F±00	
% organic						2.002102	2.202102	1 7%	
% standard dev	iation					13%	13%	1.17	
Body Feed Fac	tor	1.00				Includes un	certainty in in	ventory of T42	2+T43
				C-14	Body-		Equivalent	Equivalent	
Production	Start	End	MWhth	Bq/MWh thermal	feed factor	C-14, Bq total	Hours, (EFPH)	Years, (EFPY)	
F1+F2	2007-04-01	2008-12-31	79657363			1.96E+12	27205	3.11	
	2009-01-01	2009-06-30	22912785			5.63E+11	7825	0.89	
	2009-06-30	2010-06-30	33569293			8.25E+11	11465	1.31	
	2010-07-01	2012-04-30	78813153			1.94E+12	26917	3.07	
F1+F2 Sum	2007-04-01	2012-04-30	214952594			5.29E+12	73413	8.38	
F1	2007-04-01	2008-12-31	41856702	2 46F+04		1.03E+12	14295	1.63	
	2009-01-01	2009-06-30	11501105	2.46E+04		2.83E+11	3928	0.45	
	2009-07-01	2000 00 00	23867156	2.46E+04		5.87E+11	8151	0.40	
	2010-07-01	2012-04-30	40062529	2.46E±04		9.85E±11	13683	1.56	
F1 Sum	2007-04-01	2012-04-30	117287492	2.46E+04		2.88E+12	40057	4 57	
	2007 04 01	2012 04 00	117201402	2.402104		2.002112	40007	4.07	
F2	2007-04-01	2008-12-31	37800661	2 46F±04		Q 30F±11	12010	1 47	
	2009-01-01	2000-12-01	11411680	2.40E+04		2 81F±11	3807	0.44	
	2009-07-01	2010-06-30	9702137	2.46E±04		2.30 = 11	3314	0.38	
	2010-07-01	2012-04-30	38750624	2.40E+04		0 53E±11	13235	1 51	
F2 Sum	2007-04-01	2012-04-30	97665102	2.46E±04		2 40E±12	33356	3.81	

*Table A1-3. Summary of C-14 in powder resins at F12 produced in the period April 2007-April 2012. Calculation according to Mass Balance Model. (F12-1, sheet F12acc model 1).* 

# A1.3.2. Model 3, 4 and 5: Calculation using the Mean, Maximum and Minimum concentration in TC56

With the discontinuous transfer of resins from T42+T43 to TC56, a third calculation model has been used. This model simply uses the mean, maximum and minimum values of the concentrations in the samples analyzed. These concentrations are multiplied with the total



consumption of powder resins for the years 2008-april 2012. A BFF = 1.33 has been applied here to account for inert material and corrosion products; see *Table A1-1* for a calculation of the BFF.

The calculations are summarized *in Table A1-4A*, *B and C*, respectively, where the calculated annual accumulations for the years 2008-2010 are presented.

Body-fee	d factor	1.33	Resin unce	ertainty	10%	Mean	Mean	Mean	Mean	Mean	Mean	Mean
				Calc. Produc-	Total				% of			
Produc-				tion Bq	resin				produc-	Total	Inorg	Org
tion	Start	End	MWhth	total	mass, kg	Total Bq	Inorg, Bq	Org Bq	tion	Bq/MWhth	Bq/MWhth	Bq/MWhth
F1+F2	2008-01-01	2008-12-31	4.14E+07	1.02E+12	5940	1.17E+10	1.15E+10	2.03E+08	1.1%	2.82E+02	2.77E+02	4.92E+00
	2009-01-01	2009-12-31	3.92E+07	9.64E+11	5371	1.06E+10	1.04E+10	1.84E+08	1.1%	2.69E+02	2.64E+02	4.69E+00
	2010-01-01	2010-12-31	3.47E+07	8.54E+11	5985	1.18E+10	1.16E+10	2.05E+08	1.4%	3.39E+02	3.33E+02	5.90E+00
	2011-01-01	2012-04-30	6.14E+07	1.51E+12	9248	1.82E+10	1.79E+10	3.17E+08	1.2%	2.96E+02	2.91E+02	5.16E+00
Mean/												
Sum	2008-01-01	2012-04-30	1.77E+08	4.34E+12	26544	5.22E+10	5.12E+10	9.09E+08	1.2%	2.95E+02	2.90E+02	5.15E+00
% organ	ic							1.7%				
% standa	ard deviation	n				104%	105%	78%				

Table A1-4A. Summary of C-14 in powder resins at F12 produced in the period 2008-2012, based on the consumption of powder resins and Mean Concentration. Mean Concentration Model. (F12-1, sheet F12accmodel345).

Body-fee	d factor	1.33	Resin unce	ertainty	10%	Max	Max	Max	Max	Max	Max	Max
				Calc. Produc-	Total				% of			
Produc-				tion Bq	resin				produc-	Total	Inorg	Org
tion	Start	End	MWhth	total	mass, kg	Total Bq	Inorg, Bq	Org Bq	tion	Bq/MWhth	Bq/MWhth	Bq/MWhth
F1+F2	2008-01-01	2008-12-31	4.14E+07	1.02E+12	5940	3.48E+10	3.45E+10	6.08E+08	3.4%	8.41E+02	8.34E+02	1.47E+01
	2009-01-01	2009-12-31	3.92E+07	9.64E+11	5371	3.15E+10	3.12E+10	5.50E+08	3.3%	8.03E+02	7.95E+02	1.40E+01
	2010-01-01	2010-12-31	3.47E+07	8.54E+11	5985	3.51E+10	3.48E+10	6.13E+08	4.1%	1.01E+03	1.00E+03	1.76E+01
	2011-01-01	2012-04-30	6.14E+07	1.51E+12	9248	5.42E+10	5.37E+10	9.47E+08	3.6%	8.83E+02	8.75E+02	1.54E+01
Mean/												
Sum	2008-01-01	2012-04-30	1.77E+08	4.34E+12	26544	1.56E+11	1.54E+11	2.72E+09	3.6%	8.80E+02	8.73E+02	1.54E+01
% organi	c							1.7%	Max. value is max. for each : tot, inorg,			norg, org
% standa	rd deviation	n				10%	10%	10%	%			

Table A1-4B. Summary of C-14 in powder resins at F12 produced in the period 2008-2012, based on the consumption of powder resins and Maximum Concentration. Mean Concentration Model. (F12-1, sheet F12accmodel345).

Body-fee	d factor	1.33	Resin unce	ertainty	10%	Min	Min	Min	Min	Min	Min	Min
Produc-				Calc. Produc- tion Bq	Total resin				% of produc-	Total	Inorg	Org
tion	Start	End	MWhth	total	mass, kg	Total Bq	Inorg, Bq	Org Bq	tion	Bq/MWhth	Bq/MWhth	Bq/MWhth
F1+F2	2008-01-01	2008-12-31	4.14E+07	1.02E+12	5940	5.53E+07	1.74E+07	3.18E+07	0.0054%	1.34E+00	4.20E-01	7.69E-01
	2009-01-01	2009-12-31	3.92E+07	9.64E+11	5371	5.00E+07	1.57E+07	2.88E+07	0.0052%	1.27E+00	4.00E-01	7.34E-01
	2010-01-01	2010-12-31	3.47E+07	8.54E+11	5985	5.57E+07	1.75E+07	3.21E+07	0.0065%	1.60E+00	5.04E-01	9.23E-01
	2011-01-01	2012-04-30	6.14E+07	1.51E+12	9248	8.61E+07	2.70E+07	4.95E+07	0.0057%	1.40E+00	4.41E-01	8.07E-01
Mean/												
Sum	2008-01-01	2012-04-30	1.77E+08	4.34E+12	26544	2.47E+08	7.76E+07	1.42E+08	0.0057%	1.40E+00	4.39E-01	8.05E-01
% organi	с							58%	Min. valu	e is min. for	each : tot, ir	norg, org
% standa	rd deviatio	า				26%	51%	27%	6			

Table A1-4C. Summary of C-14 in powder resins at F12 produced in the period 2008-2012, based on the consumption of powder resins and Minimum Concentration. Mean Concentration Model. (F12-1, sheet F12accmodel345).



Produc- tion	Start	End	MWhth	Calc. Produc- tion Bq total	Total resin mass, kg	332-resin, kg	324-resin, kg	342- resin, kg
F1	2008-01-01	2008-12-31	20754453	5.10E+11	4305	4305	275	150
	2009-01-01	2009-12-31	22681798	5.58E+11	4830	4830	175	135
	2010-01-01	2010-12-31	23961956	5.89E+11	4410	4410	275	195
	2011-01-01	2012-04-30	28787036	7.08E+11				
	Start	End	MWbth	Calc. Produc- tion Bq total	Total resin	332-resin,	324-resin,	342- resin ka
F2	2008-01-01	2008-12-31	20630832	5.07E+11	3780	3780	200	180
	2009-01-01	2009-12-31	16539635	4.07E+11	4095	4095	325	195
	2010-01-01	2010-12-31	10755992	2.64E+11	2835	2835	325	165
	2011-01-01	2012-04-30	32568814	8.01E+11				

Table A1-4D. Annual energy production and resin use for the years 2008-2010.(F12-1, sheet F12acc model 345).

## A1.4. Discussion

The record keeping for the resins handled during the period has been incomplete. Using other ways of information (personal interviews, record of activity measurements of waste containers; *Attachment file F12-1, sheet TC56*) has filled the gaps in an acceptable way. Since the tank inventory turns negative when the solidifications continue in 2010, several additions are obviously missing. Therefore, data from T42 and T43 have been used as supplement to obtain a reasonable mass balance.

For the calculations based on annual resin consumption, delivery data from the plant warehouse to the units F1 and F2 have been used<sup>49</sup>.

*Table A1-5* and *Figure A1-5* give a summary of the accumulation of C-14 in the CCU resins based on the different calculation models used. With the structure of the resin handling, it is not possible to give a general mass balance for F1+F2, as for the other BWR units. *Table A1-5* has columns for "Calculation Model" 7 and 8, where

- 7) Calculations from *Ref.* 4 corrected for BFF and ds
- 8) One sample collected from a back-flushing tank, representing one filter

<sup>&</sup>lt;sup>49</sup> Supplied by E. Karlsson FKA, April 2013. (*F12-1, sheet pulvermassor*). The values used in version 4 onwards differs from those used in version 3.



Calculation Model	1	3	4	5	7	8
	Best		Inter-	Тоо		Compari-
Classification	estimate	High	mediate	Low***	Too high	son
				Minimum		
	Mass	Maximum	Mean	conc. +		Back-
	balance	conc. +	conc. +	annual	Ref. 4 +	flushing
	TC56+	annual	annual	resin	BFF + ds-	tank (one
F1+F2	T43+T42	resin use*	resin use	use**	corr.	filter)
Equivalent Full Power Hours, F1	40057					218
Equivalent Full Power Hours, F2	33356					
Equivalent Full Power Years, F1	4.57					0.025
Equivalent Full Power Years, F2	3.81					
Body-Feed Factor, Average (BFF)	1.00	1.33	1.33	1.33	1.33	1
Dry Solids Correction Factor in Sample					1.04	
Organic fraction	1.7%	1.7%	1.7%	58%	16%	0.70%
Total accumulation in CCU	0.93%	3.6%	1.2%	0.0057%	5.1%	7.7%
Organic accumulation in CCU	0.016%	0.063%	0.021%	0.0033%	0.82%	0.054%
Total Bq/MWhth	2.30E+02	8.80E+02	2.95E+02	1.40E+00	1.26E+03	1.90E+03
Inorganic Bq/MWhth	2.26E+02	8.73E+02	2.90E+02	4.39E-01	1.06E+03	1.89E+03
Organic Bq/MWhth	3.97E+00	1.54E+01	5.15E+00	8.05E-01	2.02E+02	1.34E+01
Total Std. Dev.	13%	10%	104%	26%		11%
Inorganic Std. Dev.	13%	10%	105%	51%		11%
Organic Std. Dev.	14%	10%	78%	27%		26%
*Max. value is maximum for each : tot,	inorg, org					
**Min. value is minimum for each : tot	, inorg, org					
*** Close to dried resin (two samples o	only)					

Table A1-5. Comparison of accumulation of C-14 in CCU resins, obtained by the different calculation models from this study, and Ref. 4. The Rightmost column shows data for freshly back-flushed CCU resin as a comparison. (F12-1, sheet Summary).

**Model 1** using the **Mass Balance over T42+T43+TC56 using all concentration values** may be considered **as the Best Estimate for the accumulation before drying and solidification**. The reason for this is that it is based on the samples taken before solidification. However, there are some variations in the concentrations that are not well understood. The fraction accumulated, based on this calculation model is 0.93 % with a standard deviation of 13 %.

## Model 2 using the Mass Balance over T42+T43+TC56 using samples with the three highest concentrations was used for preliminary calculations.

**Model 3** summarizes the results using the **Maximum Concentration** in TC56 and the annual consumption of CCU resin in the years 2008- April 2012. The fraction accumulated, based on this calculation model is 3.6 % with a standard deviation of 10 %. This may be considered **as the Upper Bound for the accumulation.** 

**Model 4** summarizes the results using the **Mean Concentration** in TC56 and the annual consumption of CCU resin in the years 2008- April 2012. The fraction accumulated, based on this calculation model is 1.2 %. This may be considered **as an intermediate value for the accumulation.** The standard deviation is high, 104 %, due to the large span between the maximum and minimum concentrations.



**Model 5** summarizes the results using the **Minimum Concentration** in TC56 and the annual consumption of CCU resin in the years 2008-April 2012. The fraction accumulated, based on this calculation model is 0.0057 %. This may be considered **as a too low estimate for the accumulation.** However, based on analysis of two samples from the resin dryer, it may be close to the value for the solidified resins. See *Appendix 21* for details of dried samples.

Model 6 summarizes the results from Ref. 4. (Not shown in Table A1-5).

Model 7 summarizes the results from *Ref. 4*, corrected for BFF and ds in analysis. It has the second highest values, next to the comparison sample.

**Model 8** is a comparison sample, collected in the back-flushing tank. It has the highest values, which is expected, since the sample has been collected early in the handling chain, before air bubbling.

It is noticeable that the accumulation rate for organic C-14 is fairly stable for calculation models 1, 3 and 4; it varies only between 4 and 15 Bq/MWh<sub>th</sub>, while the accumulation rates for inorganic C-14 varies between 226 and 873 Bq/MWh<sub>th</sub>. This variation is most likely due to stripping of carbon dioxide in air bubbling or tank stirring.

The standard deviation for accumulated activity includes the standard deviations in the samples and a 10 % standard deviation in the mass quantity.

The standard deviation in the Mass Balance calculations is based on the accumulated standard deviation for the samples.

The previous results (*Ref. 4*) were based on a small number of samples taken over a limited period of time, while the present results are based on some 24 samples collected during some 4 years. Furthermore, the previous samples were taken from the tank "Back-flushing tank" (*Figure A1-1*) where it had not been exposed to air bubbling and stirring<sup>50</sup>.

*Table A1-5* Model 8 shows data based on one sample withdrawn from the Back-flushing tank. This sample shows an accumulation even higher than what was found in *Ref. 4*, which supports that the previous value for the accumulation was too high.

The samples analyzed in this study have been withdrawn just before solidification, which means that they are representative for the amount of activity going to the resin dryer, prior to drying.

There are some peculiarities in the concentration values from this study; *Figure A1-4* and *Table A1-2* show that there is a decrease in the total concentration of C-14 from the samples collected in 2009 to those collected in 2010. In 2012 there are also such variations. Unfortunately, there is a period in between these two sampling periods, where no samples were taken. The reason for this was necessary campaigns for solidification of other resins.

Some of the resin samples may not be representative for the bulk of the resin; e.g. the extracted samples have in some instances been left in the sampling bottle connected to a slight underpressure, which may extract volatile species of C-14 (see *Appendix 21*, footnote before *Table A21-5*).

<sup>&</sup>lt;sup>50</sup> Christina Lillfors-Pintér. Report from visit to FKA 2011-11-30. See Attachment file F12-4, -5 and -6.



The concentration of the organic fraction does not decrease as much as the total (or actually, the inorganic fraction). The reason for this is unclear; one possibility might be air bubbling, which lowers the pH-value by introducing  $CO_2$  in the collection tanks T42 and T43 at F1.

*Appendix 21* presents analysis data form dried samples and release measurements of C-14 in the ventilation duct from the dryer. The two dried samples analyzed so far indicate a high release of C-14; see *Appendix 21* for details. The remaining accumulation in dried resins is included in *Figure A1-5* for comparison.



Figure A1-5. The accumulation rate of C-14 in CCU resins at F1+F2 for the different calculation models, including dried resins. NOTE: Logarithmic scale! (F12-1, sheet D Summary dried).



## APPENDIX 2 Results from F3: Accumulation in Wet CCU Resins

## A2.1. Resin Handling

All calculations for F3 are included in an excel-book. It can be found as *Attachment File F3-1* to this report. Reference to the various sheets will be made at tables, figures etc.



*Figure A2-1. Overview of handling of spent powder resin at F3*<sup>51</sup>. (A-5, pict. 4)

In the Condensate Clean-up system (CCU) of F3, powder resin (sulfonate resin in cation part) is loaded and operated for periods usually ranging from 6 to 16 weeks. During operation, a body-feed of inert fibers is added to the filters to improve the filtering properties. When the resin is exhausted, it is flushed to a temporary tank in the unit ("back-flushing tank"). From this tank, it is transferred to tank TD42 in the Waste Building.

The storage tank TD42 also receives spent powder resin from the Spent Fuel Pool clean-up system and the Waste Handling system. However, these additional amounts are much lower than the CCU resins. According to *Ref. 4*, these resins are not expected to contain substantial activities of C-14. A schematic overview is given in *Figure A2-1*.

Resin is brought from the storage tank TD42 to a dryer/evaporator, where the concentration of dry solids is increased to a proper value before solidification in bitumen. The dryer/evaporator also receives concentrate from the evaporation of waste waters, i. a. floor drains. This concentrate may also contain some C-14; however, this activity is not covered by this study.

<sup>&</sup>lt;sup>51</sup> Christina Lillfors-Pintér. Personal communication after visit to FKA 2011-11-30.



## A2.2. Mass Balance for Spent Resins

The record describing the additions and removals of resin was delivered as pdf-copies of a handwritten log book until mid-2011. The data in the log have been transferred to an *excel-sheet TD42* in *Attachment File F3-1*. From mid-2011 data have been delivered in excel-sheets

All resin masses added to TD42 are given as dry weight (dw) and do not include body-feed and accumulated iron oxides. Since the removals to solidification and samples are based on total dry weight, the additions of resin have to be corrected. The calculation is based on the addition of inert body-feed material and the accumulation of iron oxides. *Table A2-1* shows a summary of such accumulation, which yields a single correction factor, here called the Body-Feed Factor (BFF), which is used to multiply the additions of powder resin to obtain the total dry weight added (*Sheet BodyfeedF3*).

		Fe3O4	Fe2O3	Values fro
	Weeks/month	4.357	4.357	J-O Helme
Fe	atomic mass	56	56	
0	atomic mass	16	16	
Iron oxide	mole mass	232	160	
Iron oxide/Fe		1.381	1.429	
Fe in condensate	ppb	10	10	8-10
ionic fraction		0.1	0.1	
Volume filtered	m3	1.20E+06	1.20E+06	1.00E+06
Iron oxide	kg	14.9	15.4	
Ionic Fe	kg	1.2	1.2	
	kg/period & 7			
Body-feed	filters	30	30	30
Number of filters		7	7	7
Body-feed period	Weeks	1.429	1.429	10 d
Op. Time	Months	4.5	4.5	4
Op. Time	Weeks	19.607	19.607	
Body-feed/filter	kg	58.821	58.821	
Iron oxide	kg	1.49E+01	1.54E+01	
Ionic Fe	kg	1.20E+00	1.20E+00	
Body-feed	kg dw	58.821	58.821	
Powder resin	kg dw	70	70	70
Total	kg dw	1.45E+02	1.45E+02	
Fraction (iron oxide+ic	onic Fe)	1.11E-01	1.14E-01	
Fraction body-feed		4.06E-01	4.04E-01	
Fraction powder resin		4.83E-01	4.81E-01	
Total Body-Feed Facto	r (BFF)	2.07E+00	2.08E+00	

Table A2-1. Calculation of the correction for body feed and iron oxideaccumulation (BFF). (F3-1, sheet XBodyfeedF3).

The BFF has been used to calculate the mass balance over TD42 based on the log data for additions and removals.



*Figure A2-2* shows the additions, removals and the inventory in TD42. The inventory is given as total dw and powder resin dw, since the logs use both ways for record keeping. In the calculations below, the total dw is used throughout.



*Figure A2-2. The log data for resin storage tank TD42 in graphics. (F3-1, sheet D TD42; data are in Sheet TD42).* 

## A2.3. Mass balance for C-14 (Calculation model MBM)



Figure A2-3. Schematic mass balance over tank TD42. (A-5, pict. 12).

The Mass Balance Model (MBM) is used to calculate the amounts of C-14 disposed in solidified waste from the tank TD42. In early September 2008 the tank was empty. Then resin was added from the unit and removed for solidification. In connection with the solidifications (but not all of them) samples were withdrawn for analysis. Since the tank content is mixed by water injection, it is assumed that the content is fairly homogeneous. *Table A2-2* and *Figure A2-4* show the concentrations obtained in the analyses of the batched samples. These concentrations are used to calculate the quantities of C-14 brought to solidification. The long



period without samples is due to modification works in the WHB. During this period, no solidification and sampling could be done.



Figure A2-4. Concentrations of C-14 used for calculation of the quantities of C-14 in the CCU resins. (F3-1, sheet D F3conc).

Analysis data									
				Inorg,	Org,			SD	SD
			Total,	Bq/kg	Bq/kg		SD tot,	inorg,	org,
Batch	Start date	End date	Bq/kg dw	dw	dw	Org, %	%	%	%
F3-09B1		2009-09-22	2.41E+06	2.36E+06	5.17E+04	2.1%	2.1%	2.4%	12%
F3-10B1		2010-01-17	2.82E+06	2.75E+06	7.56E+04	2.7%	1.9%	2.0%	29%
F3-10B2		2010-05-01	4.74E+06	4.66E+06	8.17E+04	1.7%	4.5%	4.4%	16%
F3B3		2010-08-18	7.42E+05	7.08E+05	3.39E+04	4.6%	6.7%	7.0%	5.4%
F3B4		2010-09-04	2.91E+05	2.60E+05	3.03E+04	10%	4.3%	4.2%	6.9%
F3B5		2011-01-16	1.02E+06	9.81E+05	3.50E+04	3.4%	4.9%	5.2%	5.7%
F3-2		2011-03-05	1.40E+06	1.36E+06	3.95E+04	2.8%	9.3%	9.5%	2.7%
F3-1		2011-03-14	1.96E+06	1.93E+06	3.98E+04	2.0%	2.2%	2.1%	5.0%
F3-3		2011-05-11	1.20E+06	1.16E+06	3.84E+04	3.2%	3.4%	3.3%	5.3%
F3-4		2012-07-03							
F3-5		2012-10-11	4.59E+04	8.76E+03	3.72E+04	81%	3.2%	9.9%	3.2%
F12-6		2012-12-21	4.40E+06	4.36E+06	3.93E+04	0.89%	1.1%	1.1%	23%
Mean			1.91E+06	1.87E+06	4.57E+04	2.4%	78%	79%	36%
Geom. mean			1.15E+06	9.59E+05	4.33E+04	3.8%			
Max. conc			4.74E+06	4.66E+06	8.17E+04	1.7%	4.5%	4.4%	16%
Min conc.			4.59E+04	8.76E+03	3.03E+04	66%	3.2%	10%	6.9%
Median conc.			1.40E+06	1.36E+06	3.93E+04	2.8%			
Number of poin	ts	12	11	11	11				
Sample F3-4 too	Sample F3-4 too small for analysis								
Mean conc. from	n kokiller/ta	nk	2.85E+06	2.80E+06	4.94E+04	1.7%	4.9%	4.9%	7.2%

Table A2-2. Concentration data used in the calculations. (F3-1, sheet F3acc).<sup>52</sup>

<sup>&</sup>lt;sup>52</sup>Samples withdrawn for other purposes indicate that the concentration of the resins being back-flushed is around 5E6 Bq/kg dw with an organic fraction of about 1 %. (A-1, sheet FKAX+SKB).


Resin uncertain	ty			10.0%				
Calculated amo	ounts of C-14							
Kal-10/6a mla	Detak	Dette			0/ - 6 / - / - 1	Tatal Da		0 D
KOKIII/tank	Batch	Date		Kg dw	% of total		Inorg Bq	
F3091701	F3-09B1	2009-02-28		643	2.4%	1.55E+09	1.52E+09	3.32E+07
F3091711	F3-09B1	2009-09-22		726	2.7%	1.75E+09	1.71E+09	3.75E+07
F3091712	F3-09B1	2009-10-03		611	2.3%	1.47E+09	1.44E+09	3.16E+07
F3091713	F3-10B1	2009-12-12		560	2.5%	1.58E+09	1.54E+09	4.24E+07
F3091715	F3-10B1	2010-01-01		678	3.0%	1.91E+09	1.86E+09	5.13E+07
F3101701	F3-10B1	2010-01-13		653	2.9%	1.84E+09	1.79E+09	4.94E+07
F3101702	F3-10B1	2010-01-17		728	3.2%	2.06E+09	2.00E+09	5.51E+07
F3101703	F3-10B1	2010-02-27		656	2.9%	1.85E+09	1.80E+09	4.96E+07
F3101704	F3-10B1	2010-03-13		689	3.0%	1.95E+09	1.89E+09	5.21E+07
F3101705	F3-10B1	2010-03-27		682	3.0%	1.93E+09	1.87E+09	5.16E+07
F3101706	F3-10B2	2010-05-01		661	4.9%	3.14E+09	3.08E+09	5.40E+07
F3101707	F3-10B2	2010-05-08		592	4.4%	2.81E+09	2.76E+09	4.84E+07
F3101711	F3B3	2010-07-28		561	0.6%	4.16E+08	3.97E+08	1.90E+07
F3101712	F3B3	2010-08-18		627	0.7%	4.65E+08	4.44E+08	2.12E+07
F3101714	F3B4	2010-09-04		580	0.3%	1.69E+08	1.51E+08	1.76E+07
F3101718	F3B5	2011-01-16		715	1.1%	7.26E+08	7.02E+08	2.50E+07
F3111701	F3B5	2011-01-30		657	1.0%	6.68E+08	6.45E+08	2.30E+07
F3111703	F3-2	2011-03-06		627	1.4%	8.77E+08	8.52E+08	2.48E+07
F3111702	F3-1	2011-04-06		522	1.6%	1.03E+09	1.00E+09	2.08E+07
F3111704	F3-1	2011-04-06		632	1.9%	1.24E+09	1.22E+09	2.52E+07
F3111705	F3-3	2011-04-17		644	1.2%	7.72E+08	7.47E+08	2.48E+07
F3111706	F3-3	2011-05-21		628	1.2%	7.53E+08	7.28E+08	2.41E+07
F3121701	F3-3	2012-07-02		621	1.2%	7.44E+08	7.20E+08	2.39E+07
F3121705	F3-5	2012-09-30		469	0.0%	2.15E+07	4.11E+06	1.74E+07
F131709	F12-6	2012-12-21		596	4.1%	2.62E+09	2.60E+09	2.34E+07
TD42 remainder	F12-6	2012-12-21		6797	46.5%	2.99E+10	2.96E+10	2.67E+08
Sum				22555	100.0%	6.42E+10	6.31E+10	1.11E+09
% of production						2.6%	2.5%	0.044%
Bq/MWhth						6.12E+02	6.01E+02	1.06E+01
% organic								1.7%
% Standard dev	viation					4.9%	4.9%	7.2%
							Equivalent	Equivalent
							Full	Full
				C-14			Power	Power
				Ba/MWh			Hours.	Years.
	Start	End	MWhth	thermal		Bo total	(EFPH)	(EFPY)
Production	2008-08-01	2010-04-14	39289935	2.40F+04		9.43F+11	11906	1.36
Production	2010-04-14	2011-01-31	18905427	2.40F+04		4.54F+11	5729	0.65
	2011-02-01	2012-11-30	46745861	2.40F+04		1.12F+12	14165	1.62
Sum	2008-08-01	2012-11-30	1.05E+08			2.52E+12	31800	3.63

Table A2-3. Summary of C-14 in powder resins at F3 produced in the period August 2008-December 2012. (F3-1, sheet F3acc).

*Table A2-3* shows the calculated accumulated activity of C-14 in the solidified resins and the residual amount of resin in TD42.

The production of C-14 in F3 has been calculated from the production rate given in the FSAR and the production of thermal energy obtained from the operational reports. The energy production is summarized in sheet *F3 Energy*. *Table A2-4* summarizes the results. The accumulation in the resins amounts to 2.6 % of the calculated production. The organic fraction is 1.7 %. The production period starts in August 2008 and ends by the end of November 2012.





### A2.4. Discussion

The record keeping for the resins handled during the period seems to be sufficiently good with respect to additions and removals for solidification. The major uncertainty factor seems to be the Body-Feed of inert material. With a nominal addition rate of inert material, it will be almost as much inert material as resin. The iron oxides will be around 10 % of the total dw. For details, see *Table A2-1* and *Figure A2-5*. The remaining accumulation in dried resins is included in *Figure A2-5* for comparison.

The Body-Feed Factor, (BFF) is probably somewhat low, since some inventory values in TD42 are slightly negative in September 2010 and April-July 2011. This negative balance may lead to a slight underestimation of the residual resin mass and activity at the end of the sampling period.

The fraction accumulated, 2.6%, is higher than 0.7 % previously reported by Magnusson et. al. (*Ref. 4*). However, the results from Ref. 4 do not include the BFF and the ds-correction. Correction of the old value by BFF=2.08 and the ds-correction, yields an uptake of 1.7 %. On the other hand, the organic fraction found in this study, 1.7 %, is lower than 24 % in the previous one.

	Mass		
	balance		
	TD42	Ref. 4 +	O3 Sum
	Sept 2009-	BFF + ds-	090913-
F3	Dec 2012	corr.	140829
Equivalent Full Power Hours (EFPH)	31800		25380
Equivalent Full Power Years (EFPY)	3.63		2.90
Body-Feed Factor, Average (BFF)	2.08	2.08	1.12
Dry Solids Correction Factor in Sample		1.16	
Organic fraction	1.7%	24%	1.4%
Total accumulation in CCU	2.6%	1.7%	2.3%
Organic accumulation in CCU	0.044%	0.40%	0.032%
Total Bq/MWhth	6.12E+02	4.05E+02	4.15E+02
Inorganic Bq/MWhth	6.01E+02	3.08E+02	4.09E+02
Organic Bq/MWhth	1.06E+01	9.72E+01	5.95E+00
Total Std. Dev.	4.9%		2.9%
Inorganic Std. Dev.	4.9%		2.9%
Organic Std. Dev.	7.2%		8.1%

Table A2-4. Comparison of accumulation of C-14 in CCU resins from this study, Ref. 4 and O3. (F3-1, sheet Summary).

The standard deviation in the concentration values is the maximum of the scatter between the three subsamples in each batch and the standard deviation in the activity measurement. The uncertainty in the activity counting is mostly lower than this scatter. The resulting standard deviation for the whole period is composed by the uncertainty in the concentration values and a 10 % uncertainty in the resin mass. The uncertainty is 4.9 % for the total activity and 7.2 % for the organic fraction. It is the compounded standard deviation for the activity contents in all the moulds.

The previous results were based on a small number of samples taken over a limited period of time, while the present results are based on some 14 samples collected over 51 months. Furthermore, the mass of the resin in this study is based on actual "production" (although corrected for calculated body-feed). The previous results were based on an estimated annual consumption of resin and did not account for the BFF and the ds-correction. Furthermore, the resins analyzed in this study cover an operation period with 3.63 EFPY (Equivalent Full Power Years). The total and organic accumulation in F3 resins is higher than the values obtained for its sister plan O3 (*Appendix 13*). This may be due to no air bubbling in TD42. O3 uses air bubbling, at least intermittently.



Figure A2-5. Comparison of accumulation of C-14 in CCU resins from this study, including dried resins, Ref. 4 and O3. (F3-1, sheet D Summary dried).

There is no obvious explanation for the discrepancy in the organic fraction between *Ref.* 4 and the present results. The present value is in the range 1-5 %, which is obtained for most BWR samples in this study.

There are some peculiarities in the concentration values from this study; *Figure A2-4* and *Table A2-2* show that there is a decrease in the concentration of inorganic C-14 in the samples collected in the latter part of 2010, in early 2011 and in October 2012, compared with the earlier and later samples. The concentration of the organic fraction does not decrease as much as the inorganic fraction. The reason for this is unclear, since there is no air bubbling in the storage tank. A possible explanation for the variations may be that the sampling bottles have been left in the sampling position for varying times; since there is a low underpressure, C-14 compounds may have been released<sup>53</sup>.

<sup>&</sup>lt;sup>53</sup> Annelie Jansson, FKA, personal communication August 2013 and *Ref. 9*.







# APPENDIX 3 Results from O1 and O2

# A3.1. Resin Handling

All calculations for O1 and O2 are included in excel-books. They can be found as *Attachment Files* to this report. Reference to the various sheets will be made at tables, figures etc.



Figure A3-1. Overview of handling of spent powder resin at O1 and O2. (A-5, pict. 5).

The storage tanks T48 and T48 are irregularly filled with spent powder resin (sulfonate resin in cation part) and inert material from the CCU at O1 and O2 (*Figure A3-1*). In addition, powder resin and inert material from the system 342 (waste water treatment) are also added. Sometimes, fresh powder resin is added to the tanks T48 and T49 to get more capacity for cleaning of floor drainage which also is treated directly in those tanks<sup>54</sup>.

### A3.2. Part 1

#### A3.2.1. Input Data and Calculations

The first sampling campaign was performed between 2008-09-08 and 2009-03-12. The sampling record was delivered as Excel-books, *Attachment file O12-1. Table A3-1* shows an example of such a record. The operational time for each CCU filter is obtained from the table. Accordingly, it is possible to calculate the thermal energy produced during the operational time for each filter. It has been assumed that C-14 distributes uniformly between the filters. The production of C-14 is calculated from the thermal energy produced (*Attachment File O12-2, Sheet O1energy* and *O2Energy*) and the production rate obtained from the FSAR. The calculated amount is divided by 4 for O1 and by 6 for O2, to account for the number of filters

 $<sup>^{54}</sup>$  K-E Ingemansson, OKG: Pers. Comm. Feb. 2012: In 2011 the following volumes of waste water was received in T48 and T49: floor drain 11638 m<sup>3</sup> in 420 batches. Conductivity: Average 115, max 540, min 1.2  $\mu$ S/cm. Water from the decontamination facility was received in 49 batches amounting to totally 600 m<sup>3</sup>. Conductivity: Average 304, max 980, min 19  $\mu$ S/cm.



in the CCU. The timelines for the samples analyzed in Part 1 are shown in *Figure A3-2*. The calculations are found in *Attachment file O12-2*.

The amount of C-14 accumulated is calculated according to the Batch Model (BM) from the concentration in the samples, Bq/kg dw, multiplied with the total contents in the tank, (kg dw). *Figure A3-3* and *Table A3-2* show the concentrations of C-14 in the resin samples. Each concentration value is the average of three individual samples of a batch. If a value is less than half the detection limit value, it is replaced by a quarter of the detection limit value (see *Appendix 10*). In most cases, this leads to an overestimation in low concentration samples. Usually, it also leads to an overestimation of the organic fraction in low concentrations. However, in the total organic activity from several tank fillings, the contribution from these low activity samples is negligible. The analysis results are found in Sheet *OKG 332*.



Figure A3-2. Timeline for the tank fillings analyzed in Part 1. The lines are separated in the vertical direction for clarity. Batch T48-2 was not sampled and analyzed. (O12-2, D Timeline).

The standard deviation is the maximum value of scatter between the three samples analyzed in each batch and the compounded uncertainty in counting the samples. The scatter is usually larger than the uncertainty in activity counting. Despite large relative standard deviations in the separate samples, the variation in the sum of activity is just 2.5 %, since the batches with the highest activities have lower standard deviations. In the calculations, a 10 % uncertainty for the resin mass has been used in addition to the uncertainties in the concentrations.

From these numbers, the accumulation expressed as a percentage of the production and as  $Bq/MWh_{th}$  is calculated. *Table A3-6* shows an example of such a calculation. *Table A3-3* shows a summary for all the samples from O1+O2 analyzed for Part 1. In the calculations, a 10 % uncertainty for the resin mass has been used in addition to the uncertainties in the concentrations.





Figure A3-3. Concentration of C-14 in CCU resin samples from O1 and O2, Part 1. (O12-2, D conc).

				C-14			Tot.	Inorg.	Org.	_
Sam-			C-14 tot,	inorg,	C-14 org,	Org-	Standard	Standard	Standard	Inorg
ple	Start	End	Bq/kg dw	Bq/kg dw	Bq/kg dw	fraction	dev.	dev.	dev.	/Org*
T48-1	2008-09-08	2008-11-12	9.39E+04	9.06E+04	3.15E+03	3%	7%	5%	7%	0/2
T48-3	2008-11-28	2008-12-08	2.69E+03	1.68E+03	1.15E+03	43%	57%	62%	57%	2/3
T48-4	2008-12-12	2009-02-06	1.90E+04	1.79E+04	1.04E+03	5%	9%	6%	9%	0/2
T48-5	2009-04-08	2009-04-16	2.34E+05	2.23E+05	1.03E+04	4%	1%	2%	1%	0/0
T48-6	2009-04-20	2009-06-02	9.79E+03	4.26E+03	5.54E+03	57%	30%	31%	30%	0/0
T48-7	2009-06-05	2009-06-22	7.23E+05	7.12E+05	1.13E+04	2%	6%	6%	6%	0/0
T49-1	2008-10-20	2009-01-17	2.95E+05	2.91E+05	3.71E+03	1%	4%	4%	4%	0/0
T49-2	2009-01-27	2009-02-11	2.39E+03	1.15E+03	1.24E+03	52%	80%	102%	80%	1/3
T49-3	2009-02-13	2009-03-12	3.97E+04	3.87E+04	9.74E+02	2%	4%	4%	4%	0/3
Mean	2008-09-08	2009-06-22	1.58E+05	1.53E+05	4.27E+03	2.7%	142%	144%	89%	
* Numb	per of subsam	ples with (0,	25*detectio	on limit) inst	tead of dete	cted value	es			

Table A3-2. Summary of concentrations in samples of CCU resins from O1 and O2, Part 1. (O12-2, sheet Summary conc).

From *Table A3-3* it can be seen that the accumulation of C-14 ranges between 0.01 % and 0.92 % of the calculated production. The mean value is 0.30 %, including a Body-Feed Factor BFF=1.06. The calculation of the BFF is described under Discussion Part 1 below.

The large variation between the batches is probably due to differences in the air bubbling of the tank contents. Three batches (tanks) account for some 85 % of the total activity; see also *Figure A3-4*.



Avfallet O1/O2 - Provtagning C14 i jonbytarmassor (332)												Sofie Englund (OKG): Varje prov som skickats oss motsvarar ett batchprov.									
<u>Ifylles vi</u>	d prov	vtagn	ing	DG	)														Dá tanke rundpun	n blivit full kõrs luftomb 1pning i ca 15 min vareft	lanmdning + ter provet tas
Provdat	um: 2	009-0	6-23																ut. Detta	ska då motsvara hela ta	inkens
Prov ID:	T48:7	7																	Efter pro	vtagning töms tanken m	iereller
Från tan	k 342	T48									In	kopp	lingsir	ntervall	•				mindre.		
Värden f	ör (Te	emp)	och	(MV	Vth) a	ange	es s	om e	tt snit	t sett d	överı	respe	ktive 3	332 jon	bytares in	kopplig	stid "D	ygn".			
Datum	332 O1 Filter	332 O2 Filter	Gån Tei Te	332 C J1-J4 gtid/M mp/M rm.e1	)1 4 Medel edel ffekt	Går Te T€	332 C C1-C ngtid/I mp/M erm.et	02 6 Medel edel ifekt	Mängd jb- massa 332	Mängd inert- massa 332	342 Filter	Mängd jb- massa 342	Mängd inert massa 342	Extra dosering av Jb- massa tilll T48	Typ av massor MB-400 Finex 875PM UP2 W	Summa 332 massori 342 T48	Summa 342 massori 342 T48	Summa 332+342 massori 342T48	SIGN Noteringar		
ÅÅMMDD	J1-J4	C1-C6	Dygn	°c	MWth	Dygn	°c	MWth	Kg	Kg	(F1-F3, F6-F7)	Kg	Kg	Kg		Kg	Kg	Kg			
2009-06-05														50	Finex	0	50	50		Ny massa direkt till T48	
2009-06-08											F7	10	2.5		Finex+UP2	0	12.5	12.5			
2009-06-08											F1	10	2.5		Finex+UP2	0	12.5	12.5			
2009-06-11											F7	10	2.5		Finex+UP2	0	12.5	12.5			
2009-06-12											F7	10	2.5		Finex+UP2	0	12.5	12.5			
2009-06-14		C1				74			60	29					Finex+UP2	89	0	89			
2009-06-16	J3		47						70						MB-400	70	0	70			
2009-06-16		C2				64			60	25					Finex+UP2	85	0	85			
2009-06-18		C3				57			60	23					Finex+UP2	83	0	83			
2009-06-18														50	Finex	0	50	50		Ny massa direkt till T48	
2009-06-20											F7	10	2.5		Finex+UP2	0	12.5	12.5			
2009-06-20														20	Finex	0	20	20		Ny massa direkt till T48	
2009-06-22		C5				68			60	27					Finex+UP2	87	0	87			
															Summa	414	182.5	596.5			
	Provuttage 000623																				
	Brour	önad.	120 -				_														
	Provi	nanga:	130 g				_														
Fördeln	ing (33	2/342):	69%				_														
Provtag	n.inforn	nation:	ex til	lväga	agångs	ätt (lu	iftom	blandr	ning före	∋?)											
	Övrigt: 2 I dunk med ca 1/20 JB						-														

Table A3-1. Example of record-keeping for CCU powder resins. In this sheet inert masses for O2 have been included.O12-1, sheet T48-7 090623).

Resin u	ncertainty					10%												
			01	02									Fract.					
			Effective	Effective	Calc.	Accum.	Accum.	Accum.	Org.	Tot.	Inorg.	Org.	of pro-					
Sam-			energy,	energy,	Produc-	C-14 tot,	C-14	C-14 org,	Frac-	std.	std.	std.	duc-	Total	Inorg.	Org.		Inorg
ple	Start	End	MWhth	MWhth	tion, Bq	Bq	inorg, Bq	Bq	tion	dev.	dev.	dev.	tion	Bq/MWhth	Bq/MWhth	Bq/MWhth	BFF	/Org*
T48-1	2008-09-08	2008-11-12	1.76E+06	1.73E+06	7.59E+10	1.07E+08	1.04E+08	3.59E+06	3.4%	12%	11%	12%	0.13%	3.08E+01	2.98E+01	1.03E+00	1.07	0/2
T48-3	2008-11-28	2008-12-08	0.00E+00	4.24E+05	8.48E+09	7.90E+05	4.54E+05	3.36E+05	43%	58%	63%	58%	0.01%	1.86E+00	1.07E+00	7.93E-01	1.04	2/3
T48-4	2008-12-12	2009-02-06	9.73E+05	1.22E+06	4.73E+10	1.64E+07	1.55E+07	8.96E+05	5.5%	13%	12%	13%	0.03%	7.48E+00	7.07E+00	4.09E-01	1.06	0/2
T48-5	2009-04-08	2009-04-16	1.50E+06	1.77E+06	7.09E+10	2.83E+08	2.70E+08	1.25E+07	4.4%	10%	10%	10%	0.37%	8.64E+01	8.26E+01	3.82E+00	1.07	0/0
T48-6	2009-04-20	2009-06-02	1.90E+06	1.63E+06	7.74E+10	1.17E+07	5.10E+06	6.64E+06	57%	32%	32%	32%	0.01%	3.33E+00	1.44E+00	1.88E+00	1.07	0/0
T48-7	2009-06-05	2009-06-22	3.87E+05	1.88E+06	4.67E+10	4.70E+08	4.63E+08	7.34E+06	1.6%	12%	12%	12%	0.92%	2.07E+02	2.04E+02	3.24E+00	1.09	0/0
T49-1	2008-10-20	2009-01-17	8.15E+05	1.07E+06	4.06E+10	3.03E+08	2.99E+08	3.82E+06	1.3%	11%	11%	11%	0.72%	1.61E+02	1.59E+02	2.03E+00	1.04	0/0
T49-2	2009-01-27	2009-02-11	0.00E+00	8.31E+05	1.66E+10	5.39E+05	2.59E+05	2.79E+05	52%	81%	102%	81%	0.00%	6.48E-01	3.12E-01	3.36E-01	1.10	1/3
T49-3	2009-02-13	2009-03-12	0.00E+00	8.55E+05	1.71E+10	2.77E+07	2.70E+07	6.79E+05	2.5%	11%	11%	11%	0.16%	3.24E+01	3.16E+01	7.95E-01	1.03	0/3
Sum	2008-09-08	2009-06-22	7.34E+06	1.14E+07	4.01E+11	1.22E+09	1.18E+09	3.61E+07	3.0%	5.8%	5.9%	7.5%	0.30%	6.51E+01	6.32E+01	1.93E+00	1.06	
Equiva	lent Full Po	wer Hours	5338	6333														
Equiva	lent Full Po	wer Years	0.61	0.72														
* Numb	er of subsam	ples with (0,	25*detectio	n limit) inst	tead of det	ected value	S											

Table A3-3. Summary of calculated accumulation of C-14 in CCU resin from O1 and O2, Part 1. The tank fillings with organic values based on ¼ of the detection limit are all below the average organic accumulation. (O12-2, sheet Summary act).



Figure A3-4. Activity contents in the tanks sampled for O1 and O2, Part 1. (O12-2, sheet D Act).

The organic fraction also varies strongly between the batches; usually samples with detection limit values have the highest organic fraction. However, Sample T48-5 has the highest organic activity, but the organic fraction is only 4.4 %. There is no obvious operational occurrence explaining this extreme value. The samples with low concentrations may have high organic fractions, but the total activity and the organic activity are low. The average organic fraction of 3.0 % is in the higher range obtained for BWR plants in this study; however *Ref. 4* reports some values in the range 15-30 % for BWR resins.



#### A3.2.2. Discussion Part 1

Oxide		Fe3O4	Fe2O3	Mean
Fe	atomic mass	56	56	
0	atomic mass	16	16	
Fe oxide	mole mass	232	160	
Fe oxide/F	e	1.381	1.429	
lonic fracti	on	0.1	0.1	
(Fe+Fe ox	ide)/Fe on resin	1.34	1.39	1.36
Concentra	tion Fe in water, ppb		01	10
Concentra	tion Fe in water, ppb		02	10

Table A3-4. Estimation of correlation factor to calculate Body-Feed Factor (BFF)for O1 and O2. (O12-2, sheet BodyfeedO12).

The Body-Feed Factor (BFF) accounts for the uptake of corrosion products on the CCU resin since the tank content has been reported only as resin and inert material added. In the calculation sheets (example in *Table A3-6*) the water mass passing the filter has been calculated from the operational time of each filter, assuming that the feed water flow rate has been equally distributed between the filters.

The calculations have been performed with the assumptions of the chemical composition shown in *Table A3-4*; the BFF has been calculated for each tank filling. The values are displayed in Table *A3-3*. The concentration of corrosion products has been assumed to be 10 ppb Fe, with 10 % in ionic form. The rest is assumed to be equally distributed between  $Fe_3O_4$  and  $Fe_2O_3$ . This yields BFF values in the range 1.02-1.07 for the samples. The calculation of the correlation factor is shown in *Table A3-6*.

O1+O2, Part 1	This study
	Mean
	conc.
	T48+T49
Equivalent Full Power Hours (EFPH), O1	5338
Equivalent Full Power Hours (EFPH), O2	6333
Equivalent Full Power Years (EFPY), O1	0.61
Equivalent Full Power Years (EFPY), O2	0.72
Body-Feed Factor, Average (BFF)	1.06
Dry Solids Correction Factor in Sample	
Organic fraction	3.0%
Total accumulation in CCU	0.30%
Organic accumulation in CCU	0.0090%
	_
Total Bq/MWhth	6.51E+01
Inorganic Bq/MWhth	6.32E+01
Organic Bq/MWhth	1.93E+00
Total Std. Dev.	5.8%
Inorganic Std. Dev.	5.9%
Organic Std. Dev.	7.5%

Table A3-5. Summary of accumulation of C-14 in CCU resins for O1 and O2, Part 1.(O12-2, sheet Summary).

*Table A3-5* summarizes the resulting accumulation, expressed as percent of production and Bq/MWh<sub>th</sub>, respectively. The table also contains values for the energy production, expressed as Equivalent Full Power Hours (EFPH) and Equivalent Full Power Yours (EFPY), to give an idea of the operation time for the units.



														Month end date	Month	O1, GWhth/month	O2, GWhth/month	days/ month		
Avfalle	et O1	/02	- P	ro۱	∕tag	nin	g (	C14 i j	onbytar	rmass	sor (3:	32)		2009-02-28	februari	388	1207	28		
Ifylles vi	d pro	vtagn	ing (	(DG	i)						<b>`</b>			2009-03-31	mars	865	1328	31		
Provdat	um: 2	009-0	6-23	;										2009-04-30	april	989	1292	30		
Prov ID:	T48:7	7												2009-05-31	mai	1022	1330	31		
Från tan	k 342	T48												2009-06-30	iuni	987	1284	30		
Värdon	är (T	(mn)	aah	/ 8.43	N+h)	anac		om ott	onitt oott i	äver reg	o no ktiv	o 222 io	nhuta	roc inkon	pliactid "F	)van"	1201	00		
varuen		inh) (	ocn	(1919	vinj	ange	:2 2	omett	sniit sett c	Jverie	speкuv	e ssz ju	пруга	ires inkop		Jygn .				
															filters	4	6			
Datum	332 O1 Filter	332 O2 Filter	Gån Tei Te	332 C J1-J Igtid/I mp/M erm.e	D1 4 Medel Iedel ffekt	332 C1- Gång ed Temp	: O2 -C6  tid/M lel 0/Med	Extra dosering av Jb- massa tilll T48	Typ av massor MB-400 Finex 875PM UP2 W	Summa 332 massor i 342 T48	Summa 342 massori 342 T48	Summa 332+342 massor i 342T48	SIGN	Startdag O1	Startdag O2	Energi-prod under period, O1 GWh	Energi-prod under period, O2 GWh	Energi, Summa O1+O2	O1 water mass, kg	O2 water mass, kg
ÅÅMMDD	J1-J4	C1-C6	Dygn	°c	MWth	Dygn	°C	Kg		Kg	Kg	Kg								
2009-06-05								50	Finex	0	50	50								
2009-06-08								00	Finex+UP2	0	12.5	12.5								
2009-06-08									Finex+LIP2	0	12.5	12.5								
2009-06-11									Finex+LIP2	0	12.5	12.5								
2009-06-12						1			Finex+UP2	0	12.5	12.5								
2009-06-14		C1				74			Finex+UP2	89	0	89			2009-04-01		3178			9.59E+08
2009-06-16	J3		47						MB-400	70	0	70		2009-04-30		1548			5.08E+08	
2009-06-16		C2				64			Finex+UP2	85	0	85			2009-04-13		2747			8.29E+08
2009-06-18		C3				57			Finex+UP2	83	0	83			2009-04-22		2445			7.39E+08
2009-06-18								50	Finex	0	50	50								
2009-06-20									Finex+UP2	0	12.5	12.5								
2009-06-20								20	Finex	0	20	20								
2009-06-22		C5				68			Finex+UP2	87	0	87			2009-04-15		2918			8.81E+08
									Summa	414	182.5	596.5			GWhth/unit	1548.4	11287.6	12836	5.08E+08	3.41E+09
			_						Analysis results					Calc. Production	C-14, Bq	3.65E+10	2.26E+11			
		Provuttag	090623	3					Accumulation, E	Bq		4.31E+08		Calc. Production	C-14, Bq/filter	9.12E+09	3.76E+10	4.67E+10		
									Fraction accumu	ulated		9.22E-03								
	F	rovmäng	130 g					1 Calve et	C-14, Bq/MWhth	<b>1</b>		3.36E+01			GWhth/filter	387	1881	2268.4	<u> </u>	
	Förda	Ining (22)	0.604					Linked	O1: Fe conc in c	condensate,	ppb	10							<u> </u>	
	Fuide	ming (332	0.094					LINKED	Corrosion produ	onuensale, cts ka	hhn	53								
	Provta	agn.inform	tt (lufto	mblar	ndning fö	re?)			BFF	sto, itg		1.09					<u> </u>			

Table A3-6. Example of complete calculation for one sample/tank contents. In this sheet inert masses for O2 have been included, but are hidden.O12-2, sheet T48-7 090623).



## A3.3. Part 2

### A3.2.1. Input Data and Calculations

A second sampling campaign was performed between 2010-09-14 and 2011-05-13. The sampling records are found in *Attachment file O12-3*. All calculations are included in an excel-book, *Attachment File O12-4* to this report. Reference to the various sheets will be made at tables, figures etc. The tables and figures for Part 2 are numbered from 21 and upwards.



Figure A3-21. Timeline for the tank fillings analyzed in Part 2. The lines are separated in the vertical direction for clarity. (O12-4, sheet D Timeline).



Figure A3-22. Concentration of C-14 in CCU resin samples from O1 and O2, Part 2. (O12-4, sheet D conc).



Sampling and analysis were performed in the same way as described in Part 1 above. The timeline for the tank fillings is shown in *Figure A3-21*. The C-14 concentrations in the resin samples are shown in *Figure A3-22* and *Table A3-21*. In this sampling campaign, there were fewer samples where detection limit values were used in the calculations.

Sample	Start	End	C-14 tot, Bg/kg dw	C-14 inorg, Bg/kg dw	C-14 org, Bg/kg dw	Org- fract.	Tot. Std. dev.	Inorg. Std. dev.	Org. Std. dev.	Inorg /Org*	Note
T49 101018	2010-09-14	2010-10-18	3.12E+04	2.12E+04	1.00E+04	32%	6%	6%	12%	0/0	
T48 101108	2010-10-08	2010-11-06	1.92E+05	1.87E+05	5.07E+03	2.6%	3%	3%	27%	0/0	
T49 101117	2010-10-20	2010-11-15	9.02E+04	8.30E+04	7.17E+03	8.0%	3%	2%	15%	0/0	
T48 101208	2010-11-09	2010-12-03	5.68E+03	4.68E+03	1.05E+03	18%	26%	25%	103%	0/3	
T49 101229	2010-11-24	2010-12-27	5.40E+04	4.86E+04	5.45E+03	10%	3%	4%	30%	0/0	
T49 110128	2011-01-03	2011-01-21	2.35E+05	2.33E+05	2.22E+03	0.9%	5%	5%	47%	0/0	
T48 110207	2010-12-14	2011-02-07	9.67E+04	8.89E+04	7.81E+03	8.1%	12%	12%	14%	0/0	
T48 110214	2011-02-14	2011-02-14	6.61E+05	6.49E+05	1.21E+04	1.8%	4%	5%	26%	0/0	
T48 110221	2011-02-18	2011-02-18	1.49E+06	1.45E+06	3.69E+04	2.5%	10%	10%	12%	0/0	
T48 110224	2011-02-24	2011-02-24	2.80E+06	2.72E+06	8.78E+04	3.1%	1%	1%	34%	0/0	**
T49 110316	2011-02-03	2011-03-14	1.83E+04	1.28E+04	5.54E+03	30%	10%	13%	27%	0/0	
T49 110321	2011-03-21	2011-03-21	3.02E+05	2.99E+05	2.83E+03	0.9%	8%	8%	45%	0/0	
T49 110412	2011-04-01	2011-04-12	8.05E+05	7.88E+05	1.72E+04	2.1%	3%	3%	8%	0/0	
T48 110516	2011-05-11	2011-05-13	8.11E+04	7.48E+04	6.26E+03	7.7%	3%	2%	17%	0/0	
Mean	2010-09-14	2011-05-13	4.90E+05	4.75E+05	1.48E+04	3.0%	154%	155%	149%	0/3	
* Number of s	subsamples	with (0,25*de	etection lim	it) instead o	of detected	values					
** Short air l	bubbling										

Table A3-21. Summary of concentrations in samples of CCU resins from O1 and O2 Part 2.(O12-4, sheet Summary conc).

The calculation of the activity contents was performed in the same way as described for Part 1 above. However, the record keeping of masses did not include the inert material added as body-feed. To compensate for this, the amount of body-feed material was calculated from the injection rate and the operating time and included in the Body-Feed Factor (BFF). In the calculations, a 10 % uncertainty for the resin mass has been used in addition to the uncertainties in the concentrations. The results are found in *Table A3-22* and *Figure A3-23*.

Resin unce	eratinty					10%												
			01	02			Accum.						Fract.					
			Effective	Effective	Calc.	Accum.	C-14	Accum.	Org.	Tot.	Inorg.	Org.	of pro-					
			energy,	energy,	Produc-	C-14 tot,	inorg,	C-14 org,	frac-	std.	std.	std.	duc-	Total	Inorg.	Org.		Inorg/
Sample	Start	End	MWhth	MWhth	tion, Bq	Bq	Bq	Bq	tion	dev.	dev.	dev.	tion	Bq/MWhth	Bq/MWhth	Bq/MWhth	BFF	Org*
T49 101018	3 2010-09-14	2010-10-18	4.67E+05	3.31E+05	1.76E+10	2.85E+07	1.94E+07	9.17E+06	32%	11%	12%	15%	0.16%	3.58E+01	2.43E+01	1.15E+01	1.06	0/0
T48 101108	3 2010-10-08	2010-11-06	0.00E+00	6.74E+05	1.35E+10	3.45E+07	3.36E+07	9.13E+05	3%	10%	10%	29%	0.26%	5.12E+01	4.99E+01	1.35E+00	1.06	0/0
T49 101117	2010-10-20	2010-11-15	2.45E+05	5.46E+05	1.67E+10	9.79E+07	9.01E+07	7.79E+06	8%	10%	10%	18%	0.59%	1.24E+02	1.14E+02	9.84E+00	1.07	0/0
T48 101208	3 2010-11-09	2010-12-03	0.00E+00	0.00E+00	0.00E+00	5.08E+06	4.14E+06	9.38E+05	18%	28%	27%	103%					1.00	0/3
T49 101229	2010-11-24	2010-12-27	0.00E+00	1.50E+05	2.99E+09	2.54E+07	2.29E+07	2.57E+06	10%	11%	11%	31%	0.85%	1.70E+02	1.53E+02	1.72E+01	1.03	0/0
T49 110128	3 2011-01-03	2011-01-21	0.00E+00	1.96E+06	3.93E+10	1.66E+08	1.64E+08	1.57E+06	1%	11%	11%	48%	0.42%	8.42E+01	8.34E+01	7.98E-01	1.32	0/0
T48 110207	2010-12-14	2011-02-07	6.21E+05	0.00E+00	1.46E+10	8.72E+07	8.02E+07	7.04E+06	8%	16%	16%	17%	0.60%	1.40E+02	1.29E+02	1.13E+01	1.01	0/0
T48 110214	2011-02-14	2011-02-14	0.00E+00	8.50E+05	1.70E+10	1.21E+08	1.19E+08	2.20E+06	2%	11%	11%	28%	0.71%	1.42E+02	1.39E+02	2.59E+00	1.52	0/0
T48 110221	2011-02-18	2011-02-18	9.35E+05	0.00E+00	2.20E+10	1.30E+08	1.27E+08	3.23E+06	2%	14%	14%	16%	0.59%	1.39E+02	1.36E+02	3.46E+00	1.25	0/0
T48 110224	2011-02-24	2011-02-24	9.76E+05	0.00E+00	2.30E+10	2.48E+08	2.40E+08	7.75E+06	3%	10%	10%	36%	1.08%	2.54E+02	2.46E+02	7.95E+00	1.26	0/0
T49 110316	6 2011-02-03	2011-03-14	0.00E+00	0.00E+00	0.00E+00	1.14E+07	7.98E+06	3.46E+06	30%	14%	16%	28%					1.00	0/0
T49 110321	2011-03-21	2011-03-21	0.00E+00	2.27E+05	4.54E+09	2.32E+07	2.30E+07	2.18E+05	1%	13%	13%	46%	0.51%	1.02E+02	1.01E+02	9.59E-01	1.28	0/0
T49 110412	2 2011-04-01	2011-04-12	0.00E+00	1.87E+06	3.74E+10	3.17E+08	3.11E+08	6.77E+06	2%	10%	10%	13%	0.85%	1.70E+02	1.66E+02	3.62E+00	1.55	0/0
T48 110516	2011-05-11	2011-05-13	5.39E+04	0.00E+00	1.27E+09	2.20E+07	2.03E+07	1.70E+06	8%	10%	10%	20%	1.73%	4.08E+02	3.76E+02	3.15E+01	1.00	0/0
Sum	2010-09-14	2011-05-13	3.30E+06	6.61E+06	2.10E+11	1.32E+09	1.26E+09	5.53E+07	4.2%	4.1%	4.1%	8%	0.63%	1.33E+02	1.27E+02	5.58E+00	1.17	0/3

Table A3-22. Summary of calculated accumulation of C-14 in CCU resin from O1 and O2, Part 2. The tank fillings with organic values based on ¼ of the detection limit are all below the average organic accumulation. (O12-4, sheet Summary act).

The tank filling with the highest activity concentration and activity contents, T48 110224, had been air bubbled only for a short time, compared to the standard procedure, prescribing 30 minutes<sup>55</sup>. It accounts for some 20 % of the activity in part 2.



Figure A3-23. Activity contents in the tanks sampled in Part 2. (O12-4, sheet D Act).

The results for Part 1 and Part 2 are summarized in *Table A3-23*. Part 2 has a higher accumulation and higher organic fraction than Part 1. There is no obvious explanation for this.

Sample	Start	End	O1 Effective energy, MWhth	O2 Effective energy, MWhth	Calc. Produc- tion, Bq	Accum. C-14 tot, Bq	Accum. C-14 org, Bq	Accum. C-14 org, Bq	Org- frac- tion	Tot. std. dev.	Inorg. std. dev.	Org. std. dev.	Fract. of pro- duc- tion	Total Bq/MWhth	Inorg. Bq/MWhth	Org. Bq/MWhth	BFF
Part 1	2008-09-08	2009-06-22	7.34E+06	1.14E+07	4.01E+11	1.22E+09	1.18E+09	3.61E+07	3.0%	2.5%	2.6%	17%	0.30%	6.51E+01	6.32E+01	1.93E+00	1.06
Part 2	2010-09-14	2011-05-13	3.30E+06	6.61E+06	2.10E+11	1.32E+09	1.26E+09	5.53E+07	4.2%	4.1%	4.1%	7.6%	0.63%	1.33E+02	1.27E+02	5.58E+00	1.17
Sum			1.06E+07	1.80E+07	6.11E+11	2.54E+09	2.45E+09	9.14E+07	3.6%	2.4%	2.5%	8.2%	0.42%	8.86E+01	8.54E+01	3.19E+00	1.12
Equivale	nt Full Pow	er Hours	7736	10005													
Equivale	nt Full Pow	er Years	0.88	1.14													

Table A3-23. Summary of accumulation of C-14 in CCU resins for O1 and O2: Part 1 and Part 2. (O12-4, sheet Summary P1+P2).

#### A3.2.2. Discussion Part 1 and Part 2

Part 2 shows a higher accumulation of total C-14 and organic C-14, compared with Part 1. The total energy exposure, expressed as EFPY (Equivalent Full Power Years), corresponds to 0.88 for O1 and 1.14 for O2.

<sup>&</sup>lt;sup>55</sup> Pers. communication. Christina Lillfors-Pintér and K-E Ingemansson OKG, 2012-01-02.





01+02	This study	This study	This study
	Part 1	Part 2	Part 1+2
Equivalent Full Power Hours (EFPH), O1	5338	2399	7736
Equivalent Full Power Hours (EFPH), O2	6333	3673	10005
Equivalent Full Power Years (EFPY), O1	0.61	0.27	0.88
Equivalent Full Power Years (EFPY), O2	0.72	0.42	1.14
Equivalent Full Power Years (EFPY), O1 + O2	1.33	0.69	2.03
Body-Feed Factor, Average (BFF)	1.06	1.17	1.12
Dry Solids Correction Factor in Sample			
Organic fraction	3.0%	4.2%	3.6%
Total accumulation in CCU	0.30%	0.63%	0.42%
Organic accumulation in CCU	0.0090%	0.026%	0.015%
Total Bq/MWhth	6.51E+01	1.33E+02	8.86E+01
Inorganic Bq/MWhth	6.32E+01	1.27E+02	8.54E+01
Organic Bq/MWhth	1.93E+00	5.58E+00	3.19E+00
Total Std. Dev.	2.5%	4.1%	2.4%
Inorganic Std. Dev.	2.6%	4.1%	2.5%
Organic Std. Dev.	17%	7.6%	8.2%

Table A3-24. Summary of accumulation of C-14 in CCU resins for O1 and O2, Part 1 and Part 2. (O12-4, sheet Summary 1+2).

### A3.4. Part 3

#### A3.4.1. Input Data and Calculations

The third sampling campaign was performed between 2011-07-10 and 2012-12-10. The sampling record was delivered as Excel-books in the same way as for Part 1 and 2. *Table A3-1* shows an example of such a record. All calculations are included in an excel-book, *Attachment File O12-5*. The tables and figures for Part 3 are numbered from 31 and upwards.

The operational time for each CCU filter is obtained from the table. Accordingly, it is possible to calculate the thermal energy produced during the operational time for each filter. It has been assumed that C-14 distributes uniformly between the filters. The production of C-14 is calculated from the thermal energy produced (*O12-5, sheets Energy month* and *Energy day*<sup>56</sup>) and the production rate obtained from the FSAR. The calculated amount is divided by 4 for O1 and by 6 for O2, to account for the number of filters in the CCU).

Sampling, analysis and calculations were performed in the same way as for Part 1 and 2 above. The timelines for the samples analyzed in Part 3 are shown in *Figure A3-31*.

<sup>&</sup>lt;sup>56</sup> To simplify data extraction, the monthly energy production was evenly distributed over each month.







Figure A3-31. Timeline for the tank fillings analyzed in Part 3. The lines are separated in the vertical direction for clarity. (O12-5, sheet D Timeline).



Figure A3-32. Concentration of C-14 in CCU resin samples from O1 and O2, Part 3. (O12-5, sheet D conc).

The concentrations are presented in *Figure A3-32* and *Table A3-31*. In this campaign only 10 subsamples have used the detection limit value (divided by 4).

				C-14 tot,	C-14 inorg,	C-14 org,	Org-	Tot. Std.	Inorg.	Org. Std.	Inorg/
Sample	Sample	Start	End	Bq/kg dw	Bq/kg dw	Bq/kg dw	fract.	dev.	Std. dev.	dev.	Org*
012-24	T49 110810	2011-07-11	2011-08-10	3.72E+05	3.58E+05	1.36E+04	3.7%	1.3%	1.5%	7.1%	0/0
012-25	T49 110906	2011-08-22	2011-09-06	4.34E+04	3.65E+04	6.97E+03	16%	7.9%	8.8%	35%	0/0
012-26	T48 110911	2011-08-30	2011-09-11	2.51E+05	2.36E+05	1.46E+04	5.8%	4.2%	3.4%	18%	0/0
012-27	T49 111007	2011-09-11	2011-10-07	2.23E+05	2.17E+05	5.83E+03	2.5%	4.2%	2.7%	72%	0/0
012-29	T48 111021	2011-09-22	2011-10-21	9.36E+04	8.46E+04	8.96E+03	10%	2.4%	2.2%	10%	0/0
012-28	T49 111108	2011-10-08	2011-11-08	1.94E+05	1.93E+05	7.63E+02	0.39%	3.7%	3.7%	116%	0/2
012-30	T48 111128	2011-10-27	2011-11-28	8.20E+03	7.49E+03	7.18E+02	8.9%	14%	13%	115%	0/3
012-31	T49 111229	2011-11-14	2011-12-29	1.60E+05	1.58E+05	2.05E+03	1.3%	3.1%	4.2%	84%	0/0
012-32	T49 120113	2012-01-13	2012-01-13	7.43E+05	7.32E+05	1.09E+04	1.5%	1.2%	1.6%	33%	0/0
012-33	T49 120120	2012-01-20	2012-01-20	6.61E+05	6.54E+05	7.75E+03	1.2%	1.4%	1.5%	11%	0/0
012-34	T49 120127	2012-01-26	2012-01-27	4.89E+04	4.72E+04	1.67E+03	3.3%	6.5%	4.3%	77%	0/2
012-35	T48 120203	2011-12-30	2012-02-03	8.15E+04	7.87E+04	2.82E+03	3.4%	4.6%	2.9%	60%	0/0
012-25	T49 120215	2012-02-06	2012-02-15	4.34E+04	3.65E+04	6.97E+03	16%	7.9%	8.8%	35%	0/0
012-36	T48 120319	2012-02-16	2012-03-19	7.51E+04	7.22E+04	2.87E+03	3.7%	19%	19%	75%	0/0
012-37	T49 120420	2012-03-21	2012-04-20	2.23E+05	2.12E+05	1.04E+04	4.7%	47%	47%	50%	0/0
012-38	T48 120506	2012-04-15	2012-05-06	4.23E+05	4.12E+05	1.03E+04	2.4%	1.5%	3.1%	65%	0/0
012-39	T49 120516	2012-05-04	2012-05-16	1.03E+05	9.02E+04	1.30E+04	13%	47%	48%	55%	0/0
012-40	T48120607	2012-05-18	2012-06-07	1.71E+03	8.31E+02	8.83E+02	52%	73%	103%	104%	0/2
012-41	T49 120627	2012-06-08	2012-06-27	1.71E+04	1.18E+04	5.30E+03	27%	40%	32%	58%	0/0
012-25	T49 120808	2012-07-29	2012-08-08	4.34E+04	3.65E+04	6.97E+03	16%	7.9%	8.8%	35%	0/0
012-42	T48 120829	2012-08-10	2012-08-29	1.05E+05	1.04E+05	8.77E+02	0.84%	4.5%	4.5%	105%	0/1
012-43	T49 120912	2012-08-30	2012-09-12	1.71E+05	1.66E+05	5.23E+03	3.1%	7.0%	7.8%	48%	0/0
012-44	T48 120927	2012-09-12	2012-09-27	1.05E+05	9.88E+04	6.72E+03	6.6%	6.3%	11%	83%	0/0
012-45	T49 121010	2012-09-27	2012-10-10	5.96E+04	5.14E+04	8.27E+03	14%	5.1%	13%	85%	0/0
012-46	T49 121210	2012-10-29	2012-12-10	2.19E+05	2.05E+05	1.37E+04	6.2%	3.5%	6.9%	92%	0/0
	Mean			1.79E+05	1.72E+05	6.72E+03	3.8%	105%	108%	65%	0/10

Table A3-31. Summary of concentrations in samples of CCU resins from O1 and O2 Part 3.(O12-5, sheet Conc).

The calculated activity contents are shown in *Figure A3-33* and *Table A3-32*. In the calculations, a 10 % uncertainty for the resin mass has been used in addition to the uncertainties in the concentrations.

#### A3.4.2. Discussion Part 3

Almost all CCU resins originate from unit O2, since O1 was shut down most of the time. Some of the tanks did not contain any CCU resin at all. In *Table A3-32* these tanks are recognized by having blank entries for accumulation rate (Bq/MWh<sub>th</sub>).

#### A3.4.3. Summary and Discussion Part 1, Part 2 and Part 3

*Table A3-33* and *Figure A3-34* show a summary of the accumulation of C-14 for the three parts analyzed. The standard deviations reported include the standard deviation in the concentrations and a 10 % standard deviation in the resin masses.

Summing all activity accumulated during the three campaigns yields values for the accumulation with reasonably low standard deviations, even if there is almost a factor of two between the highest and lowest values in the respective campaigns. The energy production amounts to 1.19 EFPY for O1 and 2.13 EFPY for O2.





Figure A3-33. Activity contents in the tanks sampled in Part 3. (O12-5, sheet D Acc).

Resin uncerte	ainty					10%													
Reall uncerte			O1 Effective energy,	O2 Effective energy,	Calc. Produc-	Resin, kg	Accum. C-14 tot,	Accum. C-14	Accum. C-14 org,	Org. frac-	Tot. std.	Inorg. std.	Org. std.	Fract. of produc-	Total	Inorg.	Org. Bq/MWht		Inorg
Sample	Start	End	MWhth	MWhth	tion, Bq	dw	Bq	inorg, Bq	Bq	tion	dev.	dev.	dev.	tion	Bq/MWhth	Bq/MWhth	h	BFF	Org*
T49 110810	2011-07-11	2011-08-10	5.60E+05	4.48E+05	2.21E+10	5.85E+02	2.34E+08	2.25E+08	8.57E+06	3.7%	10%	10%	12%	1.1%	2.32E+02	2.24E+02	8.50E+00	1.08	0/0
T49 110906	2011-08-22	2011-09-06	0.00E+00	0.00E+00	0.00E+00	4.85E+02	2.11E+07	1.77E+07	3.38E+06	16%	13%	13%	36%					1.00	0/0
T48 110911	2011-08-30	2011-09-11	0.00E+00	2.64E+06	5.27E+10	4.95E+02	1.86E+08	1.75E+08	1.08E+07	5.8%	11%	11%	21%	0.35%	7.04E+01	6.62E+01	4.11E+00	1.50	0/0
T49 111007	2011-09-11	2011-10-07	7.07E+05	0.00E+00	1.67E+10	6.55E+02	1.49E+08	1.45E+08	3.89E+06	2.6%	11%	10%	73%	0.89%				1.02	0/0
T48 111021	2011-09-22	2011-10-21	9.81E+04	0.00E+00	2.31E+09	9.15E+02	8.58E+07	7.76E+07	8.21E+06	10%	10%	10%	14%	3.7%	8.74E+02	7.91E+02	8.37E+01	1.00	0/0
T49 111108	2011-10-08	2011-11-08	1.46E+06	0.00E+00	3.44E+10	8.60E+02	1.72E+08	1.72E+08	6.77E+05	0.39%	11%	11%	117%	0.50%	1.18E+02	1.18E+02	4.64E-01	1.03	0/2
T48 111128	2011-10-27	2011-11-28	4.81E+05	0.00E+00	1.13E+10	8.00E+02	6.64E+06	6.06E+06	5.82E+05	8.8%	17%	16%	115%	0.059%	1.38E+01	1.26E+01	1.21E+00	1.01	0/3
T49 111229	2011-11-14	2011-12-29	3.33E+05	1.71E+06	4.20E+10	1.11E+03	2.07E+08	2.05E+08	2.65E+06	1.3%	10%	11%	85%	0.49%	1.02E+02	1.00E+02	1.30E+00	1.17	0/0
T49 120113	2012-01-13	2012-01-13	0.00E+00	2.22E+05	4.43E+09	6.00E+01	5.59E+07	5.51E+07	8.17E+05	1.5%	10%	10%	35%	1.3%	2.52E+02	2.49E+02	3.69E+00	1.25	0/0
T49 120120	2012-01-20	2012-01-20	0.00E+00	3.58E+05	7.15E+09	6.00E+01	5.64E+07	5.57E+07	6.61E+05	1.2%	10%	10%	15%	0.79%	1.58E+02	1.56E+02	1.85E+00	1.42	0/0
T49 120127	2012-01-26	2012-01-27	0.00E+00	0.00E+00	0.00E+00	1.40E+02	7.10E+06	6.86E+06	2.43E+05	3.4%	12%	11%	78%					1.04	0/2
T48 120203	2011-12-30	2012-02-03	0.00E+00	1.20E+06	2.41E+10	7.65E+02	6.94E+07	6.70E+07	2.40E+06	3.5%	11%	10%	61%	0.29%	5.76E+01	5.56E+01	1.99E+00	1.11	0/0
T49 120215	2012-02-06	2012-02-15	0.00E+00	0.00E+00	0.00E+00	4.15E+02	1.80E+07	1.51E+07	2.89E+06	16%	13%	13%	36%					1.00	0/0
T48 120319	2012-02-16	2012-03-19	0.00E+00	1.20E+06	2.41E+10	8.00E+02	6.72E+07	6.47E+07	2.57E+06	3.8%	22%	21%	75%	0.28%	5.59E+01	5.37E+01	2.13E+00	1.12	0/0
T49 120420	2012-03-21	2012-04-20	0.00E+00	1.32E+06	2.64E+10	7.15E+02	1.80E+08	1.72E+08	8.42E+06	4.7%	48%	48%	51%	0.68%	1.37E+02	1.30E+02	6.39E+00	1.13	0/0
T48 120506	2012-04-15	2012-05-06	0.00E+00	0.00E+00	0.00E+00	5.70E+02	2.41E+08	2.35E+08	5.85E+06	2.4%	10%	10%	66%					1.00	0/0
T49 120516	2012-05-04	2012-05-16	0.00E+00	1.05E+06	2.09E+10	3.95E+02	4.84E+07	4.23E+07	6.10E+06	13%	48%	49%	56%	0.23%	4.63E+01	4.04E+01	5.83E+00	1.19	0/0
T48120607	2012-05-18	2012-06-07	0.00E+00	6.38E+05	1.28E+10	4.70E+02	8.82E+05	4.28E+05	4.55E+05	52%	74%	104%	105%	0.0069%	1.38E+00	6.70E-01	7.12E-01	1.10	0/2
T49 120627	2012-06-08	2012-06-27	0.00E+00	6.06E+05	1.21E+10	4.40E+02	8.33E+06	5.75E+06	2.58E+06	31%	41%	33%	59%	0.069%	1.38E+01	9.49E+00	4.26E+00	1.11	0/0
T49 120808	2012-07-29	2012-08-08	0.00E+00	0.00E+00	0.00E+00	3.00E+02	1.30E+07	1.09E+07	2.09E+06	16%	13%	13%	36%					1.00	0/0
T48 120829	2012-08-10	2012-08-29	0.00E+00	5.41E+05	1.08E+10	4.60E+02	5.61E+07	5.56E+07	4.68E+05	0.83%	11%	11%	106%	0.52%	1.04E+02	1.03E+02	8.66E-01	1.16	0/1
T49 120912	2012-08-30	2012-09-12	0.00E+00	4.17E+05	8.35E+09	3.25E+02	6.18E+07	5.99E+07	1.89E+06	3.1%	12%	13%	49%	0.74%	1.48E+02	1.44E+02	4.53E+00	1.11	0/0
T48 120927	2012-09-12	2012-09-27	0.00E+00	5.48E+05	1.10E+10	4.65E+02	5.23E+07	4.90E+07	3.33E+06	6.4%	12%	15%	83%	0.48%	9.55E+01	8.95E+01	6.08E+00	1.07	0/0
T49 121010	2012-09-27	2012-10-10	0.00E+00	7.27E+05	1.45E+10	2.80E+02	1.98E+07	1.70E+07	2.74E+06	14%	11%	16%	85%	0.14%	2.72E+01	2.34E+01	3.77E+00	1.18	0/0
T49 121210	2012-10-29	2012-12-10	4.67E+03	1.96E+06	3.92E+10	9.50E+02	2.38E+08	2.23E+08	1.49E+07	6.3%	11%	12%	92%	0.61%	1.21E+02	1.14E+02	7.59E+00	1.15	0/0
Sum	2011-07-11	2012-12-10	3.64E+06	1.56E+07	3.97E+11	1.35E+04	2.25E+09	2.16E+09	9.72E+07	4.3%	4.8%	4.9%	17%	0.57%	1.17E+02	1.12E+02	5.06E+00	1.10	0/10

Table A3-32. Summary of accumulation of C-14 in CCU resins for O1 and O2: Part 3. The tank fillings with organic values based on ¼ of the detection limit are all below the average organic accumulation. (O12-5, sheet Acc).



				Part
				1+2+3
				Sept 2008
01+02	Part 1	Part 2 *	Part 3	Dec 2012
Equivalent Full Power Hours (EFPH), O1	5338	2399	2649	10385
Equivalent Full Power Hours (EFPH), O2	6333	3673	8654	18660
Equivalent Full Power Years (EFPY), O1	0.61	0.27	0.30	1.19
Equivalent Full Power Years (EFPY), O2	0.72	0.42	0.99	2.13
Equivalent Full Power Years (EFPY), O1 + O2	1.33	0.69	1.29	3.32
Body-Feed Factor, Average (BFF)	1.06	1.17	1.10	1.11
Dry Solids Correction Factor in Sample				
Organic fraction	3.0%	4.2%	4.3%	4.0%
Total accumulation in CCU	0.30%	0.63%	0.57%	0.54%
Organic accumulation in CCU	0.0090%	0.026%	0.024%	0.022%
Total Bq/MWhth	6.51E+01	1.33E+02	1.17E+02	1.14E+02
Inorganic Bq/MWhth	6.32E+01	1.27E+02	1.12E+02	1.09E+02
Organic Bq/MWhth	1.93E+00	5.58E+00	5.06E+00	4.56E+00
Total Std. Dev.	5.8%	4.1%	4.8%	2.8%
Inorganic Std. Dev.	5.9%	4.1%	4.9%	2.8%
Organic Std. Dev.	7.5%	7.6%	17%	7.8%





Figure A3-34. Summary of accumulation of C-14 in CCU resins for O1 andO2: Part 1, 2 and 3, compared with results from Ref. 4. (O12-5, sheet D Summary).



### A3.5. Part 4

### A3.5.1. Input Data and Calculations

The fourth sampling campaign was performed between  $2013-01-22^{57}$  and 2013-07-08. The sampling record was delivered as Excel-books in the same way as for previous parts. *Table A3-1* shows an example of such a record. All calculations are included in an excel-book, *Attachment File O12-7*. The tables and figures for Part 4 are numbered from 41 and upwards.

The operational time for each CCU filter is obtained from the table. Accordingly, it is possible to calculate the thermal energy produced during the operational time for each filter. It has been assumed that C-14 distributes uniformly between the filters. The production of C-14 is calculated from the thermal energy produced (*O12-7, sheets Energy month* and *Energy day*<sup>58</sup>) and the production rate obtained from the FSAR. The calculated amount is divided by 4 for O1 and by 6 for O2, to account for the number of filters in the CCU).

Sampling, analysis and calculations were performed in the same way as for previous parts above. The timelines for the samples analyzed in Part 4 are shown in *Figure A3-41*.



Figure A3-41. Timeline for the tank fillings analyzed in Part 4. The lines are separated in the vertical direction for clarity. (O12-7, sheet D Timeline).

The concentrations are presented in *Figure A3-42* and *Table A3-41*. In this campaign only 3 subsamples have used the detection limit value (divided by 4).

The calculated activity contents are shown in *Figure A3-43* and *Table A3-42*. In the calculations, a 10 % uncertainty for the resin mass has been used in addition to the uncertainties in the concentrations. The accumulation is 0.63 % of the production, which is

<sup>&</sup>lt;sup>57</sup> The filling of the first tank sampled started 2012-11-30.

<sup>&</sup>lt;sup>58</sup> It was not possible to precisely distribute the energy production to all the tanks. Therefore, the total energy production for the sampling period was used to calculate sum and average values.

the highest value together with part 2. The organic fraction is 14 %, which is the highest value so far. See the next section for a discussion on this topic.



Figure A3-42. Concentration of C-14 in CCU resin samples from O1 and O2, Part 4. (O12-7, sheet D conc).

Sample	Sample	Start	End	C-14 tot, Bq/kg dw	C-14 inorg, Bq/kg dw	C-14 org, Bq/kg dw	Org- fract.	Tot. Std. dev.	Inorg. Std. dev.	Org. Std. dev.	Inorg/ Org*
012-47	T48 130122	2012-11-30	2013-01-22	2.92E+04	2.63E+04	2.90E+03	10%	11%	9%	33%	0/0
012-48	T49 130211	2013-01-17	2013-02-11	1.05E+04	9.79E+03	7.47E+02	7.1%	11%	8%	104%	0/3
012-49	T48 130312	2013-02-06	2013-03-11	1.46E+05	1.42E+05	4.42E+03	3.1%	6.1%	7%	25%	0/0
012-50	T49 130415	2013-03-12	2013-04-15	7.51E+04	7.28E+04	2.31E+03	2.9%	10%	8%	96%	0/0
012-51	T48 130508	2013-04-26	2013-05-08	2.61E+05	2.54E+05	6.50E+03	2.5%	3%	3.8%	82%	0/0
012-52	T48 130521	2013-05-17	2013-05-21	4.95E+05	4.00E+05	9.52E+04	19%	6.2%	13%	63%	0/0
012-53	T49 130530	2013-05-03	2013-05-29	1.06E+05	9.92E+04	6.61E+03	6.1%	5.8%	3.3%	54%	0/0
012-54	T49 130613	2013-05-31	2013-06-13	7.20E+04	6.31E+04	8.90E+03	12%	2.3%	13%	95%	0/0
012-55	T48 130618	2013-05-23	2013-06-18	9.08E+04	8.82E+04	2.57E+03	2.7%	5.4%	3.0%	100%	0/0
012-56	T49 130710	2013-06-14	2013-07-08	1.83E+05	9.85E+04	8.44E+04	46%	1.9%	2%	5%	0/0
	Mean			1.47E+05	1.25E+05	2.15E+04	15%	92%	89%	160%	0/3

Table A3-41. Summary of concentrations in samples of CCU resins from O1 and O2 Part 4. (O12-7, sheet Conc).

#### A3.5.2. Discussion Part 4

Almost all CCU resins originate from unit O2, since O1 was shut down most of the time. Two samples have unusual high fractions and concentrations of organic C-14; T48 130521 (19 %, 9.52E+04 Bq/kg dw) and T49 130710 (46 % and 8.44E+04 Bq/kg dw). The former sample

was one single CCU filter from O2. The latter one was resins from the shutdown period of O2. The energy production was 0.01 EFPY for O1 and 0.37 EFPY for O2.



Figure A3-43. Activity contents in the tanks sampled in Part 4. (O12-7, sheet D Acc).

Resin uncerta	ainty					10%													
Sample	Start	End	O1 Effective energy, MWhth	O2 Effective energy, MWhth	Calc. Produc- tion, Bq	Resin, kg dw	Accum. C-14 tot, Bq	Accum. C-14 inorg, Bq	Accum. C-14 org, Bq	Org. frac- tion	Tot. std. dev.	Inorg. std. dev.	Org. std. dev.	Fract. of produc- tion	Total Bq/MWhth	Inorg. Bq/MWhth	Org. Bq/MWhth	BFF	Inorg/ Org*
T48 130122	2012-11-30	2013-01-22	0.00E+00	1.29E+06	2.57E+10	8.40E+02	2.78E+07	2.51E+07	2.77E+06	10%	15%	13%	35%	0.11%	2.16E+01	1.95E+01	2.15E+00	1.13	0/0
T49 130211	2013-01-17	2013-02-11	0.00E+00	1.92E+05	3.85E+09	5.50E+02	6.12E+06	5.69E+06	4.34E+05	7.1%	15%	13%	105%	0.16%	3.19E+01	2.96E+01	2.26E+00	1.06	0/3
T48 130312	2013-02-06	2013-03-11	0.00E+00	1.68E+06	3.35E+10	5.85E+02	1.10E+08	1.06E+08	3.31E+06	3.0%	12%	12%	27%	0.33%	6.53E+01	6.34E+01	1.97E+00	1.28	0/0
T49 130415	2013-03-12	2013-04-15	0.00E+00	1.30E+06	2.59E+10	6.60E+02	5.66E+07	5.48E+07	1.74E+06	3.1%	14%	13%	97%	0.22%	4.37E+01	4.23E+01	1.34E+00	1.14	0/0
T48 130508	2013-04-26	2013-05-08	0.00E+00	6.18E+05	1.24E+10	3.55E+02	1.04E+08	1.02E+08	2.60E+06	2.5%	10%	11%	83%	0.85%	1.69E+02	1.65E+02	4.21E+00	1.13	0/0
T48 130521	2013-05-17	2013-05-21	0.00E+00	4.81E+05	9.62E+09	1.75E+02	1.05E+08	8.45E+07	2.01E+07	19%	12%	16%	64%	1.09%	2.18E+02	1.76E+02	4.19E+01	1.21	0/0
T49 130530	2013-05-03	2013-05-29	0.00E+00	7.02E+05	1.40E+10	6.20E+02	7.12E+07	6.67E+07	4.45E+06	6.2%	12%	11%	54%	0.51%	1.01E+02	9.51E+01	6.33E+00	1.08	0/0
T49 130613	2013-05-31	2013-06-13	0.00E+00	3.80E+05	7.61E+09	4.80E+02	3.70E+07	3.24E+07	4.57E+06	12%	10%	17%	96%	0.49%	9.72E+01	8.52E+01	1.20E+01	1.07	0/0
T48 130618	2013-05-23	2013-06-18	0.00E+00	4.37E+05	8.73E+09	1.06E+03	1.01E+08	9.80E+07	2.86E+06	2.8%	11%	10%	101%	1.2%	2.31E+02	2.25E+02	6.55E+00	1.05	0/0
T49 130710	2013-06-14	2013-07-08	0.00E+00	2.33E+05	4.66E+09	6.85E+02	1.33E+08	7.14E+07	6.11E+07	46%	10%	10%	11%	2.8%	5.69E+02	3.07E+02	2.63E+02	1.06	0/0
Sum	2012-11-30	2013-07-08	0.00E+00	7.30E+06	1.19E+11	6.01E+03	7.51E+08	6.47E+08	1.04E+08	14%	4.0%	4.3%	15%	0.63%	1.26E+02	1.09E+02	1.75E+01	1.11	0.00
Energy total	2012-11-30	2013-07-08	9.10E+04	5.87E+06	From ope	rational re	ports												
Energy diff			-9.10E+04	1.44E+06															
Energy diff			-100.0%	24.5%															
	Energ	v correction	factor		8.16E-01														

Table A3-42. Summary of accumulation of C-14 in CCU resins for O1 and O2: Part 4. The tank fillings with organic values based on ¼ of the detection limit are all below the average organic accumulation. The distribution of energy production on the individual tanks was not precise; for the sums and averages the total energy produced during the period was used. (O12-7, sheet Acc).

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	Part 1	Part 2*	Part 3*	Part 4*	Part 1.4	B2 Ref 4
	Sep2008-	Sep 2010-	Jul 2011-	Jan2013-	Sep 2008-	+ BFF
01+02	Mar 2009	May 2011	Dec 2012	Jun 2013	Jun 2013	+ ds-corr.
Equivalent Full Power Hours (EFPH), O1	5338	2399	2691	66	10493	
Equivalent Full Power Hours (EFPH), O2	6333	3673	9454	3259	22719	
Equivalent Full Power Years (EFPY), O1	0.61	0.27	0.31	0.01	1.20	
Equivalent Full Power Years (EFPY), O2	0.72	0.42	1.08	0.37	2.59	
Equivalent Full Power Years (EFPY), O1 + O2	1.33	0.69	1.39	0.38	3.79	
Body-Feed Factor, Average (BFF)	1.06	1.17	1.10	1.11	1.11	1.07
Dry Solids Correction Factor in Sample						1.16
Organic fraction	3.0%	4.2%	4.3%	14%	5.2%	16%
Total accumulation in CCU	0.30%	0.63%	0.57%	0.63%	0.49%	0.50%
Organic accumulation in CCU	0.0090%	0.026%	0.024%	0.087%	0.026%	0.080%
Total Bq/MWh <sub>th</sub>	6.51E+01	1.33E+02	1.17E+02	1.26E+02	1.03E+02	1.06E+02
Inorganic Bq/MWh <sub>th</sub>	6.32E+01	1.27E+02	1.12E+02	1.09E+02	9.74E+01	8.93E+01
Organic Bq/MWh <sub>th</sub>	1.93E+00	5.58E+00	5.06E+00	1.75E+01	5.29E+00	1.70E+01
Total Std. Dev.	5.8%	4.1%	4.8%	4.0%	2.3%	
Inorganic Std. Dev.	5.9%	4.1%	4.9%	4.3%	2.3%	
Organic Std. Dev.	7.5%	7.6%	17%	15%	9.5%	
* Inert masses for O2 calculated from operation	nal time a	nd dosage	rate			

*Table A3-43. Summary of accumulation of C-14 in CCU resins for O1 andO2, Part 1, 2, 3, and 4, compared with corrected results from Ref. 4. (O12-5, sheet Summary).* 



Figure A3-44. Summary of accumulation of C-14 in CCU resins for O1 and O2: Part 1-4, compared with results from Ref. 4. (O12-5, sheet D Summary).



### A3.6.Summary and Discussion Parts 1-4

*Table A3-43* and *Figure A3-44* show a summary of the accumulation of C-14 for the four parts analyzed. The standard deviations reported include the standard deviation in the concentrations and a 10 % standard deviation in the resin masses.

The accumulation seems to stabilize around 0.5 % of the production. The organic fraction is around 5 %. Part 4 had an unusual high organic fraction; the reason for this is unclear.

Summing all activity accumulated during the four campaigns yields values for the accumulation with reasonably low standard deviations, even if there is almost a factor of two between the highest and lowest values in the respective campaigns. The energy production amounts to 1.20 EFPY for O1 and 2.59 EFPY for O2.





# APPENDIX 4 Results from R1

### A4.1. Resin Handling

All calculations for R1 are included in an excel-book. It can be found as *Attachment Files R1-* 2, 3 and -4 to this report. Reference to the various sheets will be made at tables, figures etc.



Figure A4-1. Overview of handling of spent powder resin at R1. (A-5, pict. 2).

		Fe3O4	Fe2O3	Fe3O4	Fe2O3
Fe	atomic mass	56	56	56	56
0	atomic mass	16	16	16	16
Iron oxide	mole mass	232	160	232	160
Iron oxide/Fe		1.381	1.429	1.381	1.429
Fe in condensate	ppb	10	10	10	10
ionic fraction		0.1	0.1	0.1	0.1
Op. Time	days	42	42	35	35
Flowrate	kg/s	154	154	154	154
Volume filtered	m3	5.59E+05	5.59E+05	4.66E+05	4.66E+05
Iron oxide	kg	6.9	7.2	5.8	6.0
Ionic Fe	kg	0.6	0.6	0.5	0.5
Iron oxide	kg	6.95E+00	7.19E+00	5.79E+00	5.99E+00
Ionic Fe	kg	5.59E-01	5.59E-01	4.66E-01	4.66E-01
Powder resin	kg dw/ filter	90	90	90	90
Inert material	kg dw/ filter				
Powder res.+Inert. Mater.	kg dw/ filter	90	90	90	90
Total	kg dw/ filter	9.75E+01	9.77E+01	9.63E+01	9.65E+01
Fraction (iron oxide+ionic	Fe)	7.70E-02	7.92E-02	6.50E-02	6.69E-02
Fraction powder resin		9.23E-01	9.21E-01	9.35E-01	9.33E-01
Total Body-Feed Factor (B	FF)	1.08E+00	1.09E+00	1.07E+00	1.07E+00

*Table A4-1. Calculation of the correction for iron oxide accumulation (BFF). The BFF depends on the operation time for the filter. (R1-2, sheet BodyfeedR1).* 



In the Condensate Clean-up system (CCU) of R1 (8 filters per unit), 90 kg powder resin dw (sulfonate resin in cation part) is loaded and operated for periods usually lasting 6 weeks<sup>59</sup>. When the resin is exhausted, it is flushed to a temporary tank in the unit. From this tank, the resin is transferred by flushing to tank T41 in the Waste Handling Building. A schematic overview is given in *Figure A4-1*. From the storage tank T41 resin is taken to solidification in concrete.

## A4.2. Part 1: Resins from 2009 and 2010

### A4.2.1. Resin Mass Balance

The records describing the additions and removals of resin are found in *Attachment File R1-1* as excel-sheets.

All resin masses added to T41 are given as dry weight (dw) and do not include accumulated iron oxides. Since the removals to solidification and samples are based on total dry weight, the additions of resin have to be corrected. The calculation is based on the accumulation of iron oxides. *Table A4-1* shows a summary of such accumulation, which yields a single correction factor, here called the Body-Feed Factor (BFF), which is used to multiply the additions of powder resin to obtain the total dry weight added (*R1-2, Sheet BodyfeedR1*).



Figure A4-2. The log data for resin storage tank T41 in graphics. The sample size in grams is read on the right axis. The symbols for batches indicate the time for the last sample in the batch, not the size of the batch. (R1-2, sheet D T41; data are in Sheet T41).

<sup>&</sup>lt;sup>59</sup> From the cycle 2013-2014 the operating time is determined by the pressure drop, 1.5 bar. The average time during that cycle was 60 days.

*Figure A4-2* shows the additions, removals and the inventory in T41. In addition, the samples withdrawn for analyses are also indicated. All weights for additions, removal and inventory are given as total dw. Sample size is given as ww. The batches are indicated at the last sample included in the batch.

The plant was shut-down between 2009-03-15 and 2010-03-06 for an extensive refurbishment. The resins added to T41 from 2009-10-01 to 2010-03-08 have not been exposed to C-14 from operation. They either originate from failed filter loadings or from cleaning the condensate system water during start-up.

#### A4.2.2. Mass balance and Calculations for C-14 (Calculation model MBM)

To calculate the amounts of C-14 disposed in solidified waste, a mass balance over the tank T41 is calculated. In early January 2009 the tank had a small amount of low activity resin. The operation period in this year lasted only to mid-March, when the unit shut down for extensive refurbishment works, lasting to March 2010. During operation and the next few weeks following shut-down, resin was disposed into T41. *Figure A4-2* indicates each addition and removal of resin for solidification.



Figure A4-3. Schematic mass balance over tank T41. (A-5, pict. 11).

Samples were withdrawn for analysis in connection with the solidifications or backflushings (but not all of them). The samplings are indicated (open circles) in *Figure A4-2*; totally 51 in 2009 and 2010. Most of the samples were combined to batches for analysis; the final date for a batch is indicated by triangles. Since the tank content is mixed by air bubbling and by pumping, it is assumed that the content is fairly homogeneous.

In 2010, operation started in March and continued to early October with a break in July and August for inspection works. The resin sampling and mass balancing were continued to the end of November, when T41 was almost empty.

*Table A4-2* and *Figure A4-4* show the concentrations obtained in the analyses of the batched samples. These concentrations are used to calculate the quantities of C-14 brought to solidification.

*Table A4-3A* shows the calculated quantities of C-14 in the solidified resins and the residual amount of resin in T41 in 2009.





*Figure A4-4. Concentrations of C-14 used for calculation of the quantities of C-14 in the powder resins*<sup>60</sup>. (*R1-2, sheet D conc*).

			Sample Weighting	Total, Ba/ka	Inora	Org			SD inorg	
Batch	Start date	End date	factor	dw	Bq/kg dw	Bq/kg dw	Org, %	SD tot, %	%	SD org, %
R1-09B1		2009-01-23	1.00	2.43E+04	2.04E+04	3.86E+03	16%	5.8%	13%	58%
R1-09B2	2009-01-26	2009-03-06	1.00	3.32E+06	3.27E+06	4.54E+04	1.4%	4.6%	4%	30%
R1-09B3	2009-03-13	2009-03-19	1.00	3.90E+06	3.85E+06	4.98E+04	1.3%	2.5%	2.9%	28%
R1-10B1		2010-03-10	1.00	7.37E+03	6.74E+03	6.28E+02	8.5%	13%	11%	103%
R1-10B2	2010-03-15	2010-04-21	1.00	3.87E+05	2.75E+05	1.12E+05	29%	24%	38%	68%
R1-10B3	2010-04-22	2010-06-02	1.00	1.96E+06	1.93E+06	3.34E+04	1.7%	1.1%	1.1%	18%
R1-10B4	2010-06-02	2010-07-19	1.00	2.37E+06	2.33E+06	4.12E+04	1.7%	2.0%	1.7%	14%
R1-10B5	2010-07-19	2010-08-03	1.00	2.15E+06	2.12E+06	3.24E+04	1.5%	1.4%	1.1%	34%
R1-10B6	2010-08-04	2010-10-01	1.00	1.79E+06	1.77E+06	1.82E+04	1.0%	1.7%	2.0%	27%
R1-10B7	2010-10-02	2010-11-29	1.00	1.66E+06	1.64E+06	2.53E+04	1.5%	2.8%	3.0%	22%

Table A4-2. Concentration data used in the calculations. The sample weighting factor was introduced in order to handle dilution effects from non-radioactive resins; however, with the present calculation model it has not been used. (R1-2, sheet R1acc).

The production of C-14 in R1 has been calculated from the production rate given in the FSAR and the production of thermal energy obtained from the operational reports. The energy production is calculated as the difference of the cumulated production by the end of March and the beginning of January 2009. The calculation is to be found in *Table A4-3C*. *Table A4-3A* summarizes the results for 2009. *Table A4-3B* summarizes the results for 2010, using the

<sup>&</sup>lt;sup>60</sup> The low concentrations in the beginning and the middle of the diagram are explained in the following way: Due to long shut-downs, the storage tank had been emptied from resins used in operation. The resins in the tank were either fresh resins from failed loadings of the filters or resins used to clean condensate system water prior to start. Cf. tank inventory in Figure A4-2.



cumulated energy production between March and November 2010. These values are also found in *Table A4-3C*.

Resin unce	ertainty		10%				
				% of			
Batch	Start date	End date	kg dw	total	Total Bq	Inorg, Bq	Org Bq
	Initial						
R1-09B1	inventory	2009-01-23	-200	-0.1%	-4.86E+06	-4.09E+06	-7.72E+05
R1-09B2	2009-01-26	2009-03-06	570	30.6%	1.89E+09	1.87E+09	2.59E+07
R1-09B3	2009-03-13	2009-03-19	100	6.3%	3.90E+08	3.85E+08	4.98E+06
	Residual						
R1-09B3	inventory	2009-03-26	1000	63.1%	3.90E+09	3.85E+09	4.98E+07
Sum			1470	100.0%	6.17E+09	6.09E+09	7.99E+07
% of produ	iction				6.7%	6.7%	0.087%
Bq/MWHth	l				1.59E+03	1.57E+03	2.05E+01
% organic							1.3%
Standard I	Deviation, %	D			7.4%	7.4%	21%
Sum with	BFF=	1.09			6.71E+09	6.62E+09	8.68E+07
% of production					7.3%	7.2%	0.095%
Bq/MWHth					1.72E+03	1.70E+03	2.23E+01

Table A4-3A. Summary of C-14 in CCU resins at R1 produced in the period January-March 2009. (R1-2, sheet R1acc).

Resin unc	ertainty		10%				
				% of			
Batch	Start date	End date	kg dw	total	Total Bq	Inorg, Bq	Org Bq
	Initial						
R1-10B1	inventory	2010-03-10	-690	-0.1%	-5.09E+06	-4.65E+06	-4.34E+05
R1-10B2	2010-03-15	2010-04-21	520	2.2%	2.01E+08	1.43E+08	5.80E+07
R1-10B3	2010-04-22	2010-06-02	975	21.2%	1.92E+09	1.88E+09	3.26E+07
R1-10B4	2010-06-02	2010-07-19	410	10.8%	9.72E+08	9.55E+08	1.69E+07
R1-10B5	2010-07-19	2010-08-03	345	8.2%	7.43E+08	7.32E+08	1.12E+07
R1-10B6	2010-08-04	2010-10-01	1180	23.4%	2.11E+09	2.09E+09	2.14E+07
R1-10B7	2010-10-02	2010-11-29	1760	32.5%	2.93E+09	2.88E+09	4.46E+07
	Residual						
R1-10B7	inventory	2010-11-29	90	1.7%	1.50E+08	1.47E+08	2.28E+06
Sum			4590	100.0%	9.02E+09	8.83E+09	1.87E+08
% of produ	uction				3.6%	3.5%	0.074%
Bq/MWHth	1				8.41E+02	8.23E+02	1.74E+01
% organic							2.1%
Standard	Deviation, %	D			4.9%	4.9%	23%
Sum with	BFF=	1.09			9.79E+09	9.59E+09	2.03E+08
% of produ	uction				3.9%	3.8%	0.080%
Bq/MWHth					9.13E+02	8.94E+02	1.89E+01

Table A4-3B. Summary of C-14 in CCU resins at R1 produced in the period March-October 2010. (R1-2, sheet R1acc).

The calculation is performed in the following way: The C-14 initially present in T41 is calculated from the first sample and the tank inventory of resin. It is indicated by a minus-sign in the tables, since it was produced outside of the production period. The removals of resin to solidification corresponding to a batch are combined to calculate the amount of C-14. When the last batch has been analyzed, the residual resin in T41 is assumed to have the same



concentration as the last batch. *Table A4-3D* shows the average accumulation for the years 2009 and 2010. The uncertainty (standard deviation) in the calculated accumulation includes the uncertainty in the sample concentrations and 10 % uncertainty for the resin mass.

Start	End	MWhth start	MWhth end	MWhth	C-14 produc- tion rate, Bq/MWHth	Calculated production Bq total	Equivalent Full Power Hours, (EFPH)	Equivalent Full Power Years, (EFPY)
2009-01-01	2009-03-19	4.85E+08	4.89E+08	3892369	2.35E+04	9.16E+10	1497	0.17
2010-03-01	2010-11-30	4.89E+08	5.00E+08	10724615	2.35E+04	2.52E+11	4125	0.47
Sum				14616984	2.35E+04	3.44E+11	5622	0.64

*Table A4-3C. Summary of energy production and calculated production of C-14 in R1 in January-March 2009 and March-November 2010. (R1-2, sheet R1acc).* 

								Fraction									
			Calc.	Accum.	Accum.	Accum.	Org.	of		Tot.		Inorg.		Org.			1 1
			Produc-	C14-tot,	C14-	C14-org,	Frac-	produc-	Total	std.	Inorg.	std.	Org.	std.			
Start	End	MWhth	tion, Bq	Bq	inorg, Bq	Bq	tion	tion	Bq/MWhth	dev.	Bq/MWhth	dev.	Bq/MWhth	dev.	EFPH	EFPY	BFF
2009-01-01	2009-03-19	3.89E+06	9.16E+10	6.71E+09	6.62E+09	8.68E+07	1.3%	7.3%	1.72E+03	7.4%	1.70E+03	7.4%	2.23E+01	21%	1497	0.17	1.09
2010-03-01	2010-11-30	1.07E+07	2.52E+11	9.79E+09	9.59E+09	2.03E+08	2.1%	3.9%	9.13E+02	4.9%	8.94E+02	3.8%	1.89E+01	23%	4125	0.47	1.09
Sum		1.46E+07	3.44E+11	1.65E+10	1.62E+10	2.89E+08	1.8%	4.8%	1.13E+03	4.2%	1.11E+03	3.8%	1.98E+01	17%	5622	0.64	

Table A4-3D. Summary of combined accumulation of C-14 in CCU resins for 2009 and 2010. (R1-2, sheet R1Big sum).

### A4.3. Part 2: Resins from 2011



Figure A4-21. The log data for resin storage tank T41 for 2011 in graphics. The sample volume of slurry, multiplied by 100, is read on the right axis (liters). The symbols for batches (triangles on a constant level) indicate the samples in the batch, not the size of the batch. (R1-3, sheet D T41; data are in Sheet T41).



#### A4.3.1. Resin Mass Balance and Calculations

The records describing the additions and removals of resin are found in *Attachment File R1-3*, together with the calculations. The tables and figures for Part 2 are numbered from 21 and upwards. All analyses and calculations were performed in the same way as for part 1.

The samplings started in early April 2011, about 3.5 months after the start of the fuel cycle. This delay was caused by a misunderstanding regarding the sampling program. *Figure A4-21* shows the mass balance and samplings. *Figure A4-22* and *Table A4-21* show the concentration values.



Figure A4-22. Concentrations of C-14 used for calculation of the quantities of C-14 in the powder resins. (R1-3, sheet D conc).

Batch	Start date	End date	Sample Weighting factor	Total, Bq/kg dw	Inorg, Bq/kg dw	Org, Bq/kg dw	Org, %	SD tot, %	SD inorg, %	SD org, %
R1-11B1		2011-04-04	1.00	3.26E+06	3.22E+06	3.44E+04	1.3%	1.1%	1.1%	3.2%
R1-11B2	2011-04-04	2011-04-29	1.00	2.97E+06	2.93E+06	3.95E+04	1.3%	1.1%	1.2%	2.9%
R1-11B3	2011-04-29	2011-06-01	1.00	3.30E+06	3.27E+06	2.72E+04	0.83%	1.1%	1.1%	3.8%
R1-11B4	2011-06-01	2011-06-29	1.00	3.46E+06	3.43E+06	2.87E+04	0.82%	1.1%	1.1%	3.7%
R1-11B5	2011-06-29	2011-07-28	1.00	3.31E+06	3.27E+06	4.30E+04	1.3%	1.1%	1.1%	3.1%
R1-11B6	2011-07-28	2011-09-01	1.00	3.37E+06	3.33E+06	4.18E+04	1.2%	1.1%	1.1%	2.9%
R1-11B7	2011-09-01	2011-09-22	1.00	2.98E+06	2.95E+06	2.91E+04	1.0%	1.1%	1.1%	3.5%
R1-11B8	2011-09-22	2011-09-29	1.00	2.68E+06	2.64E+06	3.28E+04	1.2%	2.6%	2.0%	60%

Table A4-21. Concentration data used in the calculations for the 2011 samplings. The sample weighting factor was introduced in order to handle dilution effects from non-radioactive resins; however, with the present calculation model it has not been used. (R1-3, sheet R1acc).



Start	End	MWhth start	MWhth end	MWhth	C-14 produc- tion rate, Bq/MWHth	Calculated production Bq total	Equivalent Full Power Hours, (EFPH)	Equivalent Full Power Years, (EFPY)
2011-03-31	2011-09-30	5.05E+08	5.16E+08	10707363	2.35E+04	2.52E+11	4118	0.47
2010-02-28	2011-03-31	*		1017531	2.35E+04	2.40E+10	391	0.04
2011-03-31	2011-09-30			11724894	2.35E+04	2.76E+11	4510	0.51
* Single filter	rs in the begin	ning of the s						

Table A4-22. Summary of energy production and calculated production of C-14 in<br/>R1 in April-September 2011. (R1-3, sheet R1acc).

*Table A4-22* shows the energy production during the sampling period. Since some of the filters started their operation before the sampling period, an individual calculation of the "energy exposure" before 2012-04-01 was performed for these filters.

*Table A4-23* shows the accumulated amounts of C-14 during the 2011 sampling period. The BFF should have been 1.07, but due to some uncertainties in the record keeping, 1.09 has been used to compensate for possible loss of one or two filter batches. *Table A4-24* summarizes the accumulation for all the years 2009-2011.

Resin Unc	esin Uncertainty		10%				
				% of			
Batch	Start date	End date	kg dw	total	Total Bq	Inorg, Bq	Org Bq
	Initial						
R1-11B1	inventory	2011-04-04	-890	-25%	-2.90E+09	-2.87E+09	-3.06E+07
R1-11B2	2011-04-04	2011-04-29	85	2.2%	2.52E+08	2.49E+08	3.36E+06
R1-11B3	2011-04-29	2011-06-01	250	7.1%	8.25E+08	8.18E+08	6.80E+06
R1-11B4	2011-06-01	2011-06-29	0	0.0%	0.00E+00	0.00E+00	0.00E+00
R1-11B5	2011-06-29	2011-07-28	125	3.6%	4.14E+08	4.08E+08	5.37E+06
R1-11B6	2011-07-28	2011-09-01	510	15%	1.72E+09	1.70E+09	2.13E+07
R1-11B7	2011-09-01	2011-09-22	430	11%	1.28E+09	1.27E+09	1.25E+07
R1-11B8	2011-09-01	2011-09-22	215	5.0%	5.75E+08	5.68E+08	7.06E+06
	Residual						
R1-11B8	inventory	2011-09-29	3505	81%	9.38E+09	9.26E+09	1.15E+08
Sum			5120	100%	1.15E+10	1.14E+10	1.41E+08
% of produ	iction				4.2%	4.1%	0.051%
Bq/MWHth	l				9.85E+02	9.73E+02	1.20E+01
% organic							1.2%
Standard I	tandard Deviation, %				9.0%	8.9%	50%
Sum with	BFF=	1.09			1.26E+10	1.25E+10	1.54E+08
% of produ	iction				4.6%	4.5%	0.056%
Bq/MWHth					1.08E+03	1.06E+03	1.31E+01

Table A4-23. Summary of C-14 in CCU resins at R1 accumulated in the period April-September 2011. (R1-3, sheet R1acc).

Start	End	MWhth	Calc. Produc- tion, Bq	Accum. C14-tot, Bq	Accum. C14- inorg, Bq	Accum. C14-org, Bq	Org. Frac- tion	Frac- tion of produc- tion	Total Bq/MWhth	Tot. std. dev.	Inorg. Bq/MWhth	Inorg . std. dev.	Org. Bq/MWhth	Org. std. dev.	EFPH	EFPY	BFF
2009-01-01	2009-03-19	3.89E+06	9.16E+10	6.71E+09	6.62E+09	8.68E+07	1.3%	7.3%	1.72E+03	7.4%	1.70E+03	7.4%	2.23E+01	21%	1497	0.17	1.09
2010-03-01	2010-11-30	1.07E+07	2.52E+11	9.79E+09	9.59E+09	2.03E+08	2.1%	3.9%	9.13E+02	4.9%	8.94E+02	3.8%	1.89E+01	23%	4125	0.47	1.09
2011-03-31	2011-09-30	1.17E+07	2.76E+11	1.26E+10	1.25E+10	1.54E+08	1.2%	4.6%	1.08E+03	9.0%	1.06E+03	8.9%	1.31E+01	50%	4510	0.51	1.09
Sum		2.63E+07	6.20E+11	2.91E+10	2.87E+10	4.43E+08	1.5%	4.7%	1.10E+03	4.6%	1.09E+03	4.4%	1.68E+01	21%	10131	1.16	

Table A4-24. Summary of combined accumulation of C-14 in CCU resins for 2009, 2010 and 2011(R1-2, sheet Big sum).





### A4.3.2. Discussion Part 1 and Part 2

	Mass balance T41	Mass balance T41	Mass balance T41	Mass balance T41
Unit	R1	R1	R1	R1
Year	2009	2010	2011	2009-2011
Equivalent Full Power Hours	1497	4125	4510	10131
Equivalent Full Power Years	0.17	0.47	0.51	1.16
Body-Feed Factor, Average (BFF)	1.09	1.09	1.09	
Dry Solids Correction Factor in Sample				
Organic fraction	1.3%	2.1%	1.2%	1.5%
Total accumulation in CCU	7.3%	3.9%	4.6%	4.7%
Organic accumulation in CCU	0.095%	0.080%	0.056%	0.071%
Total Bq/MWhth	1.72E+03	9.13E+02	1.08E+03	1.10E+03
Inorganic Bq/MWhth	1.70E+03	8.94E+02	1.06E+03	1.09E+03
Organic Bq/MWhth	2.23E+01	1.89E+01	1.31E+01	1.68E+01
Total Std. Dev.	7.4%	4.9%	9.0%	4.6%
Inorganic Std. Dev.	7.4%	3.8%	8.9%	4.4%
Organic Std. Dev.	21%	23%	50%	21%

Table A4-25. Summary of accumulation of C-14 in CCU resins at R1 (R1-2, sheet Summary).

The record keeping for the resins handled during the period seems to be is adequate with a few exceptions. The only uncertainty is the Body-Feed Factor, BFF, which accounts for accumulation of iron oxides. It has been calculated in *Table A4-2* and has the value 1.09, which has been included in the calculations.

*Table A4-25* gives a summary of the accumulation of C-14 in the CCU resins separately for the three periods and combined. It can be seen that the amount accumulated varies a factor of two between the years. The organic fraction also shows this variation. The variation is mainly in the inorganic part. A likely explanation for this is discussed below.

The standard deviation includes the uncertainty in counting and a 10 % uncertainty from the determination of resin masses.

Furthermore, the mass of the resin in this study is based on actual record keeping (although corrected for calculated body-feed).

The organic fraction is in the range 1-5 %, where most of the BWR samples are found.

The concentrations are low at the beginning of the periods in 2009 and 2010, since there is some initial amount of resin, which has not been in operation. This initial amount will dilute the concentration in the additions of resin. The diagrams show that the concentration increases during the cycle and levels out at the end. In 2011, the sampling started several months into the fuel cycle, when the concentration had stabilized.

The total accumulation of C-14 is governed by the inorganic fraction. In 2010 it is lower than in 2009, with 2011 in between. The explanation to this may be the air bubbling, where



## A4.4. Part 3: Resins from 2012 and 2013

#### A4.4.1. Resin Mass Balance and Calculations

The records describing the additions and removals of resin are found in *Attachment File R1-4*, together with the calculations. The tables and figures for Part 3 are numbered from 31 and upwards. All analyses and calculations were performed in the same way as for part 1 and 2.



*Figure A4-31. The log data for resin storage tank T41 for 2012-13 in graphics. (R1-4, sheet D T41; data are in Sheet T41).* 

The samplings started in November 2011 and continued to the end of April 2013 with a break for the annual outage in August-September 2012. Since there are good records of the mass handling and frequent sampling, the accumulation has been calculated for each fuel cycle. *Figure A4-31* shows the mass balance for powder resins from 2009 to mid-June 2013. *Figure A4-32, Table A4-31* and *Table A4-33* show the concentration values.

<sup>&</sup>lt;sup>61</sup> Air bubbling tests performed in T41 did not show any significant release of C-14. The reason for this is unclear; the high operating temperature and the thermal degradation of the anion resin may be one contributing factor. The pH-value in the resin slurry another.


			Sample	Total,						
			Weighting	Bq/kg	Inorg,	Org,			SD inorg,	
Batch	Start date	End date	factor	dw	Bq/kg dw	Bq/kg dw	Org, %	SD tot, %	%	SD org, %
R1-12B1	2011-11-21	2011-11-21	1.00	3.00E+06	2.98E+06	2.37E+04	0.77%	3.1%	2.7%	66%
R1-12B2	2011-11-26	2011-11-26	1.00	2.36E+06	2.14E+06	2.19E+05	12%	18%	30%	97%
R1-12B2:2	2011-11-29	2011-11-29	1.00	2.49E+06	2.46E+06	2.59E+04	1.1%	5.7%	6.2%	53%
R1-12B3	2011-12-01	2011-12-29	1.00	2.12E+06	2.09E+06	2.69E+04	1.3%	14%	15%	23%
R1-12B4	2012-01-09	2012-02-03	1.00	2.49E+06	2.48E+06	1.32E+04	0.53%	1.1%	1.1%	25%
R1-12B5	2012-02-10	2012-03-02	1.00	1.96E+06	1.95E+06	9.49E+03	0.49%	11%	11%	7.6%
R1-12B6	2012-03-08	2012-03-08	1.00	2.38E+06	2.35E+06	2.26E+04	0.95%	2.2%	2.0%	15%
R1-12B6:2	2012-03-08	2012-03-08	1.00	2.50E+06	2.44E+06	6.05E+04	2.4%	2.3%	2.0%	19%
R1-12B6:3	2012-03-08	2012-03-08	1.00	2.54E+06	2.48E+06	5.36E+04	2.1%	0.91%	0.93%	16%
R1-12B7	2012-03-16	2012-04-28	1.00	2.63E+06	2.48E+06	1.56E+05	5.9%	1.5%	5.1%	76%
R1-12B8	2012-05-12	2012-05-12	1.00	2.35E+06	2.29E+06	6.09E+04	2.6%	1.1%	1.1%	22%
R1-12B9	2012-07-12	2012-07-12	1.00	2.20E+06	2.03E+06	1.68E+05	7.6%	1.8%	8.4%	109%
R1-12B10	2012-07-20	2012-08-24	1.00	2.20E+06	2.15E+06	4.50E+04	2.0%	2.6%	2.5%	8.8%
R1-12B11	2012-08-31	2012-08-31	1.00	1.96E+06	1.93E+06	3.14E+04	1.5%	20%	19%	59%

Table A4-31. Concentration data used in the calculations for the 2012 samplings. The sample<br/>weighting factor was introduced in order to handle dilution effects from non-radioactive<br/>resins; however, with the present calculation model it has not been used.<br/>(R1-4, sheet R1acc aug 2012).



Figure A4-32. Concentrations of C-14 used for calculation of the quantities of C-14 in the powder resins. See footnote to Figure A4-4 for comments on the low concentrations. (R1-4, sheet D conc).

*Table A4-32* and *Table A4-34* show the energy production during the sampling period. *Table A4-35* and *Table A4-36* show the accumulated amounts of C-14 during the 2011-2013 sampling period. The BFF has been set to 1.09 in accordance with the previous parts.

	Start	End	MWhth start	MWhth end	MWhth	C-14 produc- tion rate, Bq/MWHth	Calculated production Bq total	Equiv. Full Power Hours, (EFPH)	Equiv. Full Power Years, (EFPY)
1	2011-11-01	2012-08-31	5.16E+08	5.27E+08	11094852	2.35E+04	2.61E+11	4267	0.49

Table A4-32. Summary of energy production and calculated production of C-14 inR1 in November 2011-August 2012. (R1-4, sheet R1acc aug 2012).

			Sample Weighting	Total, Bq/kg	Inorg,	Org,			SD inorg,	
Batch	Start date	End date	factor	dw	Bq/kg dw	Bq/kg dw	Org, %	SD tot, %	%	SD org, %
R1-12B11	2012-08-31	2012-08-31	1.00	1.96E+06	1.93E+06	3.14E+04	1.5%	20%	19%	59%
R1-12B12	2012-09-17	2012-11-01	1.00	2.09E+06	2.06E+06	2.66E+04	1.3%	1.4%	1.5%	9.9%
R1-12B13	2012-11-16	2012-12-28	1.00	2.35E+06	2.17E+06	1.70E+05	7.3%	1.6%	5.2%	60%
R1-12B14	2013-01-10	2013-01-25	1.00	2.46E+06	2.42E+06	4.69E+04	1.9%	1.6%	2.1%	55%
R1-12B15	2013-02-08	2013-02-22	1.00	2.45E+06	2.42E+06	3.29E+04	1.3%	1.4%	1.4%	40%
R1-12B16	2013-03-22	2013-04-14	1.00	2.70E+06	2.69E+06	1.90E+04	0.70%	1.1%	1.1%	18%
R1-12B17	2013-04-30	2013-04-30	1.00	2.98E+06	2.47E+06	5.13E+05	13%	22%	1.1%	132%
Mean				2.43E+06	2.31E+06	1.20E+05	4.9%	32%	17%	573%
Geom. Mea	an			2.41E+06	2.30E+06	5.77E+04	2.4%			
Max				2.98E+06	2.69E+06	5.13E+05	17%	22%	1%	132%
Min				1.96E+06	1.93E+06	1.90E+04	1.0%	20%	19%	18%
Median cor	nc.			2.45E+06	2.42E+06	3.29E+04	1.3%			

Table A4-33. Concentration data used in the calculations for the 2012and 2013 samplings.The sample weighting factor was introduced in order to handle dilution effects from non-<br/>radioactive resins; however, with the present calculation model it has not been used.<br/>(R1-4, sheet R1acc aug12 apr2013).

Start	End	MWhth start	MWhth end	MWhth	C-14 produc- tion rate, Bq/MWHth	Calcula- ted produc- tion, Bq total	Equiv. Full Power Hours, (EFPH)	Equiv. Full Power Years, (EFPY)
2012-08-31	2013-04-30	5 27E+08	5 41E+08	13611601	$2.35E \pm 0.4$	3 20E+11	5235	0.60

*Table A4-34. Summary of energy production and calculated production of C-14 in R1 in August 2012 - April 2013. (R1-4, sheet R1acc aug12 apr2013).* 



Resin unce	ertainty		10%				
				% of			
Batch	Start date	End date	kg dw	total	Total Bq	Inorg, Bq	Org Bq
	Initial						
R1-12B1	inventory	2011-11-21	-1615	-50%	-4.85E+09	-4.81E+09	-3.82E+07
R1-12B2	2011-11-21	2011-11-30	435	11%	1.03E+09	9.31E+08	9.54E+07
R1-12B2:2	2011-11-21	2011-11-30	0	0%	0.00E+00	0.00E+00	0.00E+00
R1-12B3	2011-11-30	2012-01-04	225	5.0%	4.77E+08	4.71E+08	6.05E+06
R1-12B4	2012-01-04	2012-02-08	580	15%	1.44E+09	1.44E+09	7.63E+06
R1-12B5	2012-02-08	2012-03-02	390	7.9%	7.64E+08	7.61E+08	3.70E+06
R1-12B6	2012-03-02	2012-03-08	17	0.41%	3.96E+07	3.92E+07	3.77E+05
R1-12B6:2	2012-03-02	2012-03-08	17	0.43%	4.17E+07	4.06E+07	1.01E+06
R1-12B6:3	2012-03-02	2012-03-08	17	0.44%	4.23E+07	4.14E+07	8.94E+05
R1-12B7	2012-03-08	2012-05-08	765	21%	2.01E+09	1.90E+09	1.20E+08
R1-12B8	2012-05-08	2012-07-04	700	17%	1.64E+09	1.60E+09	4.26E+07
R1-12B9	2012-07-04	2012-07-18	0	0%	0.00E+00	0.00E+00	0.00E+00
R1-12B10	2012-07-18	2012-08-31	810	18%	1.78E+09	1.74E+09	3.65E+07
R1-12B11	2012-08-31	2012-09-13	100	2.0%	1.96E+08	1.93E+08	3.14E+06
Residual			2555	52%	5.02E+09	4.94E+09	8.02E+07
Sum			4995	100%	9.63E+09	9.28E+09	3.59E+08
% of produ	ction				3.7%	3.6%	0.14%
Bq/MWh <sub>th</sub>					8.68E+02	8.36E+02	3.23E+01
% organic							3.7%
Standard I	Deviation, %	)			13%	14%	40%
Sum with I	BFF=	1.09			1.05E+10	1.01E+10	3.92E+08
% of produ	ction				4.0%	3.9%	0.15%
Bq/MWh <sub>th</sub>					9.48E+02	9.13E+02	3.53E+01

Table A4-35. Summary of C-14 in CCU resins at R1 accumulated in the period November 2011-August 2012 (R1-4, sheet R1acc aug 2012)

Resin unce	ertainty		10%				
Batch	Start date	End date	kg dw	% of total	Total Bq	Inorg, Bq	Org Bq
	Initial						
R1-12B11	inventory	2012-08-31	-2565	-37%	-5.04E+09	-4.96E+09	-8.05E+07
R1-12B12	2012-09-17	2012-11-01	485	7.4%	1.01E+09	9.99E+08	1.29E+07
R1-12B13	2012-11-16	2012-12-28	875	15%	2.05E+09	1.90E+09	1.49E+08
R1-12B14	2013-01-10	2013-01-25	1225	22%	3.02E+09	2.96E+09	5.75E+07
R1-12B15	2013-02-08	2013-02-22	0	0%	0.00E+00	0.00E+00	0.00E+00
R1-12B16	2013-03-22	2013-04-14	250	5.0%	6.76E+08	6.71E+08	4.75E+06
R1-12B16	2013-04-30	2013-04-30	125	2.5%	3.38E+08	3.36E+08	2.37E+06
R1-12B17	Residual	2013-04-30	4285	85%	1.16E+10	1.15E+10	8.14E+07
Sum			4680	100%	1.36E+10	1.34E+10	2.27E+08
% of produ	iction				4.3%	4.2%	0.071%
Bq/MWh <sub>th</sub>					1.00E+03	9.86E+02	1.67E+01
% organic							1.7%
Standard I	Deviation, %	, D			22%	12%	67%
Sum with	BFF=	1.09			1.49E+10	1.46E+10	2.48E+08
% of produ	iction				4.6%	4.6%	0.077%
Bq/MWh <sub>th</sub>					1.09E+03	1.08E+03	1.82E+01

Table A4-36. Summary of C-14 in CCU resins at R1 accumulated in the period August 2012 - April 2013. (R1-4, sheet R1acc aug12 apr2013).



Table A4-37 and Figure A4-33 summarize the accumulation for all the years 2009-2013.

From the *Table A4-37* and *Figure A4-33* it can be seen that the accumulation seems to stabilize around 4.5 % of the production with an organic fraction of 2 %. There is no obvious indication that the new configuration of the CCU with forward pumping and lower operating temperature has had any effect on the accumulation of C-14.

	Mass balance	Mass balance	Mass balance	Mass balance	Mass balance	Mass balance
	T41	T41	T41	T41	T41	T41
Unit	R1	R1	R1	R1	R1	R1
						Mean
				2011- Aug	Aug 2012-	2009-Apr
Year	2009	2010	2011	2012	Apr 2013	2013
Equivalent Full Power Hours	1497	4125	4510	4267	5235	14399
Equivalent Full Power Years	0.17	0.47	0.51	0.49	0.60	2.24
Body-Feed Factor, Average (BFF)	1.09	1.09	1.09	1.09	1.09	1.09
Dry Solids Correction Factor in Sample						
Organic fraction	1.3%	2.1%	1.2%	3.7%	1.7%	2.0%
Total accumulation in CCU	7.3%	3.9%	4.6%	4.0%	4.6%	4.5%
Organic accumulation in CCU	0.095%	0.080%	0.056%	0.15%	0.077%	0.090%
Total Bq/MWhth	1.72E+03	9.13E+02	1.08E+03	9.48E+02	1.09E+03	1.07E+03
Inorganic Bq/MWhth	1.70E+03	8.94E+02	1.06E+03	9.13E+02	1.08E+03	1.05E+03
Organic Bq/MWhth	2.23E+01	1.89E+01	1.31E+01	3.53E+01	1.82E+01	2.12E+01
Total Std. Dev.	7.4%	4.9%	9.0%	13%	22%	7.1%
Inorganic Std. Dev.	7.4%	3.8%	8.9%	14%	12%	4.8%
Organic Std. Dev.	21%	23%	50%	40%	67%	23%

Table A4-37. Summary of accumulation of C-14 in CCU resins at R1 (R1-4, sheet Summary).

#### A4.4.2. Discussion Part 1, 2 and 3

The cycle starting in September 2012 introduced one major change in the condensate system for one turbine: The CCU system was reconfigured to operation at a significantly lower temperature, in order to reduce the thermal breakdown of the resin. This breakdown introduces sulfate ions into the reactor, which are considered to contribute to intergranular stress corrosion cracking. The high temperature drains are not cleaned by the CCU, but are introduced into the feed water at an appropriate point from a temperature point of view. This process is called Forward Pumping (FP) and increases the thermal efficiency of the plant. It may also introduce more corrosion products into the reactor coolant.





Figure A4-33. Summary of accumulation of C-14 in CCU resins at R1 (R1-4, sheet D Summary).

The accumulation of C-14 increased from 4.0 % to 4.6 % of the production, compared with the previous cycle. However, it is too early to attribute this increase to the FP, since earlier cycles have had the same accumulation (2011) or even higher (2009).

One effect of the FP has been observed: Spent CCU resins are used to clean some waste water fractions containing boric acid<sup>62</sup>. Before the introduction of FP there was no uptake of boric acid by the spent CCU resins. However, after the introduction of FP the spent CCU resins have shown an uptake of boric acid. This is an indication that the anion part of the resins still has its active groups intact in the FP train, operating at lower temperature than earlier<sup>63</sup>.

The total period with sampling from 2009 to April 2013 covers 2.24 EFPY with a total accumulation of 4.5 % of the production. The organic fraction is 2.0 %. The standard deviation in the totally accumulated C-14 activity ranges from 4.8 % for the inorganic fraction to 23 % for the organic fraction. The main factor contributing to this large value is the scatter in the concentrations measured for the organic fraction. There is no obvious explanation for this behavior.

<sup>&</sup>lt;sup>62</sup> The water with boric acid comes from the transports of PWR resins to the R1 WHB.

<sup>&</sup>lt;sup>63</sup> Pers. comm. A. Höglund, Ringhals, May 2013.



### A4.5. Part 4: Resins from 2013-2014

#### A4.5.1. Resin Mass Balance and Calculations

The records describing the additions and removals of resin are found in *Attachment File R1-4*, together with the calculations. The tables and figures for Part 4 are numbered from 41 and upwards. All analyses and calculations were performed in the same way as for parts 1, 2 and 3.



Figure A4-41. The log data for resin storage tank T41 for 2013-14 in graphics. (R1-4, sheet D T41 2014; data are in Sheet T41).

			Sample						SD	
			Weighting	Total,	Inorg,	Org,	Org,	SD	inorg,	SD
Batch	Start date	End date	factor	Bq/kg dw	Bq/kg dw	Bq/kg dw	%	tot, %	%	org, %
	Initial									
R1-12 B18	inventory	2013-06-27	1.00	2.30E+06	2.28E+06	2.78E+04	1.2%	1.5%	1.9%	28%
R1-12 B18	2013-06-27	2013-08-01	1.00	2.30E+06	2.28E+06	2.78E+04	1.2%	1.5%	1.9%	28%
R1-12 B19	2013-08-01	2013-09-12	1.00	2.33E+06	2.28E+06	4.84E+04	2.1%	3.4%	4.2%	33%
R1-12 B20	2013-09-12	2013-09-25	1.00	2.02E+06	1.99E+06	3.10E+04	1.5%	1.1%	1.4%	32%
R1-12 B21	2013-09-25	2013-11-05	1.00	1.80E+06	1.77E+06	2.74E+04	1.5%	1.4%	1.6%	11%
R1-12 B22	2013-11-05	2013-12-03	1.00	1.58E+06	1.55E+06	2.55E+04	1.6%	3.4%	3.2%	17%
R1-12 B23	2013-12-03	2013-12-19	1.00	2.34E+06	2.29E+06	4.08E+04	1.8%	4.1%	4.4%	9.8%
R1-12 B24	2013-12-19	2014-01-13	1.00	2.59E+06	2.52E+06	6.76E+04	2.6%	2.1%	3.0%	48%
R1-12 B25	2014-01-13	2014-01-30	1.00	2.43E+06	2.37E+06	5.61E+04	2.3%	5.3%	4.7%	38%
R1-12 B26	2014-01-30	2014-02-18	1.00	1.96E+06	1.83E+06	1.35E+05	6.9%	7.0%	8.9%	72%
R1-12 B27	2014-02-18	2014-02-21	1.00	2.20E+06	2.06E+06	1.46E+05	6.7%	8.2%	8.8%	1.5%
R1-12 B28	2014-02-21	2014-03-20	1.00	2.21E+06	2.10E+06	1.11E+05	5.0%	4.7%	4.7%	4.8%
	Residual	2014-03-20	1.00	2.21E+06	2.10E+06	1.11E+05	5.0%	4.7%	4.7%	4.8%

Table A4-41. Concentration data used in the calculations for the 2014 samplings. The sample weighting factor was introduced in order to handle dilution effects from non-radioactive resins; however, with the present calculation model it has not been used. (R1-4, sheet (R1-4, sheet R1acc mar2014).

The samplings started in July 2013 and continued to the end of the cycle in March 2014. *Figure A4-41* shows the mass balance for powder resins from 2009 to March 2014. *Figure A4-42* and *Table A4-41* show the concentration values.



Figure A4-42. Concentrations of C-14 used for calculation of the quantities of C-14 in the powder resins. See footnote to Figure A4-4 for comments on the low concentrations. (R1-4, sheet D conc).

*Table A4-42* shows the energy production during the sampling period. *Table A4-43* shows the accumulated amounts of C-14 during the 2013-2014 sampling period. The BFF has been set to 1.09 in accordance with the previous parts. *Table A4-44* and *Figure A4-43* summarizes the accumulation for all the years 2009-2014.

The accumulation in the latest cycle is 1.7 % of the production, with an organic fraction of 7.8 %. Expressing the organic accumulation as  $Bq/MWh_{th}$ , it is only slightly higher than the previous average. The lower accumulation is mainly explained by a new operating mode with full Forward Pumping of the condensate in both turbine trains and a substantially reduced amount of CCU resin used. See *Appendix 22* for a detailed discussion on this topic.

						Calculate		Equiv.
					C-14	d	Equiv. Full	Full
					produc-	productio	Power	Power
		MWhth	MWhth		tion rate,	n Bq	Hours,	Years,
Start	End	start	end	MWhth	<b>Bq/MWHth</b>	total	(EFPH)	(EFPY)
2013-04-30	2014-03-30	5.4E+08	5.6E+08	16550570	2.35E+04	3.90E+11	6366	0.73

*Table A4-42. Summary of energy production and calculated production of C-14 in R1 in 2013-2014. (R1-4, sheet R1acc mar2014).* 



Resin unc	ertainty		10%				
Batch	Start date	End date	kg dw	% of total	Total Bq	Inorg, Bq	Org Bq
	Initial						
R1-12 B18	inventory	2013-06-27	-3910	-121%	-9.01E+09	-8.90E+09	-1.09E+08
R1-12 B18	2013-06-27	2013-08-01	250	8%	5.76E+08	5.69E+08	6.94E+06
R1-12 B19	2013-08-01	2013-09-12	425	13%	9.90E+08	9.69E+08	2.06E+07
R1-12 B20	2013-09-12	2013-09-25	125	4%	2.53E+08	2.49E+08	3.88E+06
R1-12 B21	2013-09-25	2013-11-05	1025	32%	1.84E+09	1.82E+09	2.81E+07
R1-12 B22	2013-11-05	2013-12-03	505	16%	7.97E+08	7.84E+08	1.29E+07
R1-12 B23	2013-12-03	2013-12-19	125	4%	2.92E+08	2.87E+08	5.10E+06
R1-12 B24	2013-12-19	2014-01-13	0	0%	0.00E+00	0.00E+00	0.00E+00
R1-12 B25	2014-01-13	2014-01-30	250	8%	6.07E+08	5.93E+08	1.40E+07
R1-12 B26	2014-01-30	2014-02-18	125	4%	2.46E+08	2.29E+08	1.69E+07
R1-12 B27	2014-02-18	2014-02-21	0	0%	0.00E+00	0.00E+00	0.00E+00
R1-12 B28	2014-02-21	2014-03-20	125	4%	2.76E+08	2.62E+08	1.38E+07
	Residual	2014-03-20	4175	130%	9.21E+09	8.75E+09	4.62E+08
Sum			3220	100%	6.08E+09	5.61E+09	4.75E+08
Resin use			3573				
% of produ	uction				1.6%	1.4%	0.122%
Bq/MWh <sub>th</sub>					3.67E+02	3.39E+02	2.87E+01
% organic							7.8%
Standard	Deviation, %	, o			23%	24%	13%
Sum with	BFF=	1.09			6.64E+09	6.12E+09	5.19E+08
% of produ	uction				1.7%	1.6%	0.13%
Bq/MWh <sub>th</sub>					4.01E+02	3.70E+02	3.14E+01

Table A4-43. Summary of C-14 in CCU resins at R1 accumulated in the periodApril2013 - March 2014. (R1-4, sheet R1acc mar2014).

	Mass balance						
	T41	T41	T41	T41	T41	T42	T41
Unit	R1	R1	R1	R1	R1	R2	R1
							Mean
				2011- Aug	Aug 2012-	Apr 2013-	2009-Mar
	2009	2010	2011	2012	Apr 2013	Mar-2014	2014
Equivalent Full Power Hours	1497	4125	4510	4267	5235	6366	26000
Equivalent Full Power Years	0.17	0.47	0.51	0.49	0.60	0.73	2.97
Body-Feed Factor, Average (BFF)	1.09	1.09	1.09	1.09	1.09	1.09	1.09
Dry Solids Correction Factor in Sa	mple						
Organic fraction	1.3%	2.1%	1.2%	3.7%	1.7%	7.8%	2.6%
Total accumulation in CCU	7.3%	3.9%	4.6%	4.0%	4.6%	1.7%	3.8%
Organic accumulation in CCU	0.095%	0.080%	0.056%	0.15%	0.077%	0.13%	0.10%
kg resin used	1470	4590	5120	4995	4680	3220	24075
Total Bq/MWhth	1.72E+03	9.13E+02	1.08E+03	9.48E+02	1.09E+03	4.01E+02	9.05E+02
Inorganic Bq/MWhth	1.70E+03	8.94E+02	1.06E+03	9.13E+02	1.08E+03	3.70E+02	8.81E+02
Organic Bq/MWhth	2.23E+01	1.89E+01	1.31E+01	3.53E+01	1.82E+01	3.14E+01	2.37E+01
Total Std. Dev.	7.4%	4.9%	9.0%	13%	22%	23%	6.3%
Inorganic Std. Dev.	7.4%	3.8%	8.9%	14%	12%	24%	4.3%
Organic Std. Dev.	21%	23%	50%	40%	67%	13%	15%

Table A4-44. Summary of accumulation of C-14 in CCU resins at R1 (R1-4, sheet Summary).





Figure A4-43. Summary of accumulation of C-14 in CCU resins at R1 (R1-4, sheet D Summary).

# A4.6. Discussion Parts 1-4

From *Table A4-44* and *Figure A4-43* it can be seen that the accumulation over all sampling periods seems to stabilize around 3.8 % of the production with an organic fraction of 2.6 %.

The cycle starting in June 2013 introduced one major change in the condensate system: for both turbines, the CCU system was reconfigured to operation with forward pumping of the hot drains. This means that the CCU operates at a significantly lower temperature and with a flow-rate only some 70 % of the previous one. This will reduce the thermal breakdown of the resin. This breakdown introduces sulfate ions into the reactor, which is considered to contribute to intergranular stress corrosion cracking.

One effect of the FP was observed in the previous cycle, already with FP in one turbine train: Spent CCU resins are used to clean some waste water fractions containing boric acid. Before the introduction of FP there was no uptake of boric acid by the spent CCU resins. However, after the introduction of FP the spent CCU resins have shown an uptake of boric acid. This is an indication that the anion part of the resins still has its active groups intact in the FP train, operating at lower temperature than earlier<sup>64</sup>.

The accumulation of C-14 decreased after the introduction of FP in both turbine trains. This effect seems mainly to be related to the amount of CCU resin used to clean the condensate. This issue is discussed and explained in detail in *Appendix 22*.

<sup>&</sup>lt;sup>64</sup> Pers. comm. A. Höglund, Ringhals, May 2013.



The total period with sampling from 2009 to March 2014 covers 2.97 EFPY with a total accumulation of 3.8 % of the production. The organic fraction is 2.6 %. The standard deviation in the totally accumulated C-14 activity ranges from 4.3 % for the inorganic fraction to 15 % for the organic fraction. The main factor contributing to the largest value is the scatter in the concentrations measured for the organic fraction. There is no obvious explanation for this behavior, but the scatter has been lower in the last cycle compared to the previous ones (see *Figure A4-42*).

# A4.7. Part 5: Resins from 2014-2015

#### A4.7.1. Resin Mass Balance and Calculations

The records describing the additions and removals of resin are found in *Attachment File R1-4*, together with the calculations. The tables and figures for Part 5 are numbered from 51 upwards. All analyses and calculations were performed in the same way as for earlier parts.



*Figure A4-51. The log data for resin storage tank T41 for 2013-14 in graphics. (R1-4, sheet D T41 2015; data are in Sheet T41).* 



R	G	
U	J	ł
N	IS	
	R	NS NS

Batch	Start date	End date	Sample Weigh- ting factor	Total, Bq/kg dw	Inorg, Bq/kg dw	Org, Bq/kg dw	Org, %	SD tot, %	SD inorg, %	SD org, %
R1-2014 Batch 0	2014-07-02	2014-07-02	1.00	2.16E+06	2.00E+06	1.56E+05	7.3%	3.4%	3.7%	16%
R1-2014 Batch 1	2014-07-02	2014-09-03	1.00	2.18E+06	2.12E+06	6.51E+04	3.0%	1.1%	6.5%	80%
R1-2014 Batch 2	2014-09-03	2014-10-16	1.00	2.19E+06	2.07E+06	1.26E+05	5.7%	3.8%	6.2%	93%
R1-2014 Batch 3	2014-10-16	2014-12-05	1.00	2.28E+06	2.22E+06	6.23E+04	2.7%	1.6%	2.1%	24%
R1-2014 Batch 4	2014-12-05	2015-01-15	1.00	2.29E+06	2.25E+06	4.10E+04	1.8%	1.6%	1.7%	14%
R1-2014 Batch 5	2015-01-15	2015-02-16	1.00	2.31E+06	2.23E+06	8.72E+04	3.7%	8.6%	7.5%	48%
R1-2014 Batch 6	2015-02-16	2015-03-24	1.00	2.37E+06	2.15E+06	2.19E+05	9.3%	5.7%	8.9%	51%
R1-2014 Batch 7	2015-03-24	2015-04-28	1.00	2.60E+06	2.46E+06	1.42E+05	5.4%	5.2%	5.0%	48%
	Residual	2015-04-28	1.00	2.60E+06	2.46E+06	1.42E+05	5.4%	5.2%	5.0%	48%

Table A4-51. Concentration data used in the calculations for the 2015 samplings.(R1-4, sheet R1acc apr2015).

The samplings started in April 2014 and continued to the end of the cycle in March 2014. *Figure A4-51* shows the mass balance for powder resins from 2009 to March 2015. *Figure A4-52* and *Table A4-51* show the concentration values.



Figure A4-52. Concentrations of C-14 used for calculation of the quantities of C-14 in the powder resins. See footnote to Figure A4-4 for comments on the low concentrations. (R1-4, sheet D conc).

Start	End	MWhth start	MWhth end	MWhth	C-14 produc- tion rate, Bq/MWHth	Calculated production Bq total	Equiv. Full Power Hours, (EFPH)	Equiv. Full Power Years, (EFPY)
2014-03-30	2015-03-30	5.57E+08	5.74E+08	1.63E+07	2.35E+04	3.84E+11	6281	0.72

*Table A4-52. Summary of energy production and calculated production of C-14 in R1 in 2014-2015. (R1-4, sheet R1acc apr2015).* 



*Table A4-52* shows the energy production during the sampling period. *Table A4-53* shows the accumulated amounts of C-14 during the 2014-2015 sampling period. The BFF has been set to 1.09 in accordance with the previous parts. *Table A4-54* and *Figure A4-53* summarize the accumulation for all the years 2009-2015.

Resin uncertain	ty		10%				
Batch	Start date	End date	kg dw	% of total	Total Bq	Inorg, Bq	Org Bq
	Initial						
R1-2014 Batch 0	inventory	2014-07-02	-4210	-97%	-9.08E+09	-8.43E+09	-6.57E+08
R1-2014 Batch 1	2014-07-02	2014-09-03	275	6%	6.00E+08	5.82E+08	1.79E+07
R1-2014 Batch 2	2014-09-03	2014-10-16	270	6%	5.92E+08	5.58E+08	3.41E+07
R1-2014 Batch 3	2014-10-16	2014-12-05	380	9%	8.68E+08	8.44E+08	2.37E+07
R1-2014 Batch 4	2014-12-05	2015-01-15	295	7%	6.76E+08	6.64E+08	1.21E+07
R1-2014 Batch 5	2015-01-15	2015-02-16	300	7%	6.94E+08	6.68E+08	2.62E+07
R1-2014 Batch 6	2015-02-16	2015-03-24	425	10%	1.01E+09	9.12E+08	9.32E+07
R1-2014 Batch 7	2015-03-24	2015-04-28	300	7%	7.81E+08	7.39E+08	4.25E+07
	Residual	2015-06-10	6285	145%	1.64E+10	1.55E+10	8.89E+08
Sum			4320	100%	1.25E+10	1.20E+10	4.82E+08
% of production					3.3%	3.1%	0.125%
Bq/MWh <sub>th</sub>					7.65E+02	7.36E+02	2.95E+01
% organic							3.9%
Standard Devia	tion, %				17%	16%	94%
Sum with BFF=		1.09			1.36E+10	1.31E+10	5.26E+08
% of production					3.5%	3.4%	0.14%
Bq/MWh <sub>th</sub>					8.35E+02	8.03E+02	3.22E+01

Table A4-53. Summary of C-14 in CCU resins at R1 accumulated in the period July 2014 - April 2014. (R1-4, sheet R1acc apr2015).

								Mean
				2011- Aug	Aug 2012-	Apr 2013-	Jul 2014-	2009-Apr
	2009	2010	2011	2012	Apr 2013	Mar-2014	Apr 2015	2015
Equivalent Full Power Hours	1497	4125	4510	4267	5235	6366	6281	32280
Equivalent Full Power Years	0.17	0.47	0.51	0.49	0.60	0.73	0.72	3.68
Body-Feed Factor, Average (BFF)	1.09	1.09	1.09	1.09	1.09	1.09	1.09	1.09
Dry Solids Correction Factor in Sa	mple							
Organic fraction	1.3%	2.1%	1.2%	3.7%	1.7%	7.8%	3.9%	2.8%
Total accumulation in CCU	7.3%	3.9%	4.6%	4.0%	4.6%	1.7%	3.5%	3.8%
Organic accumulation in CCU	0.095%	0.080%	0.056%	0.15%	0.077%	0.13%	0.14%	0.11%
kg resin used	1530	4680	4230	5447	4746	3573	4485	28691
Total Bq/MWhth	1.72E+03	9.13E+02	1.08E+03	9.48E+02	1.09E+03	4.01E+02	8.35E+02	8.64E+02
Inorganic Bq/MWhth	1.70E+03	8.94E+02	1.06E+03	9.13E+02	1.08E+03	3.70E+02	8.03E+02	8.40E+02
Organic Bq/MWhth	2.23E+01	1.89E+01	1.31E+01	3.53E+01	1.82E+01	3.14E+01	3.22E+01	2.43E+01
Total Std. Dev.	7.4%	4.9%	9.0%	13%	22%	23%	17%	6.5%
Inorganic Std. Dev.	7.4%	3.8%	8.9%	14%	12%	24%	16%	5.2%
Organic Std. Dev.	21%	23%	50%	40%	67%	13%	94%	27%

Table A4-54. Summary of accumulation of C-14 in CCU resins at R1 (R1-4, sheet Summary).

The accumulation in the latest cycle is 3.5 % of the production, with an organic fraction of 3.9 %. Expressing the accumulation as Bq/MWh<sub>th</sub>, it is lower than the previous average, but higher than for the previous cycle. The higher accumulation compared with the previous cycle is mainly explained by a higher use of CCU resins and a higher x-value. See *Appendix 22* for a detailed discussion on this topic. The mean total accumulation is 3.8 % of the production with an organic fraction of 2.8 %.





Figure A4-53. Summary of accumulation of C-14 in CCU resins at R1 (R1-4, sheet D Summary).







# APPENDIX 5 Results from R2, R3 and R4

### **A5.1. Introduction**

All calculations for R2, R3 and R4 are included in an excel-book, which can be found *as Attachment File R234-1* to this report. Reference to the various sheets will be made at tables, figures etc.



Figure A5-1. Overview of handling of spent resins at R2, R3 and R4. (A-5, pict. 1).

The operational strategies for the RWCU in the Ringhals PWR units are virtually the same in all three units. At the beginning of the fuel cycle, mixed-bed resin is loaded and used during the whole cycle, including the initial part of the shut-down phase preceding the annual refueling.

# A5.2. Resin Handling and Sampling

The spent resin is back-flushed to a storage tank (SRST), from which it is transported to the R1 Waste Handling Building (WHB). For the sampling campaigns covered by this report, the SRST has been emptied before the RWCU resin has been back-flushed in order to obtain RWCU resins only.

The transport is performed by means of a lead shielded spherical transport vessel. Some 3-7 separate transports are needed to transfer a RWCU resin load. The handling system has no representative sampling system, so the samples are withdrawn by means of a 20 ml plastic bottle. 1 -2 subsamples are taken from each transport vessel load and batched on completion of the transport campaign<sup>65</sup>.

<sup>&</sup>lt;sup>65</sup> Since anion resin has a lower density than cation resin, there is a probability that the samples may contain a higher fraction of anion resin than the mixed bed. This may lead to an over estimation of C-14, since it is expected to be bound to the anion resin.

The radiation dose amounts to about 0.2-0.3 mmanSv for a campaign to sample the RWCU resin from one PWR unit<sup>66</sup>.

Totally 18 mixed bed resins have been sampled. See *Table A5-4*, where the units and the years are given. The resins have been in operation for one fuel cycle, but not during the refueling shut-down. There is one exception: The resin in R2 sampled in 2003 had been in operation for two cycles.

# A5.3. Calculation Model and Results

The calculations have been done according to the Batch Model (BM), where the concentrations of the resin samples have been multiplied by the resin dry weight (dw).

Analysis dat	ta											
Batch	Start date	End date	BFF	Total, Bq/kg dw	Inorg, Bq/kg dw	Org, Bq/kg dw	Org, %	SD tot, %	SD inorg, %	SD org, %		
R2-334-03	2001-05-31	2003-05-31	1.15	2.19E+07	1.60E+07	6.27E+06	27%	23%	28%	14%		
R2-334-08	2007-08-01	2008-05-17	1.15	6.23E+07	5.27E+07	9.68E+06	16%	3.3%	3.3%	3.6%		
R2-334-11	2010-01-01	2011-04-30	1.15	2.95E+07	1.88E+07	1.07E+07	36%	5.4%	6.4%	4.0%		
R2-334-12	2012-04-01	2012-09-15	1.15	1.81E+07	5.04E+06	1.31E+07	72%	10%	9.9%	11%		
R2-334-13	2012-11-01	2013-09-07	1.15	2.19E+07	1.48E+07	7.06E+06	32%	5.1%	5.8%	4.0%		
R2-334-14	2013-10-10	2014-08-15	1.15	2.19E+07	1.48E+07	7.06E+06	32%	5.1%	5.8%	4.0%		
Calculated	amounts of (	C-14										
Batch	Start date	End date	kg dw	Total Bq	Inorg, Bq	Org Bq		SD tot	SD inorg	SD org		
R2-334-03	2001-05-31	2003-05-31	986	2.49E+10	1.81E+10	7.11E+09		9.72E+09	7.66E+09	2.46E+09		
R2-334-08	2007-08-01	2008-05-17	181	1.30E+10	1.10E+10	2.02E+09		4.13E+09	3.49E+09	6.41E+08		
R2-334-11	2010-01-01	2011-04-30	173	5.88E+09	3.75E+09	2.14E+09		1.89E+09	1.21E+09	6.81E+08		
R2-334-12	2012-04-01	2012-09-15	181	3.77E+09	1.05E+09	2.73E+09		1.25E+09	3.48E+08	9.11E+08		
R2-334-13	2012-11-01	2013-09-07	181	4.56E+09	3.09E+09	1.47E+09		1.46E+09	9.93E+08	4.69E+08		
R2-334-14	2013-10-10	2014-08-15	181	4.56E+09	3.09E+09	1.47E+09		1.46E+09	9.93E+08	4.69E+08		
RESIN DAT	Α							1.1E+10	8.63E+09	2.86E+09		
Resin + san	nple scatter u	uncertainty	32%		Resin unce	rtainty	10%		Sample s	catter unce	ertainty	30%
				Dry sub-	Density,							
Batch	Start date	End date	Liters	stance	kg/lit	kg ww	kg dw	-				
R2-334-08	2001-05-31	2003-05-31	3130	0.45	0.7	2191	986	Several res	sin beds: C	S mixed b	ed, WP a	nd SFP
R2-334-11	2007-08-01	2008-05-17	575	0.45	0.7	403	181					
R2-334-11	2010-01-01	2011-04-30	550	0.45	0.7	385	1/3					
R2-334-12	2012-04-01	2012-09-15	575	0.45	0.7	403	181					
R2-334-13	2012-11-01	2013-09-07	575	0.45	0.7	403	181					
R2-334-14	2013-10-10	2014-08-15	575	0.45	0.7	403	181					
PRODUCTIO	JN DATA											
	<b>a</b>		MWhth	MWhth		C-14 produc- tion rate,	Calc. Produc- tion, Bq					
Batch	Start	End	Start	ena	Niwhth	Bq/Wwhth	total	EFPH	EFPY			
R2-334-03	2001-05-31	2003-05-31	3.85E+08	4.26E+08	40600707	1.49E+04	0.00E+11	15309	1.75			
R2-334-06	2007-08-01	2006-05-17	5.11E+00	5.20E+00	14693635	1.49E+04	2.22E+11	0100	0.04			
R2-334-11	2010-01-01	2011-04-30	5.43E+00	5.00E+00	6220072	1.49E+04	0.20E+10	0032	0.99			
R2-334-12	2012-04-01	2012-09-15	5.09E+00	5.70E+00	10229072	1.49E+04	9.30E+10	2349	0.27			
R2-334-13	2012-11-01	2013-09-07	5.70E+00	5.94E+00	18426182	1.49E+04	2.74E+11	6048	0.79			
112-334-14	2013-10-10	2014-00-13	J.94L+00	Inora %	Organic %	1.492+04	2.756+11	0940	0.79			
			of pro-	of pro-	of pro-	Total	Inorganic	Organic	0/_	SD tot	SD	SD org
SUMMARY			duction	duction	duction	Ba/MWhth	Ba/MWhth	Ba/MWhth	organic	%	inora %	%
R2-334-03	2001-05-31	2003-05-31	4 1%	3.0%	1 17%	6 12F+02	4 47F+02	1 75F+02	27%	39%	42%	35%
R2-334-08	2007-08-01	2008-05-17	5.8%	4 9%	0.91%	8 72F±02	7 37F±02	1.35F±02	16%	32%	32%	32%
R2-334-11	2010-01-01	2011-04-30	1.7%	1.5%	0.62%	2.57F+02	1.64F+02	9.33F+01	36%	32%	32%	32%
R2-334-12			4.40/	4.40/	0.02/0	C. 005-02	1.605.02	4 275 02	720/	33%	320/	33%
	2012-04-01	2012-09-15	41%	11%	19%	0.00E+U/	1.09E+U/	4.3/ E+U/	12/0	//)	.).)/0	//)
IR2-334-13	2012-04-01 2012-11-01	2012-09-15	4.1%	1.1%	2.9%	6.06E+02 2.49E+02	1.69E+02	4.37E+02 8.02E+01	32%	32%	32%	32%

Table A5-1. Summary of C-14 accumulation in RWCU for R2. (R234-1, sheet R2).

<sup>&</sup>lt;sup>66</sup> Anders Höglund, Ringhals, R1M, Pers. Comm. Sept. 2011.



#### A5.3.1. Results from R2

*Table A5-1* summarizes the results for R2 and samples collected in 2003, 2008 and 2011-2014.

#### A5.3.2. Results from R3

*Table A5-2* summarizes the results for R3 and samples collected in 2003, 2008-2009 and 2011-2014.

Analysis da	ta											
Batch	Start date	End date	BFF	Total, Bq/kg dw	Inorg, Bq/kg dw	Org, Bq/kg dw	Org, %	SD tot, %	SD inorg, %	SD org, %		
R3-334-03	2002-05-31	2003-05-31	1.15	5.25E+07	3.78E+07	1.52E+07	28%	16%	18%	13%		
R3-334-08	2007-05-30	2008-07-25	1.15	4.31E+07	2.92E+07	1.39E+07	32%	5.5%	5.7%	5.5%		
R3-334-09	2008-08-05	2009-09-01	1.15	3.79E+07	2.81E+07	9.75E+06	26%	5.3%	4.8%	5.3%		
R3-334-11	2010-09-28	2011-11-05	1.15	2.34E+07	1.82E+07	5.14E+06	22%	6.4%	8.0%	2.0%		
R3-334-12	2011-12-06	2012-06-09	1.15	3.09E+07	2.22E+07	9.20E+06	28%	3.6%	2.8%	3.1%		
R3-334-13	2012-07-05	2013-08-10	1.15	4.10E+07	2.78E+07	1.32E+07	32%	2.0%	2.7%	3.5%		
R3-334-14	2013-08-10	2014-06-01	1.15	2.23E+07	1.41E+07	8.26E+06	37%	5.0%	6.8%	3.4%		
Calculated	amounts of 0	C-14										
Batch	Start date	End date	kg dw	Total Bq	Inorg, Bq/kg dw	Org Bq		SD tot	SD inorg	SD org		
R3-334-03	2002-05-31	2003-05-31	268	1.62E+10	1.16E+10	4.67E+09		5.73E+09	4.24E+09	1.60E+09		
R3-334-08	2007-05-30	2008-07-25	205	1.02E+10	6.88E+09	3.27E+09		3.26E+09	2.21E+09	1.05E+09		
R3-334-09	2008-08-05	2009-09-01	205	8.92E+09	6.62E+09	2.30E+09		2.86E+09	2.12E+09	7.36E+08		
R3-334-11	2010-09-28	2011-11-05	205	5.50E+09	4.29E+09	1.21E+09		1.77E+09	1.40E+09	3.83E+08		
R3-334-12	2011-12-06	2012-06-09	205	7.28E+09	5.24E+09	2.17E+09		2.32E+09	1.66E+09	6.88E+08		
R3-334-13	2012-07-05	2013-08-10	205	9.66E+09	6.56E+09	3.10E+09		3.06E+09	2.08E+09	9.88E+08		
R3-334-14	2013-08-10	2014-06-01	205	5.26E+09	3.31E+09	1.95E+09		1.68E+09	1.07E+09	6.19E+08		
RESIN DAT	A							8.508E+09	6.13E+09	2.48E+09		
Resin + sar	nple scatter u	uncertainty	32%		Resin unce	rtainty	10%		Sample se	catter unce	ertainty	30%
				Dry sub-	Density,							
Batch	Start date	End date	Liters	stance	kg/lit	kg ww	kg dw					
R3-334-03	2002-05-31	2003-05-31	850	0.45	0.7	595	268	Åsa used 8	850 litres; R	4 value		
R3-334-08	2007-05-30	2008-07-25	650	0.45	0.7	455	205					
R3-334-09	2008-08-05	2009-09-01	650	0.45	0.7	455	205					
R3-334-11	2010-09-28	2011-11-05	650	0.45	0.7	455	205					
R3-334-12	2011-12-06	2012-06-09	650	0.45	0.7	455	205					
R3-334-13	2012-07-05	2013-08-10	650	0.45	0.7	455	205					
R3-334-14	2013-08-10	2014-06-01	650	0.45	0.7	455	205					
PRODUCTI	ON DATA											
			MWhth	MWhth		C-14 produc- tion rate,	Calc. Produc- tion, Bq					
Batch	Start	End	start	end	MWhth	Bq/MWHth	total	EFPH	EFPY			
R3-334-03	2002-05-31	2003-05-31	3.66E+08	3.85E+08	18857047	1.59E+04	2.99E+11	5941	0.68			
R3-334-08	2007-05-30	2008-07-25	4.72E+08	4.94E+08	22418288	1.59E+04	3.56E+11	7063	0.81			
R3-334-09	2008-08-05	2009-09-01	4.94E+08	5.20E+08	25987566	1.59E+04	4.13E+11	8188	0.93			
R3-334-11	2010-09-28	2011-11-05	5.46E+08	5.71E+08	25083584	1.59E+04	3.98E+11	7903	0.90			
R3-334-12	2011-12-06	2012-06-09	5.73E+08	5.85E+08	11996618	1.59E+04	1.90E+11	3780	0.43			
R3-334-13	2012-07-05	2013-08-10	5.85E+08	6.14E+08	29033559	1.59E+04	4.61E+11	9147	1.04			
R3-334-14	2013-08-10	2014-06-01	6.14E+08	6.29E+08	14776635	1.59E+04	2.35E+11	4656	0.53			
			Total %	Inorg %	Organic %							
			of pro-	of pro-	of pro-	Total	Inorganic	Organic			SD inorg,	SD org,
SUMMARY			duction	duction	duction	Bq/MWhth	Bq/MWhth	Bq/MWhth	% organic	SD tot, %	%	%
R3-334-03	2002-05-31	2003-05-31	5.4%	3.9%	1.6%	8.58E+02	6.18E+02	2.47E+02	28%	35%	36%	34%
R3-334-08	2007-05-30	2008-07-25	2.9%	1.9%	0.92%	4.53E+02	3.07E+02	1.46E+02	32%	32%	32%	32%
R3-334-09	2008-08-05	2009-09-01	2.2%	1.6%	0.56%	3.43E+02	2.55E+02	8.83E+01	26%	32%	32%	32%
R3-334-11	2010-09-28	2011-11-05	1.4%	1.1%	0.30%	2.19E+02	1.71E+02	4.82E+01	22%	32%	33%	32%
R3-334-12	2011-12-06	2012-06-09	3.8%	2.7%	1.1%	6.07E+02	4.37E+02	1.81E+02	28%	32%	32%	32%
R3-334-13	2012-07-05	2013-08-10	2.1%	1.4%	0.67%	3.33E+02	2.26E+02	1.07E+02	32%	32%	32%	32%
R3-334-14	2013-08-10	2014-06-01	2.2%	1.4%	0.83%	3.56E+02	2.24E+02	1.32E+02	37%	32%	32%	32%
				1	1	1			1	1		

Table A5-2. Summary of C-14 accumulation in RWCU for R3. (R234-1, sheet R3).



#### A5.3.3. Results from R4

Table A5-3 summarizes the results for R4 and samples collected in 2009 and 2011-2014.

Analysis da	ita											
Batch	Start date	End date	BFF	Total, Bq/kg dw	Inorg, Bq/kg dw	Org, Bq/kg dw	Org, %	SD tot, %	SD inorg, %	SD org, %		
R4-334-09	2008-06-21	2009-05-09	1.15	6.35E+07	4.36E+07	1.98E+07	31%	2.9%	2.9%	2.9%		
R4-334-11	2010-08-01	2011-05-30	1.15	8.16E+07	6.04E+07	2.12E+07	26%	4.3%	4.7%	4.7%		
R4-334-12	2011-11-15	2012-09-01	1.15	2.29E+07	1.61E+07	6.74E+06	29%	3.7%	4.4%	5.9%		
R4-334-13	2012-10-01	2013-05-20	1.15	1.90E+07	1.29E+07	6.19E+06	32%	0.65%	0.87%	0.87%		
R4-334-14	2013-06-15	2014-08-06	1.15	1.90E+07	1.29E+07	6.19E+06	33%	2.7%	4.2%	3.5%		
Calculated	amounts of 0	C-14										
					Inorg,							
Batch	Start date	End date	kg dw	Total Bq	Bq/kg dw	Org Bq		SD tot	SD inorg	SD org		
R4-334-09	2008-06-21	2009-05-09	268	1.95E+10	1.34E+10	6.10E+09		6.20E+09	4.27E+09	1.94E+09		
R4-334-11	2010-08-01	2011-05-30	268	2.51E+10	1.86E+10	6.53E+09		8.02E+09	5.94E+09	2.09E+09		
R4-334-12	2011-11-15	2012-09-01	268	7.04E+09	4.97E+09	2.08E+09		2.24E+09	1.59E+09	6.68E+08		
R4-334-13	2012-10-01	2013-05-20	268	5.86E+09	3.96E+09	1.90E+09		1.85E+09	1.25E+09	6.02E+08		
R4-334-14	2013-06-15	2014-08-06	268	5.86E+09	3.96E+09	1.90E+09		1.86E+09	1.26E+09	6.06E+08		
RESIN DAT	Ά							1.07E+10	7.7E+09	3.05E+09		
Resin + sar	mple scatter u	uncertainty	32%		Resin unce	rtainty	10%		Sample s	catter unce	rtainty	30%
Batch	Start date	End date	Liters	Dry sub- stance	Density, kq/lit	kg ww	kg dw					
R4-334-09	2008-06-21	2009-05-09	850	0.45	0.7	595	268					
R4-334-11	2010-08-01	2011-05-30	850	0.45	0.7	595	268					
R4-334-12	2011-11-15	2012-09-01	850	0.45	0.7	595	268					
R4-334-13	2012-10-01	2013-05-20	850	0.45	0.7	595	268					
R4-334-14	2013-06-15	2014-08-06	850	0.45	0.7	595	268					
PRODUCTI	ON DATA											
Detah	04-14	<b>F</b> _ 1	MWhth	MWhth	<b>N</b> /14/1-4/-	C-14 produc- tion rate,	Calc. Produc- tion, Bq	FEDI	FERV			
Batch	Start	End	start		10097677			<b>EFPH</b>				
R4-334-09	2008-06-21	2009-05-09	4.02E+00	5.02E+00	1998/6//	1.43E+04	2.00E+11	7203	0.82			
R4-334-11	2010-06-01	2011-05-30	5.2/E+00	5.40E+00	20030120	1.43E+04	2.94E+11	7430	0.85			
R4-334-12	2011-11-15	2012-09-01	5.40E+00	5.00E+00	17072561	1.43E+04	2.44E+11	6152	0.70			
R4-334-13	2012-10-01	2013-05-20	5.65E+08	5.80E+08	15063353	1.43E+04	2.15E+11	5428	0.62			
R4-334-14	2013-06-15	2014-08-06	5.80E+08	6.08E+08	2/508162	1.43E+04	3.93E+11	9913	1.13			
			Total %	inorg %	Organic %	Tetal		Ormania				00.000
CUMMARY			or pro-	or pro-	or pro-		morganic		0/	CD tot N	ວມ inorg,	SD org,
SUMMARY	2008.06.04	2000 05 00	auction	auction	auction	Bq/WWnth	Bq/WWhth	Bq/WWnth	% organic	ວບ tot, %	<b>%</b>	<b>%</b>
R4-334-09	2008-06-21	2009-05-09	0.9%	4.1%	2.1%	9.78E+02	0.72E+02	3.05E+02	31%	3∠%	32%	32%
R4-334-11	2010-08-01	2011-05-30	8.5%	6.3%	2.2%	1.22E+03	9.01E+02	3.16E+02	26%	32%	32%	32%
R4-334-12	2011-11-15	2012-09-01	2.9%	2.0%	0.85%	4.13E+02	2.91E+02	1.22E+02	29%	32%	32%	32%
R4-334-13	2012-10-01	2013-05-20	2.7%	1.8%	0.89%	3.89E+02	2.63E+02	1.26E+02	32%	32%	32%	32%
K4-334-14	2013-06-15	2014-08-06	1.5%	1.0%	0.49%	2.13E+02	1.44E+02	6.92E+01	33%	32%	32%	32%

Table A5-3. Summary of C-14 accumulation in RWCU for R4. (R234-1, sheet R4).

*Table A5-4* and *Figure A5-2* summarize the results for RWCU from the present report. The results from *Ref. 4* have been corrected for wrong resin gross density used in the previous calculations; see *Appendix 6* and 7.

#### A5.4. Discussion of Reasons for Variations in Results

The results summarized in *Table A5-4* and *Figure A5-2* vary over time and between the units. The correction of the old results from 2003 brings them down to a level comparable with those found in this study. However, the average value for R4 is higher than for R2 and R3. There are also variations between R2 and R3.

The average accumulation over 19 fuel cycles and 14.7 EFPY is 3.3 % of the production with an organic fraction of 30 % (1.0 % of the production). The standard deviation in the summed



Samp- ling year			EFPY	Accum. total	Accum. Inorg.	Accum. Org.	Org. frac- tion	Total Bq/ MWhth	Inorganic Bq/ MWhth	Organic Bq/ MWhth	Std. Dev. Total	Std. Dev. Inorg.	Std. Dev. Org.
2003	R2	2001-2003	1.75	4.1%	3.0%	1.2%	27%	6.12E+02	4.47E+02	1.75E+02	39%	42%	35%
2008	R2	2007-2008	0.64	5.8%	4.9%	0.91%	16%	8.72E+02	7.37E+02	1.35E+02	32%	32%	32%
2011	R2	2010-2011	0.99	1.7%	1.1%	0.62%	36%	2.57E+02	1.64E+02	9.33E+01	32%	32%	32%
2012	R2	2012-2012	0.27	4.1%	1.1%	2.9%	72%	6.06E+02	1.69E+02	4.37E+02	33%	33%	33%
2013	R2	2012-2013	0.79	1.7%	1.1%	0.54%	32%	2.49E+02	1.68E+02	8.02E+01	32%	32%	32%
2014	R2	2013-2014	0.79	1.7%	1.1%	0.53%	32%	2.47E+02	1.68E+02	7.98E+01	32%	32%	32%
2003	R3	2002-2003	0.68	5.4%	3.9%	1.6%	28%	8.58E+02	6.18E+02	2.47E+02	35%	36%	34%
2008	R3	2007-2008	0.81	2.9%	1.9%	0.92%	32%	4.53E+02	3.07E+02	1.46E+02	32%	32%	32%
2009	R3	2008-2009	0.93	2.2%	1.6%	0.56%	26%	3.43E+02	2.55E+02	8.83E+01	32%	32%	32%
2011	R3	2010-2011	0.90	1.4%	1.1%	0.30%	22%	2.19E+02	1.71E+02	4.82E+01	32%	33%	32%
2012	R3	2011-2012	0.43	3.8%	2.7%	1.1%	28%	6.07E+02	4.37E+02	1.81E+02	32%	32%	32%
2013	R3	2012-2013	1.04	2.1%	1.4%	0.67%	32%	3.33E+02	2.26E+02	1.07E+02	32%	32%	32%
2014	R3	2013-2014	0.53	2.2%	1.4%	0.83%	37%	3.56E+02	2.24E+02	1.32E+02	32%	32%	32%
2009	R4	2008-2009	0.82	6.9%	4.7%	2.1%	31%	9.78E+02	6.72E+02	3.05E+02	32%	32%	32%
2011	R4	2010-2011	0.85	8.5%	6.3%	2.2%	26%	1.22E+03	9.01E+02	3.16E+02	32%	32%	32%
2012	R4	2011-2012	0.70	2.9%	2.0%	0.85%	29%	4.13E+02	2.91E+02	1.22E+02	32%	32%	32%
2013	R4	2012-2013	0.62	2.7%	1.8%	0.89%	32%	3.89E+02	2.63E+02	1.26E+02	32%	32%	32%
2014	R4	2013-2014	1.13	1.5%	1.0%	0.49%	33%	2.13E+02	1.44E+02	6.92E+01	32%	32%	32%
	R2-R4	Mean	14.68	3.3%	2.3%	1.0%	30%	4.95E+02	3.50E+02	1.46E+02	8.4%	8.9%	7.7%
	R2	Mean	5.22	3.1%	2.2%	0.9%	30%	4.66E+02	3.30E+02	1.39E+02	19%	22%	17%
	R3	Mean	5.33	2.7%	1.9%	0.79%	30%	4.25E+02	3.01E+02	1.26E+02	14%	14%	13%
	R4	Mean	4.12	4.4%	3.1%	1.3%	29%	6.33E+02	4.48E+02	1.85E+02	17%	17%	16%
	Numbe	er of cycles	19										

Table A5-4. Summary of accumulation of C-14 in PWR RWCU resins, including corrected results from Ref. 4 (years 2001-2003). The sampling in 2003 for R2 was for resins used for two cycles. (R234-1, sheet New Summary).



Figure A5-2. Summary of C-14 accumulation in RWCU for all PWR units, including corrected results from Ref. 4. (R234-1, sheet D New summary).



activity is 8.4 %, including a 10 % uncertainty in the resin mass and a 30 % uncertainty in the sampling.

The maximum accumulation was 8.5 % in R4 in 2011, with an organic fraction of 26 %. The minimum value was 1.4 % in R3 in 2011 with an organic fraction of 22%. The maximum organic fraction was 72 % in R2 after a short cycle in 2012. This cycle also had the maximum organic accumulation with 2.9 % of the production. The reasons for the variations are discussed further in *Appendix 20*.

#### A5.4.1. Sampling Procedure

The sampling procedure may be one factor; the "fishing" with a liquid scintillation vial in a transport vessel without stirring may yield varying proportions between cation and anion resins. The anion resin has a slightly lower density than the cation resin. There is a possibility that the anion resin has been enriched in the top part of the transport vessel prior to the sampling, leading to an overrepresentation of anion resin, which may overestimate the accumulation of  $C-14^{67}$ .

On the other hand: If the majority of the anion resin is in one or a few transport vessels, it may lead to an underrepresentation of the anion resin, leading to un underestimation of the accumulation. To account for these possibilities, the PWR samples have been assigned a sampling uncertainty of 30 %, which dominates the uncertainty in the accumulation for single years. However, summing all the accumulations reduces the uncertainty to some 10 %.

#### A5.4.2. Differences in Primary Chemistry between the Units

Differences in primary chemistry between the units might be another explanation for the differences between R2 and R3 versus R4. R2 and R3 have operated at  $pH_{300} = 7.40$ , while R4 has operated at  $pH_{300} = 7.25$  until 2011. The reason for this was to reduce the primary-to-secondary leakage through cracks in the SG tubing, which has a strong dependence on the pH-value. After the SG replacement in 2011, R4 has operated with the same primary chemistry as R2 and R3. In fact, the results for R4 from 2012 and 2013 are lower than the previous results.

# A5.4.3. Low Concentration of Inorganic C-14 (Carbonate System) in the Reactor Coolant in the First Few Months of a Fuel Cycle.

In *Appendix 20*, Pourbaix diagrams and concentration values in RC from *Ref. 4* clearly indicate that organic species dominate over inorganic in the beginning of the fuel cycle. This may explain the high organic fraction for Ringhals 2 in 2012. See *Appendix 20* for a full discussion on this topic.

#### A5.4.4. (Non)-Saturation of the Resin with Organic C-14.

(Non)-saturation of the resin with organic C-14. In *Appendix 20* there is also a discussion on saturation of the ion exchange resins with organic species early in the fuel cycle.

<sup>&</sup>lt;sup>67</sup> C-14 is assumed to be bound to anion resin.

# A5.4.5. Cold Shut-downs Followed by Extensive Boron Dilution During Restart.

The lowest accumulation values have been obtained for R3 in 2011. One possible explanation for this may be a cold shutdown for five weeks late in the fuel cycle. The strong dilution required at the restart may have removed some of the C-14 from the RWCU resin. Cf. the left part of the curves in *Figure A5-3* and the discussion below.

*Figure A5-3* is from (*Ref. 4*) and shows the concentrations of C-14 species in the RCS of Ringhals 4 in 2006. During the last part of the fuel cycle, (May-end of July) the concentration in the RWCU outlet is higher than the concentration in the RC. This means that organic C-14 compounds are removed from the ion exchanger. During this period, there is a strong dilution of the RC to reduce the concentration of boric acid. The dilution also removes C-14 compounds from the RC, lowering the concentration of C-14 compounds. It seems reasonable, that the total accumulation of organic C-14 compounds is proportional to the concentration in the RC.



Figure A5-3. Ringhals 4, 2006. Concentration of non-volatile organic C-14 in the Reactor Coolant (RC) and the RWCU outlet (CS). For the major part of the cycle, the concentration in RC (=RWCU inlet) and CS (RWCU outlet) is almost the same, indicating a saturation of the resin with respect to this type of C-14 compounds (From Ref. 4).

#### A5.4.7. High pH in the RWCU System at the End of a Fuel Cycle.

Another possible factor for the high uptake of C-14 in some cycles is operation with very low boron concentration at the end of the fuel cycle (coast-down with gradually decreasing reactor power due to decreasing reactivity in the fuel).

In a PWR the boron concentration in the Reactor Coolant (RC) is decreasing with the burn-up of the fuel. To maintain a constant pH-value at 300 °C, the concentration of lithium is regulated. The Reactor Water Clean-Up system (RWCU or CS) operates at 45 °C. The pH-values in these two systems are different due to the different temperatures; 300°C and 45 °C, respectively.

*Figures A5-4* and *A5-5* show the variation of the pH-values and the lithium concentration as a function of boron concentration. The pH value is regulated with the boundary condition that pH is at the target value  $(7.25 \text{ in these diagram})^{68}$  or increasing up to the target value (in the beginning of the fuel cycle with high boron concentration)<sup>69</sup>.



Figure A5-4. Li-concentration, pH-values in RC (300 °C) and CS (45 °C) as a function of boron concentration. Target pH in RC: 7.25. (R234-2, sheet D Sum).



Figure A5-5. Li-concentration, pH-values in RC (300 °C) and CS (45 °C) as a function of boron concentration. Target pH in RC: 7.25. Zoom in at low boron concentrations in Figure A5-4. (R234-2, sheet D Sum zoom).

 $<sup>^{68}</sup>$  From 2011, the target value is 7.40 at 300 °C.

<sup>&</sup>lt;sup>69</sup> There are also maximum limits for Li; hence, in the beginning of the fuel cycle with high boron concentration, the pH-value at 300 °C will fall below the target value, (7.25) in these diagrams.



*Figure A5-5* is a zoom-in on the last period of the fuel cycle with low boron concentrations. During coast-down operation, the boron concentration is usually < 5 ppm; accordingly, the pH-value in the RWCU is in the range 8.5-9.2. The operational time at these conditions may last from one week to a couple of months. The high pH-value may allow a more extensive accumulation of hydrogen carbonate ( $pK_{a1}$ =6.37); see *Appendix 8* for further details.

# A5.4.8. Exposure of the RWCU Resin to Acidic and Oxidizing conditions at Shut-down

The RWCU is nowadays (2015)<sup>70</sup> taken out of operation prior to refueling shut-down before the RC chemistry is made acidic and oxidizing<sup>71</sup>; thus, hydrogen carbonate and organic acids may remain on the resin. The diagrams are calculated for an operational pH-value of 7.25; for operation at 7.40 the pH-value in the RWCU will be slightly higher.

#### A5.4.9. Possible Formation of Inorganic C-14 from Organic Acids Accumulated on the Ion Exchanger

The ion exchange resins and the species adsorbed are exposed to very high radiation levels, which may a cause a degradation of the organic species; either through direct radiolysis or via radicals formed by irradiation of water. This might be an explanation why most PWR resin beds have more inorganic than organic C-14 (*Ref. 11*).

# A5.4.10. Amount of Anion Resin Normalized to Equivalent Full Power Hours (EFPH)

 $f = ax_a$ 

In *Appendix 22* a regression of the accumulation of C-14 vs. the amount of anionic resin normalized to the energy production gave a good fit for BWR CCU resins.

The regression for the accumulation factor for PWR RWCU resins may be written as

The accumulation for one fuel cycle is

 $Q = Eax_a \qquad \qquad Eq. (A5-2)$ 

Eq. (A5-1)

Which may be simplified to

fQE

$$Q = an_a P \qquad \qquad Eq. (A5-3)$$

All equations are applicable to organic, inorganic and total accumulation

The symbols are

accumulation factor of C-14, Bq/MWh <sub>th</sub>
accumulation of C-14, Bq
energy production, MWh <sub>th</sub>

<sup>&</sup>lt;sup>70</sup> This operational mode has been used at least from the year 2000, and possibly from the early 1990s. It has not been possible to establish the exact year when it was introduced. However, it means that all PWR resins analyzed for this report have been exposed to this operational mode. Earlier, before an acidic shut-down chemistry was introduced, the RWCU mixed bed was used in the shut-down period as long as it had a satisfactory accumulation of corrosion products and fission products. Then it was replaced by a fresh mixed bed without Li.

<sup>&</sup>lt;sup>71</sup> Bernt Bengtsson & Pernilla Svanberg, Ringhals AB, personal communication, July 2013.









Figure A5-6. Regression of accumulation of organic C-14 vs. the parameter  $x_a$ ; anion equivalents used, normalized to the energy production in EFPH. (R234-1, sheet D regr x org).



Figure A5-7. Regression of accumulation of inorganic C-14 vs. the parameter  $x_a$ ; anion equivalents used, normalized to the energy production in EFPH. (R234-1, sheet D regr x inorg).



The same approach has been applied to RWCU resins. The justification for using Eq. (A5-1) is the following: If you use no resin at all (x=0), there will be no accumulation. Figure A5-6 shows the regression for the organic fraction (some 30 % of the total accumulation). The regression coefficient is 0.85, indicating a good fit. The fit for the inorganic fraction and the total accumulation is lower; see Figures A5-7 and -8, with regression coefficients 0.64 and 0.74, respectively. The regression data are summarized in Table A5-5.



Figure A5-8. Regression of accumulation of total C-14 vs. the parameter x; anion equivalents used, normalized to the energy production in EFPH. (R234-1, sheet D regr x tot).

The diagram for the organic fraction indicates the existence of a saturation phenomenon for organic C-14, as suggested under point 4) above, based on the observed concentrations in the RCS. The highest  $x_a$ -value is for the short cycle for R2 in 2012. The normalization to EFPH may also be interpreted as the number of anion equivalents divided by the total amount of reactor water passing the ion exchanger.

The regression has also been done for total resin mass with the same fit; this is reasonable, since the anion fraction in the resin is constant, 0.60, in the years analyzed. The organic fraction in the regression is 34 % compared to the mean value, 30 % given in *Table A5-4*. For SFR, the higher organic fraction is a conservative approach.

							Std.	Std.	Std.
				In-		Organic	Dev.	Dev.	Dev.
Reactor	RWCU	Unit	Total	organic	Organic	fraction	Total	Inorg.	Org.
R2-R4	Regression	(Bq/anion equivalents)/MWh <sub>th</sub>	5.79E+03	3.82E+03	2.00E+03	34%	14%	18%	10%
	1-param	r <sup>2</sup>	0.74	0.64	0.85				

Table A5-5. Results from regression analysis of accumulation factors to RWCU anion resins.(R234-1, sheet Accrat).

*Eqs.* A5-3 and A5-4 may be used to estimate the historic accumulation of C-14 in RWCU as an alternative to the average factors expressed as  $Bq/MWh_{th}$ . One possible advantage is that it



may estimate the organic C-14 better in fuel cycles or years with a low energy production or high resin use.



# APPENDIX 6 Determination of Dry Weight of Resins

## A6.1. Determination of Dry Weight of Fresh PWR Resins

In the PWR units, bead resin is added by volume from bags containing 25 liters. The gross density ("shipping density") of the resin was determined by weighing five bags in the storage. The results are shown in *Table A6-1*. The value 0.70 kg/liters has been used in all PWR calculations. The dry substance of the resin is given by the vendor as 0.45. The same value was verified in drying tests by Avenbrand in  $2005^{72}$ .

Plastic bag, kg <sup>*</sup>	*	1
Volume, liters		25
	Weight	
	(incl.	
	Bag),	Density
Bag #	kg	kg/liter
1	17.8	0.7120
2	17.8	0.7120
3	17.7	0.7080
4	17.9	0.7160
5	18.1	0.7240
Mean incl. bag	17.86	0.7144
Std. Dev.	0.1517	
Min	16.86	0.6744
Mean		0.6944
Value used		0.70
* estimated		

Table A6-1. Results from determination of gross density for PWR bead resins. (R234-1, sheet Density).

# A6.2. Determination of Dry Weight Fraction (Dry Solids, ds) of Resin Samples

For the samples analyzed in this report and in *Ref.4*, the dry weight fraction was determined in the following way:

A sample of 1.2-1.5 g was weighed in a plastic container (50 ml nominal volume). The container was covered by tissue paper and left in a fume cupboard for at least one week. Then it was weighed and the dry weight fraction calculated by: (weight after drying)/(weight before drying).

When this report was prepared, the question was raised, whether this procedure was adequate. To answer this question, a series of experiments were conducted. One sample was dried according to the old method for 7-9 days, and another sample was dried in an oven at 70-78

<sup>&</sup>lt;sup>72</sup> S. Avenbrand, personal communication 2005.

°C for 36-60 h. Then the samples were weighed and the ratio between the dry weight fractions was calculated. *Figure A6-1* shows the ratio between the dry weight fractions for air dried and oven dried samples. The complete data set may be found in *Attachment file A9*.



Figure A6-1. Correction for factors for Dry Weight. (A9, sheet D All)

	Dry			
	Substance	Std	Number of	
Comple	Conection	Dav		
Sample	Factor	Dev.	samples	
R1 mean	1.16	0.03	9	
F12 mean	1.04	0.03	6	
F3 mean	1.16	0.03	2	One sample excluded
O12 mean	1.05	0.04	2	
O3 mean	1.11	0.03	1	
Powder mean	1.10	0.07	20	
Powder maximum	1.19			
Powder minimum	0.90			
Powder sulphonate mean	1.14	0.04	13	
Powder sulphonate maximum	1.19			
Powder sulphonate minimum	1.03			
PWR (bead) mean	1.16	0.03	6	
PWR (bead) maximum	1.21			
PWR (bead) minimum	1.12			
All samples mean	1.11	0.07	26	
All samples maximum	1.21			
All samplesm inimum	0.90			

*Table A6-2. Summary of Dry Weight correction factors. (A9, sheet Summary)* 

The dry weight fraction for the samples dried in the oven was generally lower than for the air dried samples. Only one oven dried sample showed a lower dry weight fraction than the air



dried. This particular sample, F3-332-10 B2, has been excluded from the data set, since it is unlikely that the oven drying should give a lower dry weight fraction than air drying.

The standard deviations of the calculated ratios are in the range 0.02-0.04. The calculation is based on the assumption that the uncertainty in the weighing operations is 5 mg.

*Table A6-2* shows a summary of the correction factors. There seems to be two groups of samples: one with sulfonate resins (powder and bead) and one with carboxylate resins (F1+F2). However, the samples from O1, O2 and O3 have values in between these two main groups.

# A6.3. Selected Correction Factors

*Table A6-3* summarizes the selected correction factors. Three categories of resins have been identified: Powder sulfonate resins, powder carboxylate resins and sulfonate bead resins. The correction factors have been applied to all analytical results in the present report (Version 3 onwards) and the results reported in *Ref. 4*. The net result is an increase in the concentration values by 4 % for carboxylate resins and 16 % for sulfonate resins. All samples analyzed for Version 4 onwards have been oven dried.

Resin Category	Factor	Std. Dev.
BWR		
Powder sulphonate	1.16	0.04
Powder carboxylate	1.04	0.03
Bead	1.16	0.03
PWR		
Bead	1.16	0.03

Table A6-3. Dry Weight correction factors applied.(A9, sheet Summary)

## A6.4. Body Feed Factor for RWCU Resins in PWR

PWR mixed bed resins are in H<sup>+</sup> and OH<sup>-</sup> form when delivered. Before loading into the RWCU system, the resins are converted to  $\text{Li}^+$  and borate form. Borate anions are of the form B(OH)<sub>4</sub><sup>-</sup>. In addition, the resins also accumulate some corrosion products, but the mass of this uptake is much less than the mass from borate. The calculation of the BFF for PWR resins is shown in *Table A6-4*.

The resin beds are loaded with approximately the same capacity for cations and anions. The calculation of the BFF assumes that all sites are loaded. The BFF obtained, 1.15, is used to correct all results for PWR resins analyzed. The correction of old results is reported in *Appendix 7*.



	R2	R3	R4	Molecular masses	
Liters	575	650	850	O, g/mole	16
Drv sub-stance	0.45	0.45	0.45	B. g/mole	10.8
Density, ka/lit	0.7	0.7	0.7	H. g/eguivalent	1
ka ww	403	455	595	OH. a/equivalent	17
ka dw	181	205	268	B(OH)4, g/equivalent	78.8
Cation. liters	230	260	340	Li-7. g/eguivalent	7
Anion, liters	345	390	510	H3BO3, g/mole	61.8
Cation, kg dw	72	82	107	Cation	
Anion, kg dw	109	123	161	Volume fraction	0.40
Cation, equivalents	391	442	578	Equivalents/liter wet	1.7
Anion, equivalents	380	429	561	Anion	
H-load, initial, kg	0.4	0.4	0.6	Volume fraction	0.60
OH-load, initial, kg	6.5	7.3	9.5	Equivalents/liter wet 1.1	
Li-load, kg	2.7	3.1	4.0		
Monoborate load	29.9	33.8	44.2		
total extra load, kg	25.8	29.2	38.1		
BFF	1.14	1.14	1.14		
Corr.prod. Load, kg	2	2	2		
BFF incl. corr.prod.	1.15	1.15	1.15		

Table A6-4. Calculation of Body Feed Factor (BFF) for resins used in RWCU in PWR units.resins in Ref. 2, 3 and 4. (R234-1, sheet BFF).



# APPENDIX 7 Correction of Previous Results with Respect to Resin Density and Dry Substance Fraction in Samples

# A7.1. Introduction

In *Ref. 2, 3* and 4 results for bead resins were based on a resin density of 1.15 kg/liter and a dry weight fraction determined from samples dried in air. Unfortunately, the density used referred to the dry resin beads and not to the gross density ("shipping density" (*sd*)) of resin with a dry solids fraction (*dsf*) of 0.45.

The evaluation of correction factors for the dry weight fractions is described in *Appendix 6*. This chapter will perform a correction of the results previously reported for PWR and BWR bead resins.

*Table A7-1* shows the values used in the calculation of the dry weight in *Ref. 2, 3* and 4 (old) and the present study (corrected).

The formula for calculating the dry weight and the correction factor are

dry weight= "sd\*(volume wet resin)\*dsf

Overall Correction Factor = (dry weight corrected)/(dry weight old)= ( sd corrected.)/( sd old)\*( dsf corrected)/(dsf old)\* (Sample Dry Weight Correction Factor)\*BFF

Density old	kg/lit	1.15
Dry substance fraction (ds) old		0.45
Density corrected	kg/lit	0.70
Dry substance fraction (ds) corrected		0.45
Density Correction Factor		0.609
Sample Dry Weight Correction fact	tor	1.16
Body Feed Factor, BFF		1.15
Overall Correction Factor, PWR		0.812

Table A7-1. Calculation of correction factor for wrong density of the PWR and BWR bead resins in Ref. 2, 3 and 4. (R234-1, sheet Old).



### A7.2. Correction of PWR Bead Resins from Ref. 4

The overall effect of the density correction is that the dry weight is reduced to 60 % of the previous value. Inclusion of the Body Feed Factor and the Sample Dry Weight Correction Factor, increases the correction factor for the old values to 0.812. Accordingly, all values for accumulated activity in PWR resins will be reduced by this correction factor. *Table A7-2* shows the accumulation in some specific resin fraction for the PWR-units. The fractions "RWCU operation" should be compared with the results given in this report. *Table A7-3* shows corrected accumulation in PWR resins, as reported in *Ref. 4*. The corrected values are some 81 % of the old values.

Unit	Resin use	Old	Corrected	Operation	Org. fraction
R3	RWCU operation	7.3%	5.9%	2002-2003	29%
	RWCU 2 y operation				
R2	(incl. some other resins)	6.0%	4.9%	2001-2003	28%
R4	RWCU shut down, SFP, WP	1.6%	1.3%	2002-2003	35%
R3	RWCU shut down, SFP, WP	2.5%	2.0%	2002-2003	30%
Mean	RWCU operation	6.7%	5.4%		29%
Mean	RWCU shut down, SFP, WP	2.1%	1.7%		33%
Mean	All	8.7%	7.1%		31%

Table A7-2. Correction of C-14 accumulated in PWR resins reported in Ref. 4.(R234-1, sheet Old).

				Corrected	Org.
Unit	Resin use	Old	Corrected	organic	Fraction
R2	All (RWCU, SFP, WP)	8.3%	6.7%	2.0%	29%
R3	All (RWCU, SFP, WP)	9.8%	8.0%	2.3%	29%
R4	All (RWCU, SFP, WP)	9.0%	7.3%	2.2%	29%
Mean Ref. 4	All (RWCU, SFP, WP)	9.0%	7.3%	2.2%	29%
Mean this study	All (RWCU, SFP, WP)		5.2%	1.6%	29%

Table A7-3. Correction of C-14 accumulation in PWR resins reported in Ref. 4.(R234-1, sheet Old).

# APPENDIX 8 The Effect of Air Bubbling on C-14 Loaded Ion Exchange Resins

**Protolysis:** A weak acid, HA dissolved in water, partially splits into hydrogen ions (protons),  $H^+$ , and anions  $A^-$ . The resulting concentrations are determined by the equilibrium equation (law of mass action), where [x] denotes the concentration of species x.

$$\frac{[H^+][A^-]}{[HA]} = K_a$$

The equilibrium constant is often expressed as its negative logarithm

$$pK_a = -{}^{10} \log\left(K_a\right)$$

When  $pH < pK_a$ , the acid occurs as HA. For  $pH > pK_a$ , it occurs as A<sup>-</sup>. At  $pH = pK_a$ , both forms are equally abundant. *Figures A8-1* and *A8-2* show these relations graphically for "carbonic acid", formic acid and acetic acid.

Air bubbling in pure water dissolves  $CO_2$  from the air, which lowers the pH-value to about 5.5; see *Figure A8-1*. This pH-value is below the first pK<sub>a</sub> (6.37) in the carbonate system. Prolonged bubbling may convert C-14-bicarbonate on the resin to "carbonic acid" ( $CO_2$  + water) and remove it from the resin.



Figure A8-1. pH-concentration diagram for the carbonate system in pure water in equilibrium with  $CO_2$  in air. The pH-value is 5.5 at the intersection of H+ and hydrogen carbonate, marked by a "diamond". (A-8, sheet D carbonate3).

The  $CO_2$  will be swept away by the air. Storage tanks for CCU resins at the BWR units do not contain any other chemicals (except floor drain water in some cases), so "carbonic acid" is

expected to govern the pH-value. Measurements of pH in the tanks T48 and T49 at O1 and O2 (*Table A8-1* and *Figure A8-3*) have indicated a pH-value in this range. Measurements in bottles with resin samples in the laboratory at Ringhals have also indicated pH-values in the range 4.5 to 6 (*Table A8-1*).

The two simplest carboxylic acids, formic acid ( $pK_a$  3.77) and acetic acid  $pK_a$  4.76), have both been detected in PWR primary water at Ringhals in the concentration range 1-10 ppb, with peaks around 100 ppb occurring at transients (*Ref.* 2). *Figure A8-2* shows the protolytic equilibrium of these acids at these concentrations. At pH=5.5, the acids are completely in their anionic form. Thus, in low ionic strength water, they most likely remain on the resin. These conditions may explain why the organic fraction on resins is higher at low total (= inorganic) concentrations.



Figure A8-2. pH-concentration diagram for formic acid and acetic acid in concentrations measured in PWR primary water. If the water is in equilibrium with atmospheric CO<sub>2</sub>, the pHvalue is about 5.5, marked by a "diamond" and a square. At this pH, both acids are virtually 100 % in their anionic forms. (A-8, sheet D HFo HAc2).

From *Table A8-1* it can be found that all samples from BWR CCU resins at the laboratory at Ringhals had a pH-value below 6.37, the first  $pK_a$  for carbonic acid. The values measured at O1 and O2 had two values below the first  $pK_a$  and one value just above. The only sample with a value above the first  $pK_a$  was RWCU resin from R3. This is unexpected, since the boric acid should yield a pH value below 7. The value for R2, 5.96, corresponds to 20-30 ppm boron in the water.

*Figure A8-3* (same as *Figure 7* in the main report) shows that at O1+O2 18 tank fillings out of 23 had pH<  $pK_a$  .eleven of these were within ±0.2 pH units from the value of pure water in



equilibrium with atmospheric CO<sub>2</sub>, 5.5 (*Figure A8-3*). Two samples were above 7.1, where hydrogen carbonate is the dominating species; see *Figure A8-1*.

			Analysis	pH in sample	
Unit	Sample	Date	batch	at lab	Measurement
F3	F3101712	2011-08-03	F3B3	5.64	Sample at lab in Ringhals
F1 +F2	F3101717	2011-08-03	F12B4	5.71	Sample at lab in Ringhals
F3	F3111701	2011-08-03	F3B5	5.50	Sample at lab in Ringhals
01+02	T48 090208	2011-08-03		5.28	Sample at lab in Ringhals
01+02	T48 090416	2011-08-03		4.60	Sample at lab in Ringhals
01+02	T48 090623	2011-08-03		4.70	Sample at lab in Ringhals
01+02	T49 090211	2011-08-03		4.97	Sample at lab in Ringhals
01+02	T49 090316	2011-08-03		5.20	Sample at lab in Ringhals
01+02	T48 110804	2011-08-04		5.37	O1+O2 WHB
01+02	T49 110805	2011-08-05		6.90	O1+O2 WHB
O3		2011-08-19		6.00	O3 WHB
R2	R2 080611	2011-08-03		5.96	Sample at lab in Ringhals
R3	R3 080815	2011-08-03		7.59	Sample at lab in Ringhals

Table A8-1. Example of pH-values measured in samples at the lab and in storage tanks (O1, O2 and O3). The pH-values below the theoretical value for water in equilibrium with atmospheric  $CO_2$  (c:a 5.5) may be explained by cation resin still holding some  $H^{+73}$ . (A-7, sheet pH samples).



Figure A8-3. pH-values in storage tanks for powder resin at O1+O2, August-September 2011.<sup>74</sup> (A-7, sheet D pH O12).

<sup>&</sup>lt;sup>73</sup> Ingvar Carlsson, FKA. Pers. Comm., Jan. 2012.

<sup>&</sup>lt;sup>74</sup> Sofie Englund, OKG. Pers. Comm. Sept. 2011.




# APPENDIX 9 Analytical Procedures



Figure A9-1. Schematic system set-up used for the extraction of organic and inorganic C-14 from spent ion exchange resins and process water. The gas washing bottles are designated by 1–5 in the text. (Ref. 2).

### A9.1. Extraction of C-14 from the Resins

The analytical procedure used is essentially the same as described in *Ref.* 2. The method allows chemical separation by the formation of carbon dioxide (CO<sub>2</sub>) of inorganic (carbonate system) species, using acid stripping with 6 M H<sub>2</sub>SO<sub>4</sub>. Organic species (including carbon monoxide) are stripped using wet oxidation with 5 % K<sub>2</sub>SO<sub>8</sub> and 4 % AgNO<sub>3</sub> and heat. Pt/Al pellets<sup>75</sup> and CuO (30:70 weight ratio) constitute the catalytic bed to ensure complete conversion to CO<sub>2</sub>.

In brief, the resin sample (1.0-1.2 g ww) is put in an Erlenmeyer flask and a stirrer. The first 30 minutes is the acid stripping followed by 3 times 1 hour wet oxidation.

The C-14 species driven off from the resin sample are captured in wash bottles 2 and 4 (3, 5 are safety bottles in case 2 and 4 would be saturated). From the bottles 2 and 4, two aliquots of 3 ml each are mixed with 17 ml liquid scintillation cocktail (Hionic Fluor).

Samples from wash bottles 1, 3 and 5 are also analyzed, but not in duplicate as for 2 and 4. The mathematical formulas for calculations made by the software are presented in *Appendix 10*.

There are indications that not all organic species will be released from the resin. Magnusson et. al. (*Ref. 2*) have previously made combustion experiments indicating that there is C-14

 $<sup>^{75}</sup>$  In this context: Al stands for alumina, Al<sub>2</sub>O<sub>3.</sub>



activity left in the resins. Resins from all sites have been examined. Information about the combustion procedure is given in a section below.

After five batches/samples, or change of site (not applicable to PWR samples), the background is measured in order to detect any contamination of the system and for use in calculations of net activity. The background measurement is done using an Erlenmeyer flask without any resin. The wash bottles and equipment belonging to them are labeled with different colours/series, i.e. separated in different series, which makes it easier separating the backgrounds from different sites. The Erlenmeyer flasks are also marked with sites (FKA, OKG, R1 and PWR) and when doing a background the Erlenmeyer flask from the site to be analyzed is used.

# A9.2. Batching of Samples

The separation procedure is time consuming; thus several samples from the storage tanks are combined into a batch. However, the samples from OKG<sup>76</sup> and the Ringhals PWRs<sup>77</sup> were not batched.

By analyzing each sample from OKG, the variation between separate tank fillings could be followed. The reason for not batching the PWR samples is that only one sample/cycle is obtained. From each batch/sample, three replicates (for PWR: 5 replicates) were analyzed to give a measure of the variations in handling the resin and the separation equipment. If a subsample showed a large deviation from the others, further samples were analyzed. When time was limited, the deviating sample could be excluded if there would be an reasonable explanation for the deviation.

Concentrations in samples from tanks with continuous additions and removals vary more slowly; accordingly, samples may be combined into batches without losing information.

When batching samples, it was necessary to use samples (no more than 10 samples/batch) originating from the same operating conditions; if a reactor was shut down, the samples from that period were batched together. The weight from each sample to the batch was measured in order to make an estimate of the contribution from each sample.

#### **A9.2.1. Batching Procedure**

- decant as much water as possible from the can
- shake the can to a homogenous suspension
- filter the suspension by water suction (do not filter so hard that the resin will partly dry)
- take the resin into a new can or bottle, note the weight of the resin

When R1 2010 was batched, there was so much resin (>200 g/sample) that the resin had to be mixed on the filter and then approximately 100 g from each sample was taken to the batch.

<sup>&</sup>lt;sup>76</sup> A few samples From O1+O2 without CCU resins have been batched before analyses. The samples from O3 included samples from several filters, but were not batched further prior to analysis.

<sup>&</sup>lt;sup>77</sup> One resin bed was sampled several times and batched before analysis, as described in Appendix 5.



### **A9.3. Combustion of Extracted Resins**

Magnusson showed in her dissertation (*Ref. 2*) that not all C-14 could be extracted; the reason for this is unknown. One explanation might be that some molecules have been incorporated in the skeleton in the resin. Combustion of the analyzed resin is one method to release these molecules.

*Figure A9-2* shows the principle of the combustion system and *Figure A9-3* shows the system (without gas supply) in the laboratory.

The analyzed resin was suction filtered using a weighed,  $0.2 \mu m$  filter from Micro Filtration System. After filtration, the filter was allowed to dry in a fume cupboard over night. The dried resin was scraped off the filter and weighed.



Figure A9-2. Schematic system set-up used for the combustion of spent ion exchange resins. The gas washing bottles are designated by 1–3 in the text.

The resin was ground in a mortar before it was laid on the glass wool in the tube. Magnusson (*Ref. 2*) used oxygen in her studies, but in this study technical air was used in order to minimize the explosion risks when using pure oxygen. Using the same catalyst as in the analyses resulted in a lower concentration of CO than using only Pt/Al pellets (~30 ppm vs. >500 ppm)<sup>78</sup>.

The gas first passes bottle 1 to remove tritium and sulfur compounds. Then it entered the catalytic furnace where any remaining CO is oxidized to  $CO_2$ , which is absorbed in bottles 2 and 3. All C-14 obtained here is assumed to be of organic origin.

#### A9.3.1. Analyses of Samples from Combustion

3 ml from each bottle (duplicates from bottle 2) were analyzed the same way as the extracted samples. However, the calculations differ, see *Appendix 10*.

<sup>&</sup>lt;sup>78</sup> It was feared that CuO should react with sulfur compounds from the combustion of the resins. Actually, it did react, so the catalyst had to be replaced after the experiment. Measurements of CO during the experiments indicated that the catalyst was intact during these experiments.



#### A9.3.2. Correction Obtained from Combustion Results

The results from combustions of PWR, OKG and FKA showed that there is not more than 5% left in the resin after analyses; thus the factor, from Magnusson's previous experiments, (1.05) for the organic residues will remain unchanged. For details on the determination of the combustion correction factor, see *Appendix 17*. However, the combustion of the samples of the dryer resulted in a much higher (~17 %) of the organic fraction. This has only been showed once since only one sample from each dryer has been analyzed.



*Figure A9-3. System set-up used for the combustion of spent ion exchange resins. The gas washing bottles are designated by 1–3 in the text.* 

# A9.4. Liquid Scintillation Counting of the Samples

#### A9.4.1. Equipment

The samples are counted by a liquid scintillation counter Hidex 300 SL.

#### A9.4.1.1.Calibration:

Instead of using quench curves, efficiency calibration was made using 3\*5 standard solutions (three series with five samples in each series.) The solutions were prepared with a known amount of C-14 activity (104300 DPM) in the same matrix (3 ml 2M NaOH + 17 ml Hionic Fluor) as the measured samples. The prepared standard samples were counted 2-3 times each. From these countings, the efficiency was determined to be 0.865. with a standard deviation of 0.25 %. The energy window was chosen to maximize the Figure of Merit (efficiency<sup>2</sup>/background). After an upgrade of the instrument in February 2014 the efficiency was re-determined to 0.885.

Unquenched samples from the vendor of counter are used to regularly check the constancy of the counter. These results are stored in the counter.

#### **A9.4.2. Sample Counting**

Two aliquots of 3 ml of alkaline samples each are mixed with a liquid scintillation cocktail (Hionic Fluor). After 24 hours in darkness, the samples are counted three times, each time for one hour. Acidic samples (from bottle 1) are counted with Quicksafe 400.



#### **A9.4.3. Quality Assurance**

No gamma spectrum measurements are performed on the collected samples. It is expected that all gamma emitting nuclides remain in the Erlenmeyer flask or are captured in bottle 1. Tritium is also expected to remain in bottle 1. To verify this, the beta spectra are inspected with respect to appearance; correct C-14 endpoint energy and absence of a low energy hump from tritium in the C-14 samples.

If duplicate samples show a large scatter, new samples are counted. If the scatter persists, a new sample of the resin is extracted and counted.

The analysis results are included in the Attachment files with the calculations for each unit.







# APPENDIX 10 Calculations of Activity Data in Liquid Scintillation Counting (MicroWin 2000) and in Excel-sheets

# A10.1. Calculations in Hidex Liquid Scintillation Counter in MicroWin 2000

#### A10.1.1. Average Counts per Minute: CPMAVG

For each sample, two aliquots are counted; vial A and B Each vial is counted several times; usually three times

 $CPM_{AVG} = \frac{Mean(all \ counts \ vial \ A) + Mean(all \ counts \ vial \ B)}{2}$ 

#### A10.1.2. Standard Deviation in CPMAVG; $\sigma_{CPM}$

$$\sigma_{\text{CPM}} = \sqrt{\frac{\alpha + \beta}{n - 1}}$$

$$\alpha = (CPM_{A}P001 - CPM_{AVG})^{2} + (CPM_{A}P002 - CPM_{AVG})^{2} + (CPM_{A}P003 - CPM_{AVG})^{2}$$
  
$$\beta = (CPM_{B}P001 - CPM_{AVG})^{2} + (CPM_{B}P002 - CPM_{AVG})^{2} + (CPM_{B}P003 - CPM_{AVG})^{2}$$

n = total times vials A and B have been counted = 6 (each vial is counted 3 times)  $CPM_{A/B}P001$ ,  $CPM_{A/B}P002$  and  $CPM_{A/B}P003 = P$ late run 1, 2 and 3 (separate countings)

#### A10.1.3. Gross activity in bottle, Bq: *A*<sub>Brutto</sub>

$$\mathbf{A}_{\text{Brutto}} = \frac{CPM_{AVG}}{\eta \times V_{vial} \times s} \times V_{bottle}$$

η efficiency in counting (0.865)
the efficiency (86.5%) is a fixed value previously determined from a set of standard solutions spiked with a known amount of C-14.
V<sub>vial</sub> volume of sample in vial (3)

V<sub>bottle</sub> volume of bubble bottle (100 ml)

s conversion seconds/minute; 60



#### A10.1.4. Gross background activity in bottle, Bq: ABruttobkg

 $A_{Bruttobkg} = \frac{CPM_{AVGBKG}}{\eta \times V_{vial} \times s} \times V_{bottle}$ 

A10.1.5. Net activity in bottle, Bq; either organic or inorganic:  $A_{netto}$  $A_{Netto} = A_{Brutto} - A_{Bruttobkg}$ 

#### A10.1.6. Net concentration, Bq/kgww: CInorganic, COrganic and CCombustion

$$C_{(ln)organic} = \frac{A_{netto}}{m_{ww}} \times 1000$$

m<sub>ww</sub> sample wet weight (g)

1000 conversion from g to kg

#### A10.1.7. Detection Limit Background, CPM: DLBKG CPM

$$DL_{BKG CPM} = \frac{1.65^2}{n \times t} + \left(2 \times 1.65 \sqrt{\frac{CPM_{AVG}}{n_b \times t_b}} \times (1 + (\frac{n_b \times t_b}{n_s \times t_s})\right)$$

- 1.65 factor for 95 % confidence level
- n number of countings (6)
- t counting time, minutes; 60
- b index background
- s index sample

#### A10.1.8. Detection Limit, Bq/kg: DL

$$DL = \frac{DL_{BKG CPM} \times V_{Bottle} \times 1000}{\eta \times V_{Vial} \times s \times m_{ww}}$$

V <sub>Bottle</sub>	volume of the bubble bottle
V <sub>vial</sub>	volume of sample in vial
S	conversion seconds/minute; 60

#### A10.1.9. Statistical Uncertainty (Statunc)

$$Stat_{unc} = \frac{1}{\sqrt{number of \ counts}} = \frac{1}{CPM_{AVG} \times total \ count \ time}$$

The total counting time in minutes =  $60 \text{ minutes} \times 6 \text{ runs}$ 



# A10.1.10. Relative Uncertainty (*Rel<sub>unc</sub>*) Re $l_{unc} = \frac{CPM_{STDAV}}{CPM_{AVG}}$

#### A10.1.11. Error Brutto ,Bq also used to calculate Error Background

Error Brutto=((Stat<sub>unc</sub>>Rel<sub>unc</sub>) × Stat<sub>unc</sub>+(Stat<sub>unc</sub>< Rel<sub>unc</sub>) × Rel<sub>unc</sub>) ×  $c_{Brutto}$ 

This mathematical formula is an "IF-statement". IF ( $Stat_{unc} > Rel_{unc}$ ) is true it will be 1, if it is not true, it will be 0. IF ( $Stat_{unc} > Rel_{unc}$ ) is true, then Error Brutto will be  $Stat_{unc} \times c_{Brutto}$ . If it is not true, then Error Brutto will be  $Rel_{unc} \times c_{Brutto}$ .

#### A10.1.12. Error CPM

 $Error CPM = ((Stat_{unc} > Rel_{unc}) \times Stat_{unc} + (Stat_{unc} < Rel_{unc}) \times Rel_{unc}) \times CPM_{avg}$ 

This is just like Error Brutto, an IF-statement.

#### A10.1.13. Error Netto, Bq

*Error* Netto = 
$$\sqrt{Error Brutto^2 + Error BKG^2}$$

Error BKG error background

#### A10.1.14. Error Total Bq/kg

$$\sqrt{ \left( \left( \frac{100 \times 1000}{\eta \times V_{vial} \times s \times m} \right)^2 \times \sigma_{CPM}^2 \right) + \left( \left( \frac{CPM \times 1000}{\eta \times V_{vial} \times s \times m} \right)^2 \times 1.5^2 \right) + \left( \left( \frac{CPM \times 100 \times 1000}{\eta^2 \times V_{vial} \times s \times m} \right)^2 \times 0.002163^2 \right) + \left( \left( \frac{CPM \times 100 \times 1000}{\eta \times V_{vial}^2 \times s \times m} \right)^2 \times 0.020^2 \right) + \left( \frac{1000}{m} \right)^2 \times \sigma_{BKG}^2 + \left( \frac{CPM \times 100}{\eta \times V_{vial} \times s} - c_{BKG} \times \frac{1000}{m^2} \right)^2 \times (0.01m)^2$$

S	conversion seconds/minute; 60
100	volume in wash bottle, ml
1000	convert g to kg
$\mathbf{V}_{\text{vial}}$	volume of sample in vial $= 3 \text{ ml}$
$\sigma_{\scriptscriptstyle CPM}$	error in CPM
1.5	error when measuring volume to wash bottle, ml
0.0021	63 error in efficiency, absolute
0.020	error in pipette volume, ml
0.01m	error in mass, g



### A10.2. Calculations in Excel-sheets

#### A10.2.1. Combustion correction factor for the organic fraction *f*

 $f = \frac{a_{nettocombustion} + a_{nettoorganic}}{a_{nettoorganic}}$ 

Where

*a* activity found in the sample at combustion and extraction of organic, respectively, Bq

The value f=1.05 has been adopted for all samples, except for dried resins from FKA, where individual combustion correction factors have been determined; see *Appendix 21*. This correction factor is applied only to the organic fraction

$$c_{org\ corr} = fc_{org}$$

#### A10.2.2. Calculation of total concentration, Bq/kg<sub>ww</sub>

 $c_{total} = c_{inorg} + c_{org \ corr}$ 

#### A10.2.3. Calculation of total concentration, Bq/kgdw

$$c_{dw} = \frac{c_{total}}{ds}$$

#### A10.2.4. Calculation of standard deviation in total concentration, Bq/kg<sub>dw</sub>

$$\sigma_{total\ dw} = \frac{\sqrt{\sigma_{total\ inorg}^2 + f^2 \sigma_{org\ corr}^2}}{ds}$$

$$\sigma_{org\,dw} = \frac{f\sigma_{org\,corr}}{ds}$$



#### A10.2.5. Standard deviation used in calculations total resp. organic, Bq/kg<sub>dw</sub>

 $\sigma_{dw} = \max(\sigma_{dw}; std \ dev. (scatter) \ for \ the \ subsamples \ in \ the \ batch)$ 

#### A10.2.6. Total Standard Deviation from Activity Counting and Resin Amounts

The relative standard deviation is combined according to

$$\sigma_t = \sqrt{(\sigma_a^2 + \sigma_r^2)}$$

Where



Figure A10-1. The total standard deviation resulting from a constant standard deviation in the resin amount of 10 % and a variable standard deviation from activity counting. (A-6, sheet D Comp).

### A10.3.Using Detection Limit Values

The detection limit is calculated from the standard deviation in the background measurements. (see the paragraph Detection Limit, Bq/kg: DL above).

The DL value has a confidence level of 95 %, meaning that in 95 % of the measurement an activity exceeding the DL will be detected. In addition, there is a 5 % probability of detecting an activity less than the DL. *Ref. 9* shows the deduction of the DL value.



The net value in a measurement is obtained by subtracting a background value from the sample value. Depending on the activity levels and standard deviations in the background and sample values, a sample may show a positive or a negative net value.

To cope with this, detection limit values are handled in the same way as described in the EU recommendation for reporting releases. (*Ref. 5* and *6*; The Swedish implementation is regulated by *Ref. 7*).

The following criteria are used:

If the detected value V is greater than half the detection limit DL, the detected value V is used.

If the detected value V is less than half the detection limit DL, one quarter of the detection limit value will be used.

Or, with formulas:

$$If \left(V > \frac{D}{2}\right); use V$$
$$If \left(V < \frac{DL}{2}\right); V = \frac{DL}{k}$$
$$k = 4$$



# APPENDIX 11 Handling and Sampling of Resins in the Various Plants

# A11.1. Introduction

There are large variations between the systems for handling and sampling the spent resins in the various plants. In this chapter, a brief outline is given together with comments. *Ref. 2* has figures describing where in the process systems the ion exchangers are located.

# A11.2. PWR Ringhals R2, R3 and R4; RWCU

The original system for waste solidification of spent resins in the PWR units had a poor performance, compared to the system in the BWR WHB. Further, since the boric acid present in PWR resins will lower the strength of the solidified concrete product, it was chosen to transfer the resins to the BWR WHB and dilute the PWR resins with boron-free resins from the BWR unit.

The transport is performed by means of a lead shielded spherical transport vessel ("kula"). Some 3-7 separate transports are needed to transfer a RWCU resin load. The handling system has no representative sampling system, so the samples are withdrawn by means of a 20 ml plastic bottle. 1 -2 subsamples are taken from each transport vessel load and batched on completion of the transport campaign.

The radiation dose amounts to about 0.2-0.3 mSv for a campaign to sample the RWCU resin from one PWR unit<sup>79</sup>.



Figure A11-1. Overview of handling and sampling of spent RWCU ion exchange resin in PWR units R2, R3 and R4. (A-5, pict.1).

<sup>&</sup>lt;sup>79</sup> Anders Höglund, Ringhals, R1M, Pers. Comm. Sept. 2011.



### A11.3. BWR Ringhals R1; CCU

The transfer from the unit to the WHB is done by means of permanent piping. The sampling is done from a pipe where the resin slurry is recirculated with a pump from the storage tank after mixing by air bubbling. This sampling point is also used to determine the dry solid contents of the resin slurry, which is important to get the full strength of the cement solidification.



Figure A11-2. Overview of handling and sampling of spent CCU ion exchange resin in Ringhals 1 BWR. (A-5, pict.2 & 7).

**Collecting samples R1**: At the bottom of the storage tank there is a ventilation ring. After mixing the tank with air for approximately 20 min, the slurry is pumped through a piping system (see *Figure A11-2*). In the piping system there is a valve from which samples are taken. Before the sample is taken, some slurry is discarded in case there is old slurry in the valve.

### A11.4. BWR Forsmark F3; CCU

The handling of the CCU resins in Forsmark 3 is similar to the handling in R1. The transfer from the unit to a storage tank in the WHB is done by means of permanent piping. The tank content is mixed by gentle water injection from the bottom. Before solidification in bitumen, the resin slurry is evaporated to reduce the water contents in order to get maximum strength of the solidification.

The sampling is done by a permanent sampling system in the pipe transferring resin slurry from the storage tank to the dryer. This is the last possible sampling point before drying and solidification.





Figure A11-3. Overview of handling and sampling of spent CCU ion exchange resin in BWR Forsmark 3. (A-5, pict.4).

### A11.5. BWR Forsmark F1 and F2; CCU



Figure A11-4. Overview of handling and sampling of spent CCU ion exchange resin in BWR units Forsmark 1 and  $2^{80}$ . (A-5, pict.8).

The handling of the CCU resins in Forsmark 1 and 2 is a hybrid of the handling in R1, Ringhals PWR units and F3. Via a temporary storage tank in the unit (back-flushing tank) the resin is collected either of the tanks 342T42 and T43 in F1 WHB. These tanks are mixed by air bubbling<sup>81</sup>. Then the resin is transported to a storage tank in the WHB at F3 by similar transport vessels as used for the Ringhals PWRs. The storage tank is mixed by stirring, not by

<sup>&</sup>lt;sup>80</sup> Tank TC56 is not the normal storage tank for resins from F1+F2. Normally these resins are mixed with the corresponding resins from F3. However, TC56 was used during the sampling campaign to keep the F1+F2 resins separated from the F3 resins. Annelie Jansson, FKA, personal communication, November 2015.

<sup>&</sup>lt;sup>81</sup> Roger Jansson, Forsmark 1 Avfall, Pers. Comm. 22 Sept. 2011.



air bubbling. Before solidification in bitumen, the resin slurry is evaporated to reduce the water contents in order to get maximum strength of the solidification.

The sampling is done by a permanent sampling system in the pipe transferring resin slurry from the storage tank to the dryer. This is the last possible sampling point before drying and solidification. Sampling may be also done, without transferring resin to the dryer.

# A11.6. BWR OKG O1 and O2; CCU

The handling of CCU resins at O1 and O2 follows the following procedure: From a temporary storage tank in each unit ("back-flushing tank"), the resin is transferred via a permanent piping system to the Waste Handling Building (WHB). In the WHB the resins are stored in two tanks, T48 and T49. These tanks also receive floor drain water.

After air bubbling for 30 minutes and pump mixing in a circulating loop for another 15 minutes, a 5 liter sample will be taken out from the circulation loop. The sample is allowed to sediment for 2 hours after which the sediment will be transferred to a 250 ml bottle for analysis.

Finally, the resin slurry is transferred to concrete tanks were it will be dewatered by suction via filter rods placed in the bottom of the concrete tank. The closed down units B1 and B2 at Barsebäck had a similar procedure for handling the CCU resins.



Figure A11-5. Overview of handling and sampling of spent CCU ion exchange resin in BWR units O1 and O2. (A-5, pict.5).

# A11.7. BWR OKG O3; CCU

The handling of CCU resins at units O3 is similar to the procedures in O1 and O2: From a temporary storage tank in the unit (back-flushing tank), TB14, the resin is transferred via a permanent piping system to the Waste Handling Building, tank TB41. For the C-14 sampling campaign, only TB41 is used for CCU resins (to avoid mixing with other powder resins).



After air bubbling<sup>82</sup> and pump mixing in a circulating loop for 1 hour, a sample will be taken out from the loop. The sample is allowed to sediment for 2 hours after which the sediment will be transferred to a 250 ml bottle for analysis.

Finally, the resin slurry in the tank is transferred to concrete tanks were it will be dewatered by suction via filter rods placed in the bottom of the concrete tank.



Figure A11-6. Overview of handling and sampling of spent CCU ion exchange resin in BWR unit O3. (A-5, pict.6).

# A11.8. BWR Barsebäck B1 and B2; CCU

The handling of CCU resins at B1 + B2 has been assumed to be similar to O1 + O2. However, there was no air bubbling, just pump circulation of the resin slurry before transfer to concrete  $tanks^{83}$ .

<sup>&</sup>lt;sup>82</sup> For the resin batches analyzed in this report (2010 and 2011) the following conditions had been used: The operational procedures have not included air bubbling, just pump circulation. Pers. comm. K-E Ingemansson, 2012-01-03.

<sup>&</sup>lt;sup>83</sup> Lars Wiklund, BKAB, via Lars Håkansson, BKAB. Pers. Comm. 2012-04-12.







# APPENDIX 12 Example of Analysis Data

Samma	nstä	Ilnin	ng F	Ring	hals	33	4			TS-korr.fakto	ı 1.16	Gammal T	S-halt ska div	ideras med	faktom!				2	Det. Lim. Fra	act. For using	detected		det. Limit f	actor	
																	]		Modifierat 20	012-03-16						
	Förbränt (	delprov																								
Urspr TS-halt (Torr/våt vikt) =	0.36	<sup>5</sup> 1.16	Korrekt	tionsfaktor "	TS (Ny/Gam	Gamma	TS-halt	t i B. Korr.fa	aktor i C	länkad från L	.1!															
R2-334-08																										
Start (driftsättning) =			R2 CSE	DMMB-02:				Komm	nentar as	mz:ANHG:R	2-334-08: 13	3 klockor to	otalt, varav 3	st var tomn	ia (= bara	vatten?) o	ch 3 st med	l innehåll på	<20 liter							
Stopp (togs ur drift) =			2007-08	8-01 Tömd (	och fylld med	575 I SB	MR-72	jb-ma	ssa. 13-3	-3 = 7 st kloc	kor som pro	ovtogs. Frå	in Varje klock	a ska enligt	procedure	en tas dubl	pelprov a di	ygt 20 ml m	iassa.							
TS-halt (Torr/våt vikt) =	0.31		2008-0	5-17 Tömd (	och fylld med	1575 I SB	MR-72																			
																							Fraction of	f detection I	imit used to	replace zer
Delprov	Inorganic <sup>14</sup> C (Bq/kg w w )	Error TOTAL (Bq/kg)	Error TOTAL (%)	Detection limit (Bq/kg w w )	Organic <sup>14</sup> C (Bq/kg w w)	Error TOTAL (Bq/kg)	Error TOTAL (%)	Detection limit (Bq/kg ww)	Correcti on f= 1.05	Corrected Organic <sup>14</sup> C (Bq/kg w w)	Total <sup>14</sup> C (Bq/kg w w )	Organic Fraction (%)	Total <sup>14</sup> C (Bq/kg dry)	Gamma total (Bq/kgww)	Co-60 (Bq/kg ww)	Co-60 (Bq/kg dry)	Inorg <sup>14</sup> C (Bq/kg dry)	Corrected Organic <sup>14</sup> C (Bq/kg dw )	Error total total Bq/kg dw	Error total inorganic Bq/kg dw	Error total organic Bq/kg dw Bq/kg dw	Total, Bq/kg dw	Inorganic, Bq/kg dw	Organic, Bq/kg dw	Organic Fraction (%)	Error organic fraction
R2-334-08:1	1.70E+07	3.30E+05	1.9%	8.30E+02	2.90E+06	5.60E+04	1.9%	8.80E+02	1.05	3.05E+06	2.00E+07	15%	6.46E+07				5.48E+07	9.81E+06	1.08E+06	1.06E+06	1.89E+05	6.46E+07	5.48E+07	9.81E+06	15.2%	0.4%
R2-334-08:2	1.60E+07	3.10E+05	1.9%	9.35E+02	2.80E+06	5.40E+04	1.9%	9.85E+02	1.05	2.94E+06	1.89E+07	16%	6.10E+07				5.16E+07	9.47E+06	1.02E+06	9.99E+05	1.83E+05	6.10E+07	5.16E+07	9.47E+06	15.5%	0.4%
R2-334-08:3	1.60E+07	3.10E+05	1.9%	8.00E+02	2.90E+06	5.60E+04	1.9%	8.40E+02	1.05	3.05E+06	1.90E+07	16%	6.14E+07	4.54E+08	1.24E+08	4.00E+08	5.16E+07	9.81E+06	1.02E+06	9.99E+05	1.89E+05	6.14E+07	5.16E+07	9.81E+06	16.0%	0.4%
R2-334-08:4	1.57E+07	3.05E+05	1.9%	8.16E+02	2.70E+06	5.27E+04	2.0%	8.61E+02	1.05	2.84E+06	1.86E+07	15%	5.98E+07				5.07E+07	9.14E+06	9.99E+05	9.83E+05	1.78E+05	5.98E+07	5.07E+07	9.14E+06	15.3%	0.4%
R2-334-08:5	1.70E+07	3.20E+05	1.9%	8.92E+02	3.00E+06	5.90E+04	2.0%	9.40E+02	1.05	3.15E+06	2.02E+07	16%	6.49E+07				5.48E+07	1.02E+07	1.05E+06	1.03E+06	2.00E+05	6.49E+07	5.48E+07	1.02E+07	15.6%	0.4%
Average	1.63E+07									3.00E+06	1.93E+07	16%	6.235E+07				5.27E+07	9.68E+06	4.62E+05	4.54E+05	8.41E+04	6.23E+07	5.27E+07	9.68E+06	15.5%	0.4%
Std deviation	6.07E+05									1.20E+05	7.07E+05	3.18E-03	2.28E+06				1.86E+06	1.95E+05	0.7%	0.9%	0.9%	2.04E+06	1.75E+06	3.45E+05	0.3%	0.1%
Standard dev %	3.7%									4.0%	3.7%	2.0%	3.7%				3.5%	2.0%	3.3%	3.3%	3.6%	3.3%	3.3%	3.6%	1.8%	1.8%

Table A12-1. Analysis data from Ringhals 2 RWCU 2008. (R234-1, sheet RAB-334).

Samma	anstä	ällnir	ng F	Ring	hals	332	2			TS-korr.fakto	5 1.16	Gammal T	S-halt ska o	divideras m	ed faktorn!				2	Det. Lim. F	Fract. For u	sing detect	4	Detection I	imit factor	
Urspr TS-halt (Torr/våt vikt) =	0.40	1.16	Korrektio	onsfaktor TS	S (Ny/Gamn)	Gammal T	S-halt i E	3. Korr.fakto	or i C länl	kad från L1!									Modifierat 201	2-03-16						
R1-332-200	9		Batch 1 u provtagni	utgör ett Noll ing 2009-01	prov. Tanken -23 200 kg m	innehöll vi assa som	d använts																			
Start (driftsättning) = Stopp (togs ur drift) =			under sis	sta delen av	revision. Inge	n effektdri	ft! Se																			
TS-halt (Torr/våt vikt)	0.34		g (gansk	a blöt mass	a)	B-332-200	J9". 4,22																			
																							Fraction of	f detection l	imit used to	p replace ze
Delprov	Inorganic <sup>14</sup> C (Bq/kg w w)	Error TOTAL (Bq/kg)	Error TOTAL (%)	Detection limit (Bq/kg ww)	Organic <sup>14</sup> C (Bq/kg w w )	Error TOTAL (Bq/kg)	Error TOTAL (%)	Detection limit (Bq/kg w w)	Correctio n f= 1.05	Corrected Organic <sup>14</sup> C (Bq/kg w w )	Total <sup>14</sup> C (Bq/kg w w )	Organic Fraction (%)	Total <sup>14</sup> C (Bq/kg dry)	Gamma total (Bq/kgww)	Co-60 (Bq/kg w w )	Co-60 (Bq/kg dry)	Total Inorg <sup>14</sup> C (Bq/kg dry)	Total Org <sup>14</sup> C (Bq/kg dry)	Error total total Bq/kg dw	Error total inorganic Bq/kg dw	Error total organic Bq/kg dw Bq/kg dw	Total, Bq/kg dw	Inorganic, Bq/kg dw	Organic, Bq/kg dw	Organic Fraction (%)	Error organic fraction
R1-332-09B1:1	8.18E+03	5.37E+02	6.6%	9.26E+02	0.00E+00	4.32E+02		9.26E+02	1.05	0.00E+00	8.18E+03	0.0%	2.38E+04				2.38E+04	0.00E+00	2.05E+03	1.57E+03	1.32E+03	2.45E+04	2.38E+04	7.08E+02	2.9%	5.4%
R1-332-09B1:2	6.02E+03	4.71E+02	7.8%	8.64E+02	1.91E+03	4.19E+02	2.19E-01	8.92E+02	1.05	2.01E+03	8.03E+03	25.0%	2.34E+04	1.73E+04	1.60E+04	4.66E+04	1.75E+04	5.57E+03	1.88E+03	1.37E+03	1.28E+03	2.34E+04	1.75E+04	5.85E+03	25.0%	5.8%
R1-332-0XB1:3	6.87E+03	4.99E+02	7.3%	8.97E+02	1.61E+03	9.26E+02	5.74E-01	9.45E+02	1.05	1.70E+03	8.56E+03	19.8%	2.49E+04				2.00E+04	4.70E+03	3.19E+03	1.46E+03	2.83E+03	2.49E+04	2.00E+04	4.94E+03	19.8%	11.6%
Average	7.02E+03									1.23E+03	8.26E+03	15%	2.405E+04				2.046E+04	3.425E+03	1.41E+03	8.47E+02	1.13E+03	2.43E+04	2.05E+04	3.83E+03	15.9%	8.1%
Std deviation	7.69E+02									7.64E+02	1.94E+02	9.33E-02	5.66E+02				3.170E+03	2.998E+03	5.8%	4.1%	29.4%	1.41E+03	2.59E+03	2.24E+03	9.4%	1.3%
Standard dev %	11.0%									61.9%	2.4%	62.5%	2.4%				15.5%	87.5%	5.8%	12.7%	58.5%	5.8%	12.7%	58.5%	59.4%	59.4%

Table A12-2. Analysis data from Ringhals 1 CCU 2010. (R1-2, sheet RAB-332).



The Rightmost section is used to calculate averages, sums and standard deviations. The standard deviation is the maximum of the scatter between the subsamples and the standard deviation in the counting. If the detected value is less than half the detection limit, one quarter of the detection limit value will be used. See *Appendix 10* for explanation.



# APPENDIX 13 Results from O3

### A13.1. Resin Handling

All calculations for O3 are included in an excel-book. It can be found as *Attachment File O3-2* to this report. Reference to the various sheets will be made at tables, figures etc.



Figure A13-1. Overview of handling of spent powder resin at  $O3^{84}$ . (A-5, pict.6).

The storage tank TB41 is used to store the spent resins. The powder resin has sulfonate resin in the cation part. For some time, inert material was also added as body-feed. *Figure A13-1* schematically shows the handling procedures for the spent CCU resins. However, the first two resin batches sampled for this report were not air bubbled; see the footnote to *Figure A13-1*.

The sampling records were delivered as Excel-books, *Attachment file O3-1. Table A13-5* shows an example of such a record. The operational time for each CCU filter is obtained from the table. However, the periods have been checked with data from *Attachment file O3-4*, where the total volumes of water for each filter have been calculated by P. Arvidsson, OKG.

From daily average values for the reactor power and the flow rate through each CCU filter, the "energy exposure" for each filter was calculated (*O3-4, sheet e332f*). An example is shown in *Table A13-6*.

It has been assumed that C-14 distributes uniformly between the filters. The production of C-14 is calculated from the thermal energy produced and the production rate obtained from the FSAR.

<sup>&</sup>lt;sup>84</sup> For the two first resin batches analyzed in this report the following conditions had been used: The operational procedures have not included air bubbling, just pump circulation. Pers. comm. K-E Ingemansson, 2012-01-03.





### A13.2. Calculations and Results

Figure A13-2. Concentration of C-14 in CCU resin samples from O3. The reactor power is also displayed. The curve for the inorganic concentration is covered by the curve for the total concentration. (O3-4, sheet D Conc new).

					C14 inorg,	C-14 org,			Inorg.	
Sam-				C14 tot,	Bq/kg	Bq/kg	Org-	Tot. Std.	Std.	Org. Std.
ple	Tank log	Start	End	Bq/kg dw	dw	dw	fract.	dev.	dev.	dev.
O3-1	O3 110412	2009-09-13	2011-03-10	1.39E+06	1.36E+06	2.68E+04	1.9%	1.1%	1.2%	28%
O3-2	O3 110818	2010-01-11	2011-08-03	2.39E+06	2.36E+06	2.23E+04	0.94%	1.1%	1.2%	28%
O3-3	O3 111031	2011-04-17	2011-10-19	3.85E+06	3.79E+06	5.17E+04	1.4%	2.0%	2.4%	36%
O3-4	O3 120217	2011-09-08	2012-02-15	4.41E+06	4.37E+06	4.66E+04	1.1%	2.0%	2.4%	36%
O3-5	O3 120717	2011-11-23	2012-07-10	2.82E+06	2.80E+06	2.37E+04	0.84%	3.0%	3.1%	19%
O3-6	O3 121003	2012-02-29	2012-10-03	3.74E+06	3.72E+06	2.71E+04	0.72%	3.2%	3.0%	30%
O3-7	O3 130103	2012-07-26	2012-12-21	4.09E+06	4.06E+06	3.85E+04	0.94%	1.6%	1.7%	11%
O3-8	O3 130306	2012-10-23	2013-03-04	3.68E+06	3.62E+06	6.20E+04	1.7%	4.4%	4.5%	5.9%
O3-9	O3 130604	2013-01-09	2013-05-03	3.12E+06	3.09E+06	3.53E+04	1.1%	3.2%	3.0%	26%
O3-10	O3 130827	2013-01-30	2013-08-25	3.38E+06	3.33E+06	4.03E+04	1.2%	2.9%	2.8%	19%
O3-12	O3 12 140114	2013-10-09	2014-01-08	3.67E+06	3.63E+06	3.73E+04	1.0%	1.9%	1.7%	32.1%
O3-13	O3 13 140221	2013-11-26	2014-02-19	3.98E+06	3.88E+06	9.59E+04	2.4%	1.1%	1.9%	40.8%
O3-14	O3 14 140419	2013-12-16	2014-04-11	6.69E+06	6.54E+06	1.46E+05	2.2%	1.1%	1.3%	18.9%
O3-15	03 15 140501	2014-03-10	2014-05-01	2.55E+06	2.52E+06	2.98E+04	1.2%	8.2%	8.3%	25.1%
O3-16	O3 16 140829	2014-03-01	2014-08-29	3.63E+06	3.54E+06	9.12E+04	2.5%	4.1%	3.8%	21.7%
Mean		2009-09-13	2014-08-29	3.56E+06	3.51E+06	5.17E+04	1.5%	31%	30%	63%
O3-8: D	ry substance d	etermination	uncertain. Co	nservative v	alue used					
* Numb	er of subsample	es with (0.25*	detection limi	t) instead o	f detected	values				

Table A13-1. Summary of concentrations in samples of CCU resins from O3. No detectionlimit values have been used. (O3-2, sheet Conc).

*Figure A13-2* and *Table A13-1* show the concentrations of C-14 in the resin samples. Each concentration value is the average of three individual samples of a batch. All samples yielded values above the detection limit<sup>85</sup>.

The standard deviation in the concentrations is the largest value of the scatter between the three samples analyzed in each batch and the uncertainty in counting. The scatter is usually larger than the uncertainty in activity counting.

The amount of C-14 accumulated is calculated according to the Batch Model (BM) from the concentration in the samples, Bq/kg dw, multiplied with the total contents in the tank, (kg dw). The uncertainty in the resin mass has been assumed to be 10 %.

From this accumulated activity, the accumulation expressed as a percentage of the production and as Bq/MWh<sub>th</sub> is calculated. *Table A13-6* shows an example of such a calculation for one resin batch. *Table A13-2* shows a summary for all the samples from O3 analyzed for this report.

From *Table A13-2* it can be seen that the accumulation of C-14 in the batches ranges between 0.72 % and 5.3 % of the calculated production. The mean value is 2.3 %. The organic fraction also varies between the batches. The average organic fraction of 1.4 % is in the lower range for BWRs in this study. However, *Ref. 4* reports some values in the range 15-30 % for BWR resins. The standard deviation in the sum of accumulated C-14 is 2.9 %.



Figure A13-3. Activity contents in the tanks sampled (O3-2, sheet D Sum act).

<sup>&</sup>lt;sup>85</sup> If the detected value is less than half the detection limit, one quarter of the detection limit value has been used. In most cases, this leads to an overestimation in low concentration samples; see *Appendix 10*. Usually, it also leads to an overestimation of the organic fraction, since most detection limits values have been used in the organic fractions.

Resin	uncertainty					10%												
Sam- ple	Tank log	Start	End	Effective energy, MWhth	Calc. Produc- tion, Bq	Resin, kg dw	Accum. C- 14 tot, Bq	Accum. C-14 inorg, Bq	Accum. C-14 org, Bq	Org. frac- tion	Fract. of pro- duc- tion	Tot. std. dev.	Inorg. std. dev.	Org. std. dev.	Total Bq/MWhth	Inorg. Bq/MWhth	Org. Bq/MWhth	BFF
O3-1	O3 110412	2009-09-13	2011-03-10	8.22E+06	1.52E+11	6.79E+02	1.09E+09	1.07E+09	2.11E+07	1.9%	0.72%	10%	10%	30%	1.33E+02	1.30E+02	2.57E+00	1.16
O3-2	O3 110818	2010-01-11	2011-08-03	1.19E+07	2.19E+11	9.56E+02	2.66E+09	2.63E+09	2.49E+07	0.94%	1.2%	10%	10%	30%	2.24E+02	2.22E+02	2.09E+00	1.17
O3-3	O3 111031	2011-04-17	2011-10-19	6.72E+06	1.24E+11	7.52E+02	3.27E+09	3.22E+09	4.39E+07	1.4%	2.6%	10%	10%	38%	4.86E+02	4.79E+02	6.54E+00	1.13
O3-4	O3 120217	2011-09-08	2012-02-15	7.30E+06	1.35E+11	7.20E+02	3.59E+09	3.55E+09	3.79E+07	1.1%	2.7%	10%	10%	38%	4.92E+02	4.87E+02	5.19E+00	1.13
O3-5	O3 120717	2011-11-23	2012-07-10	7.37E+06	1.36E+11	6.30E+02	2.04E+09	2.02E+09	1.72E+07	0.84%	1.5%	10%	10%	21%	2.77E+02	2.74E+02	2.33E+00	1.15
O3-6	O3 121003	2012-02-29	2012-10-03	7.04E+06	1.30E+11	7.20E+02	3.04E+09	3.02E+09	2.20E+07	0.72%	2.3%	10%	10%	32%	4.32E+02	4.29E+02	3.13E+00	1.13
03-7	O3 130103	2012-07-26	2012-12-21	6.71E+06	1.24E+11	7.20E+02	3.30E+09	3.27E+09	3.11E+07	0.94%	2.7%	10%	10%	15%	4.93E+02	4.88E+02	4.63E+00	1.12
O3-8	O3 130306	2012-10-23	2013-03-04	6.40E+06	1.18E+11	7.20E+02	2.96E+09	2.91E+09	4.99E+07	1.69%	2.5%	11%	11%	12%	4.63E+02	4.55E+02	7.80E+00	1.12
O3-9	O3 130604	2013-01-09	2013-05-03	4.59E+06	8.48E+10	5.40E+02	1.87E+09	1.85E+09	2.11E+07	1.13%	2.2%	10%	10%	28%	4.06E+02	4.02E+02	4.60E+00	1.11
O3-10	O3 130827	2013-01-30	2013-08-25	1.08E+07	2.00E+11	1.08E+03	4.09E+09	4.04E+09	4.88E+07	1.19%	2.0%	10%	10%	22%	3.78E+02	3.73E+02	4.51E+00	1.12
03-12	O3 12 140114	2013-10-09	2014-01-08	4.98E+06	9.19E+10	6.30E+02	2.53E+09	2.51E+09	2.58E+07	1.01%	2.8%	10%	10%	34%	5.09E+02	5.03E+02	5.17E+00	1.09
03-13	O3 13 140221	2013-11-26	2014-02-19	3.51E+06	6.48E+10	4.50E+02	1.96E+09	1.91E+09	4.72E+07	2.42%	3.0%	10%	10%	42%	5.58E+02	5.45E+02	1.35E+01	1.09
03-14	O3 14 140419	2013-12-16	2014-04-11	4.76E+06	8.79E+10	6.30E+02	4.64E+09	4.54E+09	1.01E+08	2.19%	5.3%	10%	10%	21%	9.75E+02	9.54E+02	2.13E+01	1.10
O3-15	O3 15 140501	2014-03-10	2014-05-01	6.24E+05	1.15E+10	1.80E+02	4.80E+08	4.75E+08	5.61E+06	1.17%	4.2%	13%	13%	27%	7.70E+02	7.61E+02	9.00E+00	1.05
O3-16	O3 16 140829	2014-03-01	2014-08-29	8.05E+06	1.49E+11	9.10E+02	3.60E+09	3.51E+09	9.06E+07	2.50%	2.4%	11%	11%	24%	4.47E+02	4.36E+02	1.12E+01	1.09
Sum	O3 Sum 090913-14	0829		9.90E+07	1.83E+12	1.03E+04	4.11E+10	4.05E+10	5.88E+08	1.4%	2.3%	2.9%	2.9%	8.1%	4.15E+02	4.09E+02	5.95E+00	1.12

Table A13-2. Summary of calculated accumulation of C-14 in CCU resin from O3. $(O3-2, sheet Summary act)^{86}$ .

<sup>&</sup>lt;sup>86</sup> The sample O3-1 was obtained for CCU resins loaded in 2009 and back-flushed in 2011. Due to operational problems, the operational history of these resins is not typical for full power operation.



### A13.3. Discussion

Oxide		Fe3O4	Fe2O3	Mean						
Fe	atomic mass	56	56							
0	atomic mass	16	16							
Fe oxide	mole mass	232	160							
Fe oxide/Fe		1.381	1.429							
Ionic fraction		0.1	0.1							
(Fe+Fe oxide)/Fe on r	esin	1.34	1.39	1.36						
Concentration Fe in v	vater, ppb			10						
Body feed rate inert material O3* kg/d/filter 0.4										
* Only in February 201	L1-May 2011									

Table A13-3. Estimation of correlation factor to calculate accumulated mass of corrosion products from Fe concentration in the condensate. The mean value assumes equal amounts of  $Fe_3O_4$  and  $Fe_2O_3$ . (O3-2, sheet BodyfeedO3).

The Body-Feed Factor accounts for the uptake of corrosion products on the CCU resin since the tank content has been reported only as resin and inert material added<sup>87</sup>. The calculation of the uptake of corrosion products has been made according to the following model: From the accumulated water volume and the iron concentration the uptake of corrosion products has been calculated. An example is shown in *Table A13-6*.

The concentration of corrosion products has been assumed to be 10 ppb Fe, with 10 % in ionic form. The rest is assumed to be equally distributed between  $Fe_3O_4$  and  $Fe_2O_3$ . This yields a BFF in the range 1.05-1.17 with a mean value of 1.12 for the samples. The calculation of the correlation factor is shown in *Table A13-3*. The calculation of the amount of corrosion products is included in *Table A13-6*.

*Table A13-4* and *Figure A13-4* summarize the resulting accumulation, expressed as a percentage of the production and as Bq/MWh<sub>th</sub>, respectively. The values obtained for O3 are lower than the values obtained for its sister plant F3.

<sup>&</sup>lt;sup>87</sup> O3 used body-feed of inert material in the period 2011-02-01 to 2011-06-01. Pers. Comm. P. Arvidsson, OKG 2013-06-03.





Figure A13-4. Activity contents in the tanks sampled compared with results for F3. (03-2, sheet D Summary).

The organic fraction (and the accumulation rate, expressed as  $Bq/MWh_{th}$ ) is lower in this study than in *Ref.* 4. It is also lower than the values reported for F3 in *Appendix* 2. There is no obvious explanation for this.

The energy exposure of the resins amounts to 25380 EFPH (Equivalent Full Power Hours), or 2.90 EFPY (Equivalent Full Power Years).



																O3 Sum		Ref. 4 +
	03	03	03	03	03	03	03	03	03	03	O3 12	03 13	03 14	O3 15	O3 16	090913-	F3, Mass	BFF + ds-
	110412	110818	111031	120217	120717	121003	130103	130306	130604	130827	140114	140221	140419	140501	140829	140829	Balance	corr.*
Equivalent Full Power Hours (EFPH)	2109	3047	1723	1871	1891	1805	1720	1640	1178	2775	1276	899	1221	160	2065	25380	31800	
Equivalent Full Power Years (EFPY)	0.24	0.35	0.20	0.21	0.22	0.21	0.20	0.19	0.13	0.32	0.15	0.10	0.14	0.02	0.24	2.90	3.63	
Body-Feed Factor (BFF)	1.16	1.17	1.13	1.13	1.15	1.13	1.12	1.12	1.11	1.12	1.09	1.09	1.10	1.05	1.09	1.12	2.08	2.08
Dry Solids Correction Factor in Sar	nple																	1.16
Organic Fraction	1.9%	0.94%	1.4%	1.1%	0.84%	0.72%	0.94%	1.7%	1.1%	1.2%	1.0%	2.4%	2.2%	1.2%	2.5%	1.4%	1.7%	24%
Total accumulation in CCU	0.72%	1.2%	2.6%	2.7%	1.5%	2.3%	2.7%	2.5%	2.2%	2.0%	2.8%	3.0%	5.3%	4.2%	2.4%	2.3%	2.6%	1.7%
Organic accumulation in CCU	0.014%	0.011%	0.035%	0.028%	0.013%	0.017%	0.025%	0.042%	0.025%	0.024%	0.028%	0.073%	0.115%	0.049%	0.061%	0.032%	0.044%	0.40%
Resin kg dw	540	630	720	720	630	720	720	720	540	1080	630	450	630	180	910	9820		
Total Bq/MWhth	1.33E+02	2.24E+02	4.86E+02	4.92E+02	2.77E+02	4.32E+02	4.93E+02	4.63E+02	4.06E+02	3.78E+02	5.09E+02	5.58E+02	9.75E+02	7.70E+02	4.47E+02	4.15E+02	6.12E+02	3.11E+02
Inorganic Bq/MWhth	1.30E+02	2.22E+02	4.79E+02	4.87E+02	2.74E+02	4.29E+02	4.88E+02	4.55E+02	4.02E+02	3.73E+02	5.03E+02	5.45E+02	9.54E+02	7.61E+02	4.36E+02	4.09E+02	6.01E+02	2.37E+02
Organic Bq/MWhth	2.57E+00	2.09E+00	6.54E+00	5.19E+00	2.33E+00	3.13E+00	4.63E+00	7.80E+00	4.60E+00	4.51E+00	5.17E+00	1.35E+01	2.13E+01	9.00E+00	1.12E+01	5.95E+00	1.06E+01	7.43E+01
Tot. std. dev.	10%	10%	10%	10%	10%	10%	10%	11%	10%	10%	10%	10%	10%	13%	11%	2.9%	4.9%	
Inorg. std. dev.	10%	10%	10%	10%	10%	10%	10%	11%	10%	10%	10%	10%	10%	13%	11%	2.9%	4.9%	
Org. std. dev.	30%	30%	38%	38%	21%	32%	15%	12%	28%	22%	34%	42%	21%	27%	24%	8.1%	7.2%	

Table A13-4. Summary of accumulation of C-14 in CCU resins. Results from F3, Appendix 2, are included as a comparison. (O3-2, sheet Summary).



# Avfallet O3 - Provtagning C14 i jonbytarmassor (332)

<u> </u>	fylles	vid provt	tagnin	g (ke	mi)												
Provdatum:	-	2011-04-12															
Prov ID:		Prov 1															
Från tank 342	TR41									lı	nkonn	linasir	ntorval	•			
Värden för /Tei						00144			o no luti		) ionh	40700	inkon	ı. Aliantia	ערויי אירויי		
varuen for (Ter	mp) oc	ivivv u	i) ange	25 50	m eu	. sniu	sell 0	overre	speкu	ve 332	, jonby	lares	шкорр	Jiigstic	л руд	n.	
Start-datum	Slut- datum	332 O3 Filter	324 O3 Filter	Gå T€ T	332 O3 CB1-CB ngtid/Me emp/Meo erm.effe	7 edel del ekt	Mängd Jb- massa 324	Mängd jb-massa 332	Mängd inert- massa 332	342 Filter	Mängd jb-massa 342	Mängd inert massa 342	Extra dosering av Jb- massa tilll TB41	Typ av massor MB-400 Finex 875PM UP2 W	Summa 332 massor i 342 TB41	Summa 342 massori 342 TB41	Summa 324+332 + 342 massor i 342TB41
										CB11,							
ÅÅMMD	ÅÅMMDD	CB1-CB7	CC1	Dvan	°C	MWth	Κα	Ka	Κα	21, 71 CD12, 22	Κα	Κα	Κα		Ka	Ka	Ka
091218	100518	CB6	-	122	35	15253	0	90	0	0	0	0	0	F875	90	0	90
091017	100518	CB4	-	122	30	14924	0	90	0	0	0	0	0	F875	90	0	90
100622	101115	CB1	-	61	35	23093	0	90	29	0	0	0	0	F875	119	0	119
091004	110220	CB5	-	272	33	19630	0	90	0	0	0	0	0	F875	90	0	90
090913	110308	CB3	-	224	33	19800	0	90	55	0	0	0	0	F875	145	0	145
091219	110310	CB7	-	279	33	20132	0	90	55	0	0	0	0	F875	145	0	145
															0	0	0
														Summa	679	0	679
Prov-ID C-14	03-1	Komme	ntar fr. För	rfattare:													
pH Kandulati daat	5.53	Provflas	kan var stö	örre än d	e												
Mängd massa i prov	1.05	Vätsken	1 cm krop	p, 4 cm r cm	ials.												
Massa/vätska	13.64%			0111													

Table A13-5. Example of record-keeping for CCU powder resins. The data in the column "MWth" are not relevant for the calculations.(03-1, sheet 03 20110412).



	Avfa	llet	03 -	Pro	vtaq	ninc	a C14	4 i io	onbv	tarr	nas	sor	(332	2)			
	lfylles vi	d provta	agning (	kemi)			<b>j</b> • .	· · )`	J				(	-/			
Provda	tum:	2011-04-12															
Prov ID	•	Broy 1															
Erån ta	nk 312 T	D/1									Inkonr	lingein	torvall				
Fiall la	1K 342 1	D41	/ 8.41.4.1					1 - 1 -			iiikopp	ningsin 		11			
varden	for (Tem	p) ocn	(wwth)	anges s	som ett s	snitt se	tt over r	espekti	ve 332 j	onbyta	res inko	oppiigs	tia "Dy	gn≞.			
Start- datum	Slut-datum	332 O3 Filter	324 O3 Filter	Gångtio	332 O3 CB1-CB7 I/Medel Tem Term.effekt	p/Medel	Mängd Jb- massa 324	Mängd jb-massa 332	Mängd inert- massa 332	342 Filter	Mängd jb-massa 342	Mängd inert massa 342	Extra dosering av Jb- massa tilll TB41	Typ av massor MB-400 Finex 875PM UP2 W	Summa 332 massor i 342 TB41	Summa 342 massor i 342 TB41	Summa 324+332+ 342 massor i 342TB41
ÅÅMMDD	ÂÂMMDD	CB1-CB7	CC1	Dygn	°C	MWth	Kg	Kg	Kg	CB11, 21, 71 CD12, 22	Kg	Kg	Kg		Ka	Ka	Ka
2009-12-18	2010-05-18	CB6	-	122	35		0	90	0	0	0	0	0	F875	90	0	90
2009-10-17	2010-05-19	CB4	-	122	30		0	90	0	0	0	0	0	F875	90	0	90
2010-07-21	2010-11-15	CB1	-	61	35		0	90	29	0	0	0	0	F875	119	0	119
2010-05-18	2011-02-20	CB5	-	272	33		0	90	0	0	0	0	0	F875	90	0	90
2009-09-13	2011-03-08	CB3	-	224	33		0	90	55	0	0	0	0	F875	145	0	145
2009-12-19	2011-03-10	CB7	-	279	33		0	90	55	0	0	0	0	F875	145	0	145
						(	)	Summa	139	Conservat	ive!					0	0
2009-09-13	2011-03-10													Summa	679	0	679
														Summa	679	0	679
		Start, cum	End, cum	Filter,	Filter H2O,		BodyFeed		BodyFeed								
Start	Stopp	MWHt	MWHt	MWht	m3		days		kg					ID			03 110412
2009-12-18	2010-05-18	192279	5647780	7.79E+05	1.04E+06		0		0					Inert bod	y feed, kg		C
2009-10-17	2010-05-19	0	5723967	8.18E+05	9.56E+05		0		0					Corr. Pro	d., kg		130
2010-07-21	2010-11-15	8907237	11342143	3.48E+05	3.80E+05		0		0					Bff		NA71-7	1.16
2010-05-18	2011-02-20	4941064	16626202	2.205+06	1.52E+00		19		0					Total wa	energy, w		0.755+00
2009-09-13	2011-03-08	221375	16801635	2.36E+00	2.00E+00		35		14					TOLATWA	ler mass,	Ng	9.752+05
2005 12 15	2011 05 10	221373	10001055	0.00E+00	J.20L100		57		15								
				0.00E+00										x.	kg dw/kg (	condensate	6.96E-08
	Sum			8.22E+06	9.75E+06				36					,			
Stort BE	Slut BE																
2011-02-01	2011-06-01																
2011 02-01	2011 00-01																
Doshast.	0.4	kg/d/filter															

Table A13-6. Example of complete calculation for one sample/tank contents. The data in the column "MWth" are not relevant for the calculations.(O3-2, sheet O3 110412).



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# APPENDIX 14 Mass Balance for CCU Resins from F1 & F2

### A14.1. Introduction

All calculations for F1 and F2 are included in an excel-book. It can be found as Attachment *File F12-3* to this report. Reference to the various sheets will be made at tables, figures etc.



Figure A14-1. Overview of handling of spent powder resin at F1 and F2.<sup>88</sup>(A-5, Pict. 8)

In the Condensate Clean-up system (CCU) of F1 and F2 (10 filters per unit), 80 kg powder resin (carboxylic resin in cation part) + 10 kg of inert material is loaded per filter and operated for periods usually ranging from 6 to 16 weeks.

When the resin is exhausted, it is flushed to two temporary tanks in the unit, 342T42 and T43. These tanks are mixed by air bubbling. From these tanks, the resin is transferred in a shielded transport vessel (T80 with a stirrer) via tank 343T2 (with a stirrer) to tank TC56 (with a stirrer) in the Waste Building at F3. A schematic overview is given in *Figure A14-1*.

From the storage tank TC56 (continuously stirred) resin is brought to a dryer/evaporator, where the concentration of dry solids is increased to a proper value before solidification in bitumen. The dryer/evaporator also receives concentrate from the evaporation of waste waters, i. a. floor drains. In connection with this transfer, a sample is withdrawn. It is also possible to take a sample without a transfer for solidification. (This text is also included in *Appendix 1*).

### A14.2. Storage Tank Inventories and Mass Balance

#### A14.2.1. F1-342T42&T43 Original Data

The record describing the additions and removals of resin was delivered as an excel-book in *Attachment File F12-2* covering the time period from January 2007 to early January 2013. The excel-sheet contains dates for back-flushing of spent resin from each unit and the destination tank (T42 or T43), the amount of resin (kg dw), the inventory of each tank and

<sup>&</sup>lt;sup>88</sup> Christina Lillfors-Pintér. Report from visit to FKA 2011-11-30.



amounts removed to the F3 WHB for solidification. *Figure A14-2* shows the tank inventories together with the additions of resin at the back-flushings ("added") in the diagram.

The sums of the additions and removals into cumulative curves, together with the tank inventories, yield the diagram in *Figure A14-3*. A calculated mass balance, including the removals and the tank inventories is also included in the diagram. It can be seen that in the first half of the diagram, the curves for the cumulated additions and the mass balance closely follow each other. However, in July 2009, there is a sudden drop in the mass balance curve, caused by a corresponding drop in the inventory curve for T42. A close inspection of the Excel sheet revealed some comments when the tank inventories were zero. These comments are displayed in *Table A14-1*. After July 2009, there is an almost constant difference of some 10 000 kg between the cumulated additions and the mass balance.



Figure A14-2.Back-flushings of spent CCU resins ("added") and inventories in the storage tanks. Not updated with 2012 data. (F12-3, sheet D A14-2).

	A14.2.2.	F1-342T42&	T43 Data	Corrected	for Res	in Use
--	----------	------------	----------	-----------	---------	--------

	Invent	ory before b	being set to	zero.		
	Date	T42	T43	Comment	Commented by	Comment
				The inventory was set to	•	
T42 och T43 zero				zero by introducing negative	Christer	uttransport 11
contents	2007-05-07	2940	2340	values for the back-flushing.	Owenius	st kulor
						T43 tom och
				The inventory was 1590		tankspolad. Fel
				before being manually	Johan	diff 1590kg =ca
T43 zero contents	2009-04-02		1590	zeroed	Lundmark	6 kuler
T42 zero contents	2009-07-07	10395			No comment o	n difference

Table A14-1. Comments from the excel-sheet with tank inventory data for the data points with zero tank contents. (F12-3, sheet NOLLNINGAR).

To clarify this discrepancy, a compilation of the amounts of CCU powder resin being used in the years 2007-2011 was prepared in *Table A14-2*.





Figure A14-3. Inventories in the storage tanks for spent CCU resins, together with cumulated additions and removals. A mass balance curve is included together with a regression line for the cumulated additions of resin. Not updated with 2012 data. (F12-3, sheet D A14-3).

Resin use	2007	2008	2009	2010	2011
F1, kg/year		4305	4830	4410	
F2, kg/year		3780	4095	2835	
F1+F2, kg/year	6970	8085	8925	7245	6635

Table A14-2. Amounts of CCU resins used in the years 2007-2011. In the value for 2011 the value for December is missing. (F12-3, sheet supply).

A cumulation of the resin use as a function of time was compared with the regression line for the cumulated back-flushings; see *Figure A14-4*. The back-flushings are slightly higher than the cumulated resin use. However, there should also be a 10 % addition of inert filter material in the filters, so that curve will be slightly higher than the back-flushings. *Figure A14-4* strongly suggests that the zeroing of the contents in T42 in July 2009 is just a typing mistake. Removing this zero value, yields new curves in *Figure A14-5*. The balance line now closely follows the cumulated line for back-flushed resins.

#### A14.2.3. F1-342T42&T43 Corrected Data Aligned with F3-342TC56

Now, the next step is to compare the removals of resin from T42 and T43 with the additions to tank TC56 in the Waste Handling Building at F3. *Figure A14-6* shows the removals from T42 + T43 and the additions to TC56 according to the records. The figure shows all the recorded removals and additions, as well as the accumulated amounts. From the beginning of 2009 and until mid-June 2009 there is a reasonable agreement between removals and additions. After mid-June 2009, there is a growing gap between the accumulated curves.

In *Figure A14-7*, the accumulated removals from TC56 have been added. By the date 2010-05-20, the accumulated removals exceed the accumulated additions. This discrepancy persists as long as the record keeping lasts, i.e. until mid-November 2010.



Figure A14-4. Comparison of cumulated amounts of back-flushed CCU resins (from the regression curve in Figure A14-3) and the use of fresh CCU resins, with and without 10 % inert material added. (F12-3, sheet D A14-4).



Figure A14-5. Corrected inventories in the storage tanks for spent CCU resins, together with cumulated additions and removals. A mass balance curve is included together with a regression line for the cumulated additions of resin. (F12-3, sheet D A14-5).





*Figure A14-6. Removals fromTC42+T43 and additions to TC56 according to records. (F12-3, sheet D A14-9).* 



Figure A14-7. Removals fromTC42+T43, additions to and removals from TC56 according to records. (F12-3, sheet D A14-7).





Figure A14-8. Removals fromTC42+T43, additions to and removals from TC56 according to records. The additions have been corrected at 2010-04-01. (F12-3, sheet D A14-8).



Figure A14-9. Removals fromTC42+T43, additions to and removals from TC56 according to records. The additions have been corrected at 2010-04-01 and 2010-09-03 to bring them in agreement with the removals from T42+T43. (F12-3, sheet D A14-9).

Obviously, several batches of resin have not been recorded as added to TC56. *Figure A14-8* shows the accumulated contents of TC56 when 2860 kg dw (11\*260 kg dw) have been added as a correction on 2010-04-01. This correction brings the accumulated additions to TC56 in reasonable agreement with the accumulated removals from T42-T43 until September 2010. At this time, there is a new discrepancy between the removals from T42+T43. A further addition


of 1648 kg dw brings the two curves to coincide, *Figure A14-9*, which also includes the recorded level in TC56. Even if the level is not directly proportional to the contents of resin, there is a qualitative agreement with the additions and removals, making the corrections reasonable.

## A14.3. Discussion

The mass balance for the storage tanks T42+T43 in the Waste Handling Building of F1 seems to be reasonable, given that the zero value in July 2009 is disregarded. Each back-flushing is assumed to contain 105 kg dw, which is a rough estimate, based on 80 kg resin, 10 kg inert material. The rest is accumulated corrosion products. Cf. *Table A1-1*, where a value of 106 to 107 kg was calculated. Based on the recorded inventories when the tanks were emptied, the uncertainty is estimated to be in the range 10-20 %.

The tanks T42 and T43 were simultaneously emptied on 2007-05-07. This date sets the starting point for the accumulation of C-14 in the resins. The resin is located in T42 and T43 at F1 and in TC56 at F3. The only place where resin is sampled is in TC56 in connection with removals for solidification. The long hold-up times in the tanks T42 and T43 and TC56 (cf. *Figure A14-5* and *Figure A1-3*) means that it is not possible to use the sampling in TC56 to make any reliable calculations of the inventory of C-14 in T42 +T43.

*Figure A1-3* shows a strong variation in the concentrations of C-14 in the resins. With the sampling scheme used, it is impossible to decide whether the decrease in concentration has occurred in the filter during operation, in T42+T43 or in TC56.

Since T42+T43 are mixed by air bubbling, it seems reasonable that the decrease has occurred in these tanks. Indeed, there is one observation supporting the removal of C-14 from T42+T43: Airborne releases of C-14 during reactor shut-down<sup>89</sup>.

The additions to and the inventory in TC56 based on the records form the WHB at F3 seem to be underestimated. The calculations in *Appendix 1*, using the inventory of TC56 will include the corrections discussed above.

<sup>&</sup>lt;sup>89</sup> Christina Lillfors-Pintér. Report from visit to FKA 2011-11-30.



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# APPENDIX 15 Preliminary Tests to Determine Formic Acid in CCU and BTRS Resins

## A15.1. Introduction

A classical method to decompose formic acid into carbon monoxide and water is treatment with concentrated sulfuric acid. It has been suggested (*Ref. 16*) that treatment with hot 6 M sulfuric acid in presence of  $Fe_2O_3$  should decompose formic acid on the resins into carbon monoxide and water. The set-up used is shown in *Figure A15-1*.

The set-up differs from the ordinary extraction procedure, but the result will not be different. Summing the activity in bottles 4:1 and 4:2 will correspond to the activity in bottle 4.



Figure A15-1. Experimental set-up for the determination of formic acid in resins. The arrows show the flow direction. Bottle 1 contains 1.7M H<sub>2</sub>SO<sub>4</sub> and bottles 2-5 contain 2M NaOH. The manometer (not shown) is placed between the catalyst and the valve to the right of the catalyst. FM1 and FM2: Flow meters.

# A15.2. Extraction

1-1.2 g resin is put into the Erlenmeyer flask together with a stirring magnet (same as for normal wet extraction, described in *Appendix 9*). The pressure is lowered to 0.8 bar before nitrogen gas flow is started.

#### A15.2.1. Cold Acid Strip for 30 min

Bottles connected: 1, 2, 3, 4:1 and 5  $17 \text{ ml } 6M \text{ H}_2\text{SO}_4$  is added. The stirring is started and when a proper bubbling is obtained, the timer is started.

#### A15.2.2. Hot Acid Strip for 60 min

Bottles connected: 1, 4:1 and 5  $8 \text{ ml } 6M \text{ H}_2\text{SO}_4$  is added the heater is turned on at 250°C.

# A15.2.3. Wet Oxidations: three times 60 min

Bottles connected: 1, 4:2 and 5  $20 \text{ ml } \text{K}_2\text{S}_2\text{O}_8$ , followed by 2 ml 4% AgNO<sub>3</sub> and 3 ml water are added.

## A15.3. Liquid Scintillation Counting

The counting was performed as described in Appendix 9.

# A15.4. Results

The results from the tests are summarized in Table A15-1.

Sample		O3-3			O12-33			Prov 3*		BTRS**
	Conc.	Std.	Det.Lim.,	Conc.	Std.	Det.Lim.,	Conc.	Std.	Det.Lim.,	Conc.
Fractions	Bq/kg	Dev.	Bq/kg	Bq/kg	Dev.	Bq/kg	Bq/kg	Dev.	Bq/kg	Bq/kg
Inorganic	1.37E+06	2.0%	7.92E+02	1.87E+05	2.1%	7.92E+02	7.66E+05	2.0%	7.68E+02	2.10E+04
Formic Acid (FA)	1.37E+04	4.2%	7.95E+02	7.66E+03	6.4%	7.95E+02	1.31E+04	4.3%	7.70E+02	2.99E+03
Other Organics	1.69E+03	22%	7.95E+02	6.48E+02	58%	7.95E+02	1.13E+04	4.7%	7.70E+02	1.96E+05
Total organic	1.54E+04	4.5%		8.31E+03	7.5%		2.44E+04	3.1%		1.99E+05
Total C-14	1.39E+06	1.9%		1.95E+05	2.0%		7.90E+05	1.9%		2.20E+05
Total org. fract.	1.1%			4.3%			3.1%			90%
FA fraction	1.0%			3.9%			1.7%			1.4%
FA/tot.org.	89%			92%			54%			1.5%
* Sample from F1										
** Boron Thermal	3 and R4									

Table A15-1. Results from tests to determine formic acid in resins. (A-11, Sheet Hform).

The tests were performed on three BWR CCU samples and one sample from PWR BTRS. The BWR CCU samples gave a high fraction of formic acid, while the BTRS sample gave a very low fraction of formic acid.

A possible explanation for the low yield of formic acid may be the absence of iron oxides, since the BTRS resins have a position in the system where iron and iron oxides have already been separated. Another possible explanation may be that part of the C-14 in BTRS resins are present as elementary carbon; see *Figure A20-1*, right part.



# APPENDIX 16 Accumulation of C-14 in One CCU Filter: F1-332C1 Sample from Back-flushing Tank

One sample was obtained from the Back-flushing tank with the data shown in *Table A16-1*. This sample was intended as a comparison sample, since it was collected in a similar position as the samples analyzed in *Ref. 4*.

The "Filter Factor" accounts for the number of CCU filters in parallel. The accumulation has been assumed to be uniformly distributed between the filters. The accumulation amounts to 7.7 % of the production, which is the highest recorded value for any CCU resin sample.

Batch	Start Date	End date	Total, Bq/kg dw	Inorg, Bq/kg dw	Org, Bq/kg dw	Org, %	SD tot, %	SD inorg, %	SD org, %	x, kg CCU resin/EFPY	x <sub>a</sub> CCU anion eq/EFPY
FKAX-17	2010-04-16	2010-07-07	1.16E+07	1.15E+07	8.14E+04	0.71%	4.5%	4.6%	24%	0.41	0.61
F1-332C1											
Filter Factor		10.00	Resin Unce	ertainty	10%						
Produc-tion	Start	End	MWhth	Calc. Produc- tion Bq total	Total resin mass, kg	Total Bq	Inorg, Bq	Org Bq	% of produc- tion	Total Bq/MWhth	Inorg Bq/MWhth
Mean/ Sum	2010-04-01	2010-06-30	6.39E+06	1.57E+11	105	1.21E+10	1.20E+10	8.55E+07	7.7%	1.90E+03	1.89E+03
% organic								0.70%			
% standard dev	viation					11%	11%	26%			

Table A16-1. Concentrations and accumulation on one single CCU filter, F1-332C1 16 April—7 July, 2010. "Filter Factor" accounts for the number of filters in parallel in the unit. (F12-1, sheet F1Backspolntank).



Figure A16-1. Concentrations in resin samples from F12. (F3-1 sheet D conc sum F12).



*Figure A16-1* shows the concentration of all resin samples from F12. The concentration in the sample from the Backflushing tank is 1.16E7 Bq/kg dw, which should be compared with the highest value recorded in TC56 (actually in the temporary storage tank TC58, coming from F1 WHB), 4.40E6 Bq/kg dw. From this sample and the corrected values obtained in *Ref. 4*, 5.54E6 Bq/kg dw, for a sample from the same tank, it is obvious that C-14 is removed from the resin in the storage tanks T42 and T43, or in the transfer process to TC56 and in TC56 itself.



# APPENDIX 17 Correction of Organic C-14 by Sample Combustion

The Combustion correction factor (see also Appendix 10) is defined by

 $f = \frac{a_{\textit{nettocombustion}} + a_{\textit{nettoorganic}}}{a_{\textit{nettoorganic}}}$ 

where *a* is organic activity (Bq) in the sample from the extraction and the combustion experiments, respectively. The net activity is obtained by subtracting the background from the gross value. It is assumed that the activity collected from combustion is organic<sup>90</sup>. The complete data from the combustion experiments can be found in *Attachment file A-10*.

Model 1: If the net value for combustion is negative, it is set = 0 and the factor f is calculated. This means that there is no correction for combustion. This is most likely an underestimation of the correction factor.

Model 2: The detection limit (DL) value for a bottle is about 0.9 Bq. Substituting all values below the DL/2 by DL/4 in the formula yields a likely value for the Combustion correction factor. The standard deviation is calculated from the standard deviations in the results from the extraction and combustion experiments, respectively.

	Net		Net										
	activity in		activity in	Sigma		Det.	Selecte	Selec-					
	bottle	Sigma	bottle	com-		Lim.	d	ted		Model	Model		
	from	extrac-	from	bus-		(DL)	value,	value,	Model	1:	2:	Model	
	extrac-	tion,	combus-	tion,	Net	bottle,	Extrac-	Combus	2 minus	Comb./	Comb./	2 plus	Std.dev
Sample	tion, Bq	Bq	tion, Bq	Bq	sum, Bq	Bq	tion, Bq	tion, Bq	1 SD	Extr.	Extr.	1 SD	, (SD)
C_F3_332_10B2_1 101103	26	0.91	0.12	0.53	26	1.0	26	0.25	0.956	1.000	1.009	1.063	0.053
C_F3-332-10B1_1 101102	15	0.71	-0.04	0.57	15	1.1	15	0.26	0.941	1.000	1.017	1.094	0.077
C-F12-332-09B2	3.1	0.51	-1.35	0.53	3	1.0	3	0.25	0.770	1.000	1.080	1.389	0.310
C_F12_332_09B3_1 101102	3.8	0.52	-0.34	0.54	4	1.0	4	0.25	0.810	1.000	1.068	1.325	0.258
C-2-O12-10:3	2.3	0.44	-0.26	0.57	2	0.9	2	0.24	0.695	1.000	1.104	1.513	0.409
C-2-O12-19:3	36	1.04	-0.33	0.57	36	0.9	36	0.24	0.962	1.000	1.007	1.051	0.044
C-2-O3-1:3	5.0	0.50	0.17	0.57	5	0.9	5	0.24	0.856	1.000	1.047	1.238	0.191
C-2-O3-2:1	7.4	0.54	-0.23	0.57	7	0.9	7	0.24	0.899	1.000	1.032	1.164	0.132
C-2-R1-2010-B6:3	4.8	0.57	0.22	0.57	5	0.9	5	0.24	0.833	1.000	1.049	1.265	0.216
C-2-R1-2010-B7:3	11	0.90	-0.03	0.57	11	0.9	11	0.24	0.889	1.000	1.022	1.155	0.133
C_R2_334_08_5 101101	4304	84	97	1.07	4400	2.5	4304	97	0.995	1.022	1.022	1.050	0.028
C-2-R2-2011-4	4518	88	144	3.15	4663	0.9	4518	144	1.004	1.032	1.032	1.060	0.028
C_R3_334_08_1	4407	86	55	1.47	4462	1.0	4407	55	0.985	1.013	1.013	1.040	0.028
C_R3_334_09_101028	3140	61	107	2.49	3246	1.0	3140	107	1.006	1.034	1.034	1.062	0.028
C_R4_334_08_5 101101	6426	125	107	2.95	6533	1.2	6426	107	0.989	1.017	1.017	1.044	0.028
C_R4_334_09_1 101026	6944	134	68	1.76	7012	1.0	6944	68	0.982	1.010	1.010	1.037	0.028
C-2-R4-2011-2	6857	133	129	2.85	6985	0.9	6857	129	0.991	1.019	1.019	1.046	0.028
Mean all samples									1.01	1.01	1.03	1.06	0.03
Mean BWR samples									1.01	1.00	1.04	1.07	0.03
Mean PWR samples									1.01	1.02	1.02	1.03	0.01
Max BWR									1.10	1.00	1.10	1.10	
Max PWR									1.03	1.03	1.03	1.03	
Min BWR									1.01	1.00	1.01	1.01	
Min PWR									1.01	1.01	1.01	1.01	

*Table A17-1. Summary of the combustion experiments and correction factors. (A-10, sheet COMB).* 

The table shows that the PWR samples (R2, R3 and R4) have detected values for the combustion experiment, the lowest values of f and the lowest standard deviation. The PWR

<sup>&</sup>lt;sup>90</sup> Some inorganic anions may stabilize anion resins at elevated temperatures (up to 150 °C). B Torstenfeldt, SKB. Pers. Comm. May 2013.



mean value ranges between 1.01 and 1.03 with a mean value of 1.02 and a standard deviation of 0.01.

For the BWR samples, Model 2 values ranges between 1.01 and 1.10 with a mean value of 1.04 and a standard deviation of 0.03. Since the activities involved in BWR samples are less than for PWR samples, the standard deviation is higher for the BWR samples.



Figure A17-1. Graphical summary of the combustion experiments and correction factors. (A-10, sheet D COMB).

Since the BWR samples are powder resins, they have an appreciably larger surface area than the bead resins used in PWR. All BWR powder resin samples used in the combustion experiments experienced a weight loss of about 50 % in the extraction experiments<sup>91</sup>. Therefore, it is likely that the BWR resins have released the organic activity in the acid, oxidizing extraction at least to the same extent as the PWR resins do.

Consequently, a value of 1.05 for the Combustion correction factor has been chosen for all resin types<sup>92</sup>, which is in agreement with *Ref. 4*; see also *Table A17-2*.

		Std.	Number of
Sample types	Mean	Dev.	samples
Mean all samples	1.03	0.03	17
Mean BWR samples	1.04	0.03	10
Mean PWR samples	1.02	0.01	7
Selected value	1.05		

Table A17-2.	Summary of combustion	correction factors.
	(A-10, sheet COMB)	).

<sup>&</sup>lt;sup>91</sup> The PWR bead resins lost almost no weight.

<sup>&</sup>lt;sup>92</sup> FKA dried resin samples are reported in Appendix 21.



# APPENDIX 18 Releases of C-14 from Spent Resins in Storage Tanks and Resin Dryers

## A18.1. Introduction

The following question has been asked: Are the samples taken from the spent resins representative for what goes into the waste containers?

Appendix 11 gives an overview of the handling and sampling of the resins prior to solidification or packaging.

At O1 and O2 the sampling is done in the well mixed collection tank immediately before the resin is transferred to concrete tanks where they are dewatered.

At O3 the sampling is done in the well mixed collection tank immediately before the resin is transferred to concrete tanks where they are dewatered.

At R1 the sample is taken from the well mixed storage tank at regular intervals, but not necessarily in connection with concrete solidifications.

For the PWR units R2, R3 and R4 the sample is taken from the transport vessel before it is emptied into the storage tank in the WHB. In this tank the resin is regularly (frequently) mixed by air bubbling before it is taken to concrete solidification.

For F1, F2 and F3 the sample is usually taken when the resin slurry is sent to the resin dryer. From the dryer it is sent to high temperature solidification in bitumen.

# A18.2. R2, R3 and R4 Resins in the WHB of R1

*Figure A18-1* shows the release rate of C-14 from the stack at R1. All parts of the unit, including the WHB, are vented through this stack. It is obvious that the release rate decreases when the unit is shut down. Since R1 only has a very simple delay system for the ejector off-gases (sand tanks with together some 250 m<sup>3</sup> void volume), it is unlikely that there are any significant traps for C-14 during shutdown. Since the accumulation in RWCU and SFP resins is very low in BWR units (*Ref. 4*), the releases of C-14 during shut-down most likely originate from spent resins in the WHB.

*Figure A18-2* zooms in on the period with a long shut-down, March 2009 - March 2010. Fig A18-3 shows the measured release rate during shut-downs in R1, where all release data with rector power>0 % have been removed. The diagram also has lines for average release rate during shut-down for the years 2002-2013 and for the long shutdown in 2009-2010. This latter shut-down is of extra interest, since there were very little or no CCU resins in the storage tanks; see *Figure A18-4*. Even if there has been added resin to the tank, the resin has not been exposed to power operation.





Figure A18-1. Airborne release rate of C-14 from R1 and reactor power (24 hours mean). Ox means inorganic and red means organic. (R1-5 sheet D ut orig).



*Figure A18-2. Airborne release rate of C-14 from R1and reactor power (24 hours mean) in 2008-2013. ). Ox means inorganic and red means organic. (R1-5 sheet D ut 2009-2013).* 





Figure A18-3. Airborne release rate of C-14 from at shut-down in 2002-2013. ). Ox means inorganic and red means organic. (R1-5 sheet D ut SD).



Figure A18-4. Spent CCU resin in the storage tank T41 in R1 WHB. (R1-5 sheet D T41A).



Unit	Production rate, Bq/MWht	Production 2002-2012, MWht	Production 2002-2012, Bq	Average production rate, Bq/s	Unit	Release rate during shut- down	Inorg. C-14, Bq/s	Organic C-14, Bq/s	Total C-14, Bq/s	Orga- nic fraction
R1	2.35E+04	1.72E+08	4.05E+12	1.17E+04		Mean 2002-2013	3.81E+02	6.69E+01	4.48E+02	15%
R2	1.49E+04	1.82E+08	2.72E+12	7.85E+03		2009-2010	2.03E+02	3.28E+01	2.36E+02	14%
R3	1.59E+04	2.42E+08	3.84E+12	1.11E+04			Fraction	of produc	tion rate	
R4	1.43E+04	2.28E+08	3.25E+12	9.37E+03			Inorg.	Org.	Total	
R1-R4	6.86E+04		1.39E+13	3.99E+04	R1-R4	Mean 2002-2010	0.95%	0.17%	1.1%	15%
R2-R4	4.51E+04		9.82E+12	2.83E+04	R2-R4	2009-2010	0.72%	0.12%	0.83%	14%

Table A18-1. Summary of average production rates of C-14 in the Ringhals units 2002-2012(left section) and release rate of C-14 from R1 during shut-down periods (right section).(R1-5 sheet Rel rate).

*Table A18-1* shows the mean values for the release rate of C-14 (during shut-down of R1) for the whole period 2002-2013 and for 2009-2010, respectively. These release rates have been compared with the production rates in the units R1-R4 (left part of the table).

For the whole period, where resins from all four units are expected to contribute, the release rate during shut-down corresponds to 1.1 % of the Average production rate in the period. Applying the release rate in 2009-2010 on the PWR units yields a release rate of 0.83 % of the production rate. The organic fraction in the releases is some 15 %, which is higher than during normal operation (1-5 %). These data indicate that there is a certain release of C-14 from the storage tanks for spent resins. This also implies that the calculated amounts of C-14 in the waste containers (4.1 % of the production) may be overestimated by some 20-30 %.

## A18.3. O1 and O2

The WHB at O1 handles spent resins from O1 + O2. The ventilation air from the storage tanks for spent resin is released through the stack in O1<sup>93</sup>. When O1 is shut down, the airborne releases of C-14 must originate from the storage tanks for spent resins, analogous with R1.

O1 also has a simple delay system for the ejector off-gases, which is very unlikely to accumulate significant amounts of C-14. *Figure A18-5* shows the release rate of C-14 from O1 in the years 2002-April 2013. The release rate is shown as inorganic (oxidized, CO<sub>2</sub>) and reduced (organic, CH) as well as the sum (tot). In addition, the reactor power is also displayed. There are two longer periods when O1 was shut down; 2002 and Nov. 2011-Apr 2013. However, the stack sampling for C-14 was intermittent in 2002, so that period has been excluded. It has been assumed that all releases originate from O2 resins during shutdown of O1.

*Figure A18-6* shows the release rate and reactor power for the period July 2011-June 2013. It can be seen that here are short periods where O1 operated.

*Table A18-2* summarizes the release rate from November 2011- mid-April 2013. The first line also includes some periods with reactor operation. The second line contains only samplings with the reactor in the no-operational state. The third line contains the difference between the two first lines, giving the contribution from reactor operation. It has been compared with the

<sup>&</sup>lt;sup>93</sup> Karl-Erik Ingemansson OKG, personal communication, June 2013.



calculated production, shown in *Table A18-3*. There is a good agreement between these two values for the reactor production.



*Figure A18-5. Airborne release rate of C-14 from O1and reactor power (24 hours mean) in 2002-2013. CO<sub>2</sub> means inorganic and CH means organic. (O12-6 sheet D O1).* 



Figure A18-6. Airborne release rate of C-14 from O1and reactor power (24 hours mean) in November 2011-April 2013. CO<sub>2</sub> means inorganic and CH means organic. (O12-6 sheet D O1 SD).



		C-14 Release			C-14 ca	lc. Prod	No-op. release,		
		Inorganic	Organic	Total			Fraction of O2		
Start	End	Bq	Bq	Bq	01, Bq	02, Bq	production		
2011-11-01	2013-04-15	9.93E+09	1.23E+10	2.23E+10				Incl. O1 re	actor op.
2011-11-01	2013-04-02	8.77E+09	1.20E+10	2.08E+10				O1 no ope	eration
2011-11-01	2013-04-15	1.17E+09	3.40E+08	1.51E+09	2.53E+09	4.41E+11		From read	tor operat.
2011-11-01	<b>11-11-01 2013-04-15</b> 1.98E+10			1.98E+10			4.5%	Release -	alc. O1 prod.
Accumulati	fter release	e to air, fro	m Table 6			0.49%			
Fraction released of originally accumulated C-14							90%		

Table A18-2. Summary of measured airborne releases from O1 of C-14 in the period November 2011-April 2013. The releases as a fraction of the calculated production in O2 has also been calculated. CO<sub>2</sub> means inorganic and CH means organic. (O12-6 sheet Summary).

	Power pr MW	oduction, /hth	
	01	02	
2011-11-012011-12-31	0	2.36E+06	Estim. from op. data
2012	1.01E+05	1.55E+07	From monthly report
2013-01-012013-04-15	6.27E+03	4.22E+06	Estim. from op. data
Sum	1.07E+05	2.21E+07	
EFPY	0.009	1.40	
Prod. Rate, Bq/MWhth	2.36E+04	2.00E+04	
Production, Bq	2.53E+09	4.41E+11	

Table A18-3. Energy production in O1 and O2 and calculated production of C-14 in November 2011- mid-April 2013. (O12-6 sheet Summary).

The release while the reactor is shut down must originate from the resins produced in O2. The amount of C-14 released is calculated as a fraction of the calculated production in O2 and amounts to 4.5 %. The C-14 in the resins packed in concrete tanks for storage is on the average 0.49 % (*Appendix 3*). It means that the total uptake in the CCU resins is about 5 % of the production. However, it seems as about 90 % of this uptake is released during the storage and air bubbling in the WHB.

## A18.4. Assessment

#### A18.4.1. 01, 02 and 03

For O1, O2 and O3 the samples are representative for what goes into the concrete tanks, since the release from the storage tanks occurs before the sampling. The releases at shut-down also strongly indicates that C-14 is released from the stored resin in the tanks; most probably in connection with air bubbling.

#### A18.4.2. R1

For R1 the samples are also representative, despite the observed release. The representativeness is due to the frequent sampling of the resins in the storage tank. It is unlikely that concrete solidification should give a volatilization of carbon dioxide or organic acids at the high pH in the concrete slurry, even if it reaches a temperature of 120-130 °C during the hardening of the concrete.



#### A18.4.3. R2, R3 and R4

For the resins from the PWR units R2, R3 and R4 the observed release occurs after the sampling. It is likely that the solidified resins contain some 20-25 % less C-14 than the calculations from the samples indicate.

#### A18.4.4. F1, F2 and F3

Measurements in a ventilation duct from the WHB indicate a significant release of C-14 (*Ref.* 8) during the drying and the solidification in bitumen. These measurements and analyses of dried resin samples are discussed in *Appendix 21*.

For F1, F2 and F3 the major fraction is volatilized during the drying; see *Appendix 21* for details. Therefore, the values for wet resins are not representative for the solidified resins.



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## APPENDIX 19 Barsebäck CCU Accumulation: A Recalculation

## A19.1. Introduction

The reasons for recalculating the accumulation of C-14 in CCU are the following:

- Calculating a grand mean from all samples available
- Include standard deviations from the samples and the resin amount
- Include Body-Feed Factor  $(1.20)^{94}$  and dry substance correction factor (ds, 1.16, *Appendix 6*) in the calculations.
- The method differs slightly from the methods used in *Ref. 3* and 4

## A19.2. Calculations

*Table A19-1* shows input data from *Ref. 3* and 4 with the corrections mentioned above. Furthermore, a grand mean of all concentrations has been calculated from all four samples analyzed. The resulting standard deviations are the maximum of the standard deviations in the analyses and the scatter between the samples.

								corr.					Dry							
	Inorg		Det.lim.	Organic		Det.lim.	Organic	Organic,			Total	Total	substan-	Total,	Inorg	Org				
	Bq/kg	Sigma	Bq/kg	Bq/kg	Sigma	Bq/kg	corr.	Bq/kg	Dry sub-	Org.	Bq/kg	Bq/kg	ce cor-	Bq/kg	Bq/kg	Bq/kg	Org.	SD Tot,	SD Inorg,	SD Org,
Sample	ww	rel	ww	ww	rel	ww	Factor	ww	stance	fraction	ww	dw	rection	dw	dw	dw	fraction	rel	rel	rel
Ref.	3	3	3	3	3	3	4	3+4	4	calc	3+4	3+4	App. 6	calc	calc	calc	calc	calc	calc	calc
B-1:1	1.00E+05	4.0%	4.14E+02	1.29E+03	19%	4.68E+02	1.36	1.75E+03	37%	1.3%	1.02E+05	2.75E+05	1.16	3.19E+05	3.15E+05	4.04E+03	1.3%	4.6%	4.0%	19%
B-1:2	9.30E+04	3.9%	5.08E+02	1.19E+04	5.9%	6.64E+02	1.36	1.62E+04	37%	11%	1.09E+05	2.95E+05	1.16	3.42E+05	3.05E+05	3.73E+04	11%	4.1%	3.9%	5.9%
B-2:1	1.42E+05	4.0%	5.76E+02	1.47E+04	5.7%	7.54E+02	1.33	1.96E+04	47%	9.1%	1.62E+05	3.44E+05	1.16	3.99E+05	3.62E+05	3.63E+04	9.1%	4.3%	4.0%	5.7%
B-2:2	7.84E+04	4.1%	6.45E+02	1.42E+04	7.4%	8.44E+02	1.33	1.89E+04	47%	15%	9.73E+04	2.07E+05	1.16	2.40E+05	2.05E+05	3.50E+04	15%	4.3%	4.1%	7.4%
Mean											1.17E+05	2.80E+05		3.25E+05	2.97E+05	2.82E+04	8.7%	18%	19%	50%

Table A19-1. Recalculation of the analyses of the Barsebäck CCU samples<sup>95</sup>. (B-2, sheet Conc).

<sup>&</sup>lt;sup>94</sup> Same as originally calculated for O1+O2, (1.07), but later increased to 1.20 to account for inert masses added. See *Table A28-1*.

<sup>&</sup>lt;sup>95</sup> The samples were collected from dewatered resins in concrete tanks.



	B1/GWh-el, net	B1/GWh-th	B2/GWh-el, net	B2/GWh-th	C-14 production rate, Bq/MWhth (=O2)	2.00E+04			
1975	1540	4813			Waste type B.07	Anal	vsis	From x <sub>2</sub> -c	orrelation
1976	3545	11078					Ctd Day	- d -	Std Day
1977	2700	8438	2625	8203			Siu. Dev.,		Siu. Dev.,
1978	3908	12213	3836	11988	B1+B2	Value	%	Value	%
1979	2250	7031	3929	12278	Total Bq/kg dw	3.25E+05	18%	1.60E+06	9.0%
1980	3551	11097	3377	10553	Inorganic Bq/kg dw	2.97E+05	19%	1.57E+06	12%
1981	4133	12916	3803	11884	Organic Bg/kg dw	2.82E+04	50%	3.29E+04	21%
1982	3957	12366	4606	14394	Organic fraction	8 7%		2 1%	
1983	3988	12463	3/19	11622	Dedu Food Foster DFF	1.20		2.1/0	
1984	4161	13003	4021	12566	Body-reed Factor, BFF	1.20			
1985	4099	12809	4306	13456	Annual resin use per unit, kg dw	5190	10%		
1986	4372	13663	4129	12903	Annual average production, MWhth	1.17E+07			
1987	4556	14238	4448	13900	Calculated C-14 production, Bq/year	2.34E+11			
1988	4393	13720	4393	13/20					
1989	4328	13020	4200	13144	Total accumulation Bo/year	2 03E+09	20%	9 98F+09	13%
1990	4310	13409	4210	13130		1.055.00	20/0	0.705.00	1.0/
1991	2067	8050	2642	8250	inorganic accumulation, Bdyyear	1.85E+09	22%	9.78E+09	10%
1992	2007	0353	2043	8024	Organic accumulation, Bq/year	1.76E+08	51%	2.05E+08	23%
1004	4270	12656	2000	11702					
1994	4370	12206	2751	11703					
1006	4142	12044	2772	11788					
1990	2721	11650	2001	12101					
1997	4322	13506	4010	12531					
1999	2641	8253	3482	10881					
2000	2041	0200	3026	9457					
2000			4577	14302					
2002			3874	12106					
2002			2279	7122					
2003			4692	14663					
2005			1899	5934					
Sum	93416	291925	108732	339787					
Net. Efficiency	0.3200		0.3200						
Mean production	3737	11677	3749	11717					
Grand Mean, B1+B2	3743	11698							

Table A19-2 (left). Summary of energy production in the Barsebäck Units. (B-1, sheet BKAB energy). Table A19-3 (right). Calculation of the annual accumulation in CCU in one unit at Barsebäck. (B-2, sheet Summary).



## A19.3.Sampling and Energy Production

The samples were collected from already filled concrete tanks. It was not possible to relate the resins to a specific operating period. Therefore, the concentrations were used to calculate the annual accumulation from the annual use of dry CCU resin. To recalculate the accumulation to % of production and Bq/MWh<sub>th</sub>, the following procedure was used: *Table A19-2* shows a summary of the energy production from the Barsebäck units. The average annual production has been calculated for each unit and for both units. The values obtained are almost identical.

## A19.4. Calculating the Accumulation from the $x_a$ -Correlation

Since there were very few samples collected, an alternate method to estimate the accumulation is to use the  $x_a$ -correlation described in *Appendix* 22, using Eq. (A22-1). The calculations are shown in *Appendix* 28. The correlation assumes that the spent CCU sulfonate resins have not been air bubbled<sup>96</sup>. The total accumulation has been calculated from the  $x_a$ -correlation. The organic part has been calculated, using the organic fraction obtained in the analyses. This is considered conservative, since a low total concentration often has a higher *organic fraction* than a high total concentration. The results from this correlation are included in *Table A19-4*.

## A19.5. Summary and Discussion

*Table A19-3* shows the calculation of the accumulation expressed as % of production and as Bq/MWh<sub>th</sub>. The results are summarized in *Table A19-4*, together with the results from O1+O2 and the previously calculated values from *Ref. 4*. A 10 % standard deviation for the resin amount has also been included.

*Table A19-4* summarizes the accumulation of C-14 in the CCU resins. It also contains the values for O1+O2 and original and corrected values from *Ref. 4*.

The recalculated total accumulation is about 50 % higher than the corrected value from *Ref. 4*. On the other hand, the organic accumulation is almost the same. The reasons for these differences are the correction factors (BFF and ds) and how the mean values are calculated. The accumulation of inorganic and organic is higher than in O1+O2, which is reasonable, since Barsebäck did not air-bubbling the storage tanks. *Ref. 4* did not include standard deviations in the calculated accumulation. The large standard deviations in the B1+B2 values are mainly due to the

<sup>&</sup>lt;sup>96</sup> Lars Wiklund, BKAB, via Lars Håkansson, BKAB. Pers. Comm. 2012-04-12.



scatter in the few concentration values measured. The lower uncertainties in the O1+O2 values are mainly due to the larger number of samples analyzed.

The accumulation from  $x_a$ -correlation is higher than obtained from the analyses. One explanation for this may be that the sampling was performed from the top of CCU resins filled in a concrete tank, where some C-14 may have been volatilized. The  $x_a$ -correlation may be considered as an upper limit of the accumulation.

	<b>₽1</b> ±₽2	B1+B2	B2, Ref. 4		
	analyses	lation	+ ds-corr.	01+02	B2. Ref. 4
x <sub>a</sub> -value (average B1+B2 1992-2005)		0.20			,
Organic fraction	8.7%	2.1%	16%	5.2%	16%
Total accumulation in CCU	0.87%	4.3%	0.50%	0.49%	0.40%
Inorganic accumulation in CCU	0.79%	4.2%	0.42%	0.47%	0.34%
Organic accumulation in CCU	0.075%	0.088%	0.080%	0.026%	0.064%
Total Bq/MWh <sub>th</sub>	1.73E+02	8.54E+02	1.06E+02	1.03E+02	8.53E+01
Inorganic Bq/MWh <sub>th</sub>	1.58E+02	8.36E+02	8.93E+01	9.74E+01	7.16E+01
Organic Bq/MWh <sub>th</sub>	1.50E+01	1.76E+01	1.70E+01	5.29E+00	1.36E+01
Total Std. Dev.	20%	13%		2.3%	
Inorganic Std. Dev.	22%	16%		2.3%	
Organic Std. Dev.	51%	23%		9.5%	

Table A19-4. Summary of the accumulation of C-14 in Barsebäck 1 and Barsebäck 2. (B-2, sheet Summary).



# APPENDIX 20 Which Chemical Species of C-14 Could Be Expected in the Systems?

## A20.1. Introduction

One way to demonstrate the stability of chemical species is to calculate potential-pH diagrams from thermodynamic data. *Figure A20-1* shows two potential-pH diagrams, (also called Pourbaix diagrams) for the system carbon-water at 300 °C and 25 °C. The border line between two species indicates equal chemical activity of the species; for dissolved species, the chemical activity usually is close to its concentration.



Figure A20-1. Potential-pH diagrams (Pourbaix diagrams) for carbon compounds in water. The rectangular areas indicated represent the conditions in various parts of a PWR plant. Left: RC during operation at 300 °C. Right: RWCU during operation and RC, RWCU, BTRS and SFP during shut-down at 25 °C. See footnote below for actual temperatures. From Yim and Caron, Ref. 11.

# A20.2. PWR

#### A20.2.1. Power Operation

The Pourbaix diagrams in *Figure A20-1* are <u>approximating the temperatures<sup>97</sup></u> in PWR RC during operation (left part), in RWCU during operation and in RC, RWCU and SFP during shut-down (right part). The diagrams come from Yim and Caron (*Ref. 11*). The diagrams have rectangles indicating the combinations of redox potential and pH for the different operating conditions<sup>98</sup>. *Ref. 12-15* have similar information as *Ref. 11*.

<sup>&</sup>lt;sup>97</sup> RC operating temp.: 290-330 °C. RWCU operating temp.: 45 °C. SFP temp: 25-45 °C, depending on cooling water temp. and residual power in the stored fuel. RC, RWCU and SFP shutdown temp.: 25-45 °C.

<sup>&</sup>lt;sup>98</sup>The following calculation programs have been used for calculation of pH and redox-potential: BORIS (pH only), EPRI ChemWorks and SGW4Studsvik. Input data have been obtained from the Ringhals Chemistry Data Base.

*Left diagram Rectangle 1*: In RC during operation, the stable species are methane, methanol and formate ion. During operation, the pH is close to 7.40 in the Ringhals PWRs. During start-up and shut-down it may be lower, but usually not below 6.50. Accordingly, formate ion is stable for most of the cycle.



Figure A20-2. Li-concentration, pH-values in RC (300 °C) and RWCU (45 °C) as a function of boron concentration. The pH at 25 °C is included for a comparison with the Pourbaix diagram in Figure A20-1. Target pH in RC: 7.40. (R234-2, sheet D Sum).



Figure A20-3. Organic acids in the Reactor Coolant of Ringhals 4. The detection limits are: oxalate 0.8; formate 1.1 and acetate 1.2 ppb. (R234-3, sheet D R3).

*Right diagram, Rectangle 4*: In RWCU during operation the stable species is methane. In the beginning of the fuel cycle, the pH-value is around 6.50. As the concentration of boric acid decreases during the fuel cycle, the pH is increasing. In the end of a fuel cycle with coast-



down operation, the pH may approach 10; see *Figure A20-2*. In the final part of the fuel cycle, elemental carbon and hydrogen carbonate may also exist.

#### A20.2.2. Shutdown Conditions

At shutdown, with the reactor system open to the atmosphere and connected to the Spent Fuel Pools, (SFP), the chemical conditions correspond to the Rectangle 5 in the right diagram. Under these conditions, inorganic species of C-14 are dominating. Since "carbonic acid" decomposes into carbon dioxide and water, inorganic C-14 will be released to the atmosphere.

#### A20.2.3. Comparison with Analytical Data

*Figure A20-3* shows that in the reactor coolant of R4 the anions of simple organic acids have been found; formate, acetate and oxalate. Since the RC water sampled has been cooled to the same temperature as the RWCU, it is likely that the organic anions also exist in the RWCU<sup>99</sup>. Attachment *File R234-3* has data from all three PWR units showing the presence of the simple organic anions.



the Reactor Coolant (RC). (From Ref. 4).

In the last part of the previous cycle, (June and July) there is appreciably more inorganic species, which is consistent with the rightmost part of the Rectangle 4 in *Figure A20-1*.

*Figure A20-4* shows the results from analyses of C-14 the RC in Ringhals 4 (*Ref. 4*). The sampling was made at the same temperature as in the RWCU system. The central part of the diagram is the annual refueling shut-down. The rightmost part is the beginning of a fuel cycle, where the concentrations of organic "liquid" species (non volatile at room temperature) dominate over inorganic species (carbonate system), which is consistent with the information in the Pourbaix diagrams above.

<sup>&</sup>lt;sup>99</sup> Even if the acid anions are not thermodynamically stable at the temperature in the RWCU and in the sampling line, they may have been "frozen" a to quasi stable state.



In the central part of *Figure A20-4*, during the refueling shut-down, the concentrations of organic and inorganic C-14 are much lower than during operation, which is consistent with Rectangle 5 in *Figure A20-1*.

# A20.2.4. Distribution of Organic and Inorganic C-14 in the RWCU Resins from R2 in 2012

The resins generated in 2012 had an unusually high fraction of organic C-14. *Figure A20-5* shows the power history and the boron concentration in RC. The cycle started in early April 2012, had a shut-down in Jun-July and then operated for two months until mid-September. The boron concentration is shown in the inset of *Figure A20-5*. The boron concentration values during power operation were between 900 and 400 ppm. The corresponding pH-values in the RWCU at 25 °C are 6.8 to 7.0 (from *Figure A20-2*). In the Pourbaix diagram for the RWCU, this pH-interval is in the area where organic species dominate. Therefore, in the first few months of a fuel cycle, it is reasonable that there would be more organic than inorganic C-14 on the resins.



Figure A20-5. Ringhals 2, 2012. Operational period and boron concentration in RC.

#### A20.2.5-Saturation of the RWCU Resin with Respect to Organic C-14

*Figure A20-6* shows the concentration of non-volatile organic C-14 compounds in the Reactor Coolant (RC) and the RWCU outlet (CS). For the major part of the cycle, the concentration in RC (=RWCU inlet) and CS (RWCU outlet) is almost the same, indicating a saturation of the resin with respect to this type of C-14 compounds (*Ref. 4*). Whether these species are anionic (organic acids) or non-ionic (e.g. alcohols) cannot be concluded.





Figure A20-6. Ringhals 4, 2006. Concentration of non-volatile organic C-14 in the Reactor Coolant (RC) and the RWCU outlet (CS). For the major part of the cycle, the concentration in RC (=RWCU inlet) and CS (RWCU outlet) is almost the same, indicating a saturation of the resin with respect to this type of C-14 compounds (From Ref. 4).



### A20.3. BWR

Figure A20-7. Potential-pH diagrams (Pourbaix diagrams) for carbon compounds in water. The rectangular areas indicated represent the conditions in various parts of a BWR plant. Left: RC during operation at 300 °C under NWC and HWC. Right: RWCU and CCU during operation and RC, RWCU and SFP during shut-down at 25 °C. See footnote below for actual temperatures. From Yim and Caron, Ref. 11.



#### A20.3.1. Pourbaix Diagrams

*Figure A20-7* shows two potential-pH diagrams for the system carbon-water at 300 °C and 25 °C. A border line between two species indicates equal chemical activity of the species; usually the activity is close to its concentration.

The diagrams are <u>approximating the temperatures</u><sup>100</sup> in BWR RC during operation (left part), in RWCU and CCU during operation and in RC, RWCU and SFP during shut-down (right part). The diagrams come from Yim and Caron (*Ref.11*). The diagrams have rectangles indicating the combinations of redox potential and pH for the different operating conditions<sup>101</sup>. *Ref. 12-15* have similar information as *Ref. 11*.

*Left diagram Rectangles 2 (NWC), 3* and *3a (HWC)*: In RC during operation, the only stable species are carbonic acid (or carbon dioxide + water) and hydrogen carbonate ion under NWC and HWC. In some parts of the system, formate ion may be stable under HWC. During operation, the pH is close to 5.7. However, impurities (e.g. from decomposition of ion exchange resins) may alter this value up or down.

*Right diagram, Rectangle 6 (RWCU, CCU operation)*: Hydrogen carbonate is the only stable species. The pH-value during operation is around 7.0, but may be altered by impurities.

*Right diagram, Rectangle 7 (RC, RWCU, SFP shutdown):* Hydrogen carbonate is the only stable species. The pH-value during shut-down is around 6.6, due to the equilibrium with atmospheric carbon dioxide.

Even if no organic species are thermodynamically stable in the BWR systems, they have been observed on ion exchange resins. The explanation for this is probably small amounts of organic impurities in the process water, stemming from either humic acids in the make-up water or degradation products from ion exchange resins<sup>102</sup>. However, the fraction of organic compounds is lower in BWR resins than in PWR resins, which is in qualitative agreement with the information in the Pourbaix diagrams.

<sup>&</sup>lt;sup>100</sup> RC operating temp.: 286 °C. RWCU operating temp.: 45 °C. CCU temp.: 35-72 °C, depending on unit and cooling water temp. SFP temp: 25-45 °C, depending on cooling water temp. and residual power in the stored fuel. RC, RWCU and SFP shutdown temp.: 25-45 °C.

<sup>&</sup>lt;sup>101</sup>The following calculation programs have been used for calculation of pH and redox-potential: BORIS (pH only), EPRI ChemWorks and SGW4Studsvik. Input data have been obtained from the Ringhals Chemistry Data Base.

<sup>&</sup>lt;sup>102</sup> The C-14 in the process water may undergo isotopic exchange or hot-atom reactions with these organic compounds.



# A20.4. Summary

*Table A20-1* gives a summary of thermodynamically stable species and some other species detected in analyses.

Reactor type		BW	/R			PWR					
Operational			Shut-				Shut-				
state	Power o	peration	down	All	Power o	peration	down	All			
Thermody-											
namically		25 °C	25 °C	Airbor-			25 °C	Airbor-			
stable	300 °C	RWCU,	RC, SFP,	ne re-	300 °C	25 °C	RC, SFP,	ne re-			
species	RC	CCU	RWCU	leases	RC	RWCU	RWCU	leases			
С						х					
$H_2CO_3, CO_2$	х		x	*			x	*			
HCO <sub>3</sub> <sup>-</sup>	х	x	x			x					
HCOO	(x)				х	**					
СН₃ОН					х						
CH <sub>4</sub>				***		x		***			
* Measured	as "oxidize	ed species'	" in stack n	nonitorin	g						
** Analyticall	** Analytically identified in Ringhals PWRs together with acetate and oxalate										
*** Measured as "reduced species" in stack monitoring											
(): With HWC in parts of the system where the redox-potential is below - 0.5 V											

Table A20-1. Summary of thermodynamically stable species in reactor systems at various operating conditions (x) and species detected in analyses (\*, \*\*; \*\*\*). See footnotes 76 and 79 for actual temperatures in the various systems. (A-2, sheet Species).







# APPENDIX 21 Results from Dried Resin Samples from F1+F2 and F3

## A21.1. Introduction

The resins from the units F1, F2 and F3 are dried<sup>103</sup> before being solidified for deposition in the final repository SFR1. To establish the remaining C-14 activity in the dried resins, a few samples of dried resins were withdrawn from the dryer from each of the tanks TD42 and TC56. Reference samples were also taken before the drying started. It was a time consuming procedure, since the dryer had to be cooled down and opened to retrieve samples.

When the samples had been extracted according to the procedures in *Appendix 9*, the resins were also combusted to establish the remaining C-14, which conservatively will be considered as organic. The results from the combustion experiment are summarized in *Table A21-1*. The correction factors for the organic fraction in the extraction experiments were higher for these samples than for samples that had not been dried. Only two samples have been combusted.

	Extraction			Combustion				Correction factor org.			
Quanta						Fract	Fract.	Det		SD	SD
Sample	Tot, Bq	Inorg, Bq	Org, Bq	Det. Bq	DL, Bq	det.	¼DL	Det.	14DL	Det.	1/4DL
F12-12:1	2.32E+00	2.54E-01	2.07E+00	6.40E-01	8.54E-01	0.31		1.31		15%	
F3-13:3	8.02E+00	2.36E-01	7.78E+00	1.10E+00	8.53E-01	0.14		1.14		4.2%	

Table A21-1. Results from the combustion of dried samples. For F12-12:1 the negative values for the inorganic fraction are replaced by a ¼DL (Detection Limit), according to the criterion: given in Appendix 10. The combustion fraction is calculated relative to the <u>organic amount in</u> <u>the extraction</u>. All C-14 from the combustion is conservatively assumed to be organic. The yellow-marked values are adopted as best estimates. The values, 1.31 and 1.14, are above the maximum values obtained for BWR samples after extraction (1.10, Table A17-1). (E12.9, sheet Comb)

(F12-9, sheet Comb).

## A21.2. Dried resins from F12

*Table A21-2* shows a summary of the results for dried samples of carboxylate powder resins from F12. The reference sample F12-1 already has the ordinary combustion correction for wet samples. The upper part of the table shows the uncorrected values. The middle part shows the values corrected by combustion. The lower part shows the residual activity as a percentage of the reference sample. It can be seen that the inorganic fraction is removed more extensively than the organic; however, the total remaining activity is only 0.22 %.

<sup>&</sup>lt;sup>103</sup> Drying conditions: Initially at 100 °C and at an underpressure of -1.3 kPa. When 150 °C is reached, the mixing with bitumen starts. The drying time for carboxylate resins from F1+F2 is some 14 days, compared to 9-10 days for sulfonate resins from F3.



			Not corrected for extraction residual						
	Org.								
	comb.		Tot,	Inorg,			Std.dev	std.dev.	std.dev.
Sample	Factor		Bq/kg	Bq/kg	Org, Bq/kg	% org	tot	Inorg	Org
F12-13	1		3.55E+06	3.22E+06	3.30E+05	9.3%	1.3%	6.0%	64%
F12-12	1.31		6.05E+03	7.01E+02	5.35E+03	88%	18%	107%	15%
F12-14	1.31		6.42E+03	7.50E+02	5.67E+03	88%	18%	110%	14%
			Corrected	for extrac	tion residual				
			Tot,	Inorg,			Std.dev	Std.dev.	Std.dev.
Sample		Kokill	Bq/kg	Bq/kg	Org, Bq/kg	% org	tot	Inorg	Org
F12-13	Reference		3.55E+06	3.22E+06	3.30E+05	9.3%	1.3%	6.0%	64%
F12-12	Dried		7.70E+03	7.01E+02	7.00E+03	88%	18%	107%	15%
F12-14	Dried		8.18E+03	7.50E+02	7.43E+03	88%	18%	110%	14%
Mean	Dried		7.94E+03	7.25E+02	7.22E+03	91%	18%	109%	15%
							Std.dev	Std.dev.	Std.dev.
Residual after drying			Tot	Inorg	Org	% org	tot	Inorg	Org
F12-12	Dried		0.22%	0.022%	2.1%	88%	18%	107%	66%
F12-14	Dried		0.23%	0.023%	2.3%	88%	18%	110%	66%
Mean	Dried		0.22%	0.023%	2.2%	91%	18%	109%	66%

Table A21-2. Results from dried resin samples from F12. (F12-9, sheet F12dry).

## A21.3. Dried resins from F3

		Not corrected for extraction residual							
	Org.								
	comb.		Tot,	Inorg,			Std.dev	Std.dev.	Std.dev.
Sample	Factor		Bq/kg	Bq/kg	Org, Bq/kg	% org	tot	Inorg	Org
F3-11	1		1.07E+06	1.01E+06	6.12E+04	5.6%	5.9%	3.7%	43%
F3-12	1.14		1.55E+04	1.71E+03	1.38E+04	88%	29%	33%	33%
F3-13	1.14		2.15E+04	1.69E+03	1.98E+04	92%	7.5%	47%	5.5%
F3-14	1.14		2.11E+04	1.95E+03	1.92E+04	91%	4.5%	30%	3.9%
F3-15	1.14		1.65E+04	5.92E+02	1.59E+04	96%	5.6%	98%	5.8%
			Corrected	for extrac	tion residual				
			Tot,	Inorg,			Std.dev	Std.dev.	Std.dev.
Sample		Kokill	Bq/kg	Bq/kg	Org, Bq/kg	% org	tot	Inorg	Org
F3-11	Mixed R	Reference	1.07E+06	1.01E+06	6.12E+04	5.6%	5.9%	3.7%	43%
F3-12	Dried	130906	1.74E+04	1.71E+03	1.57E+04	88%	29%	33%	33%
F3-13	Dried	130908	2.43E+04	1.69E+03	2.26E+04	92%	7.5%	47%	5.5%
F3-14	Dried	130908	2.38E+04	1.95E+03	2.18E+04	91%	4.5%	30%	3.9%
F3-15	Dried	130912	1.87E+04	5.92E+02	1.81E+04	96%	5.6%	98%	5.8%
Mean	Dried		2.10E+04	1.48E+03	1.96E+04	93%	13%	43%	14%
							Std.dev	Std.dev.	Std.dev.
Residual aft	er dryin	g	Tot	Inorg	Org	% org	tot	Inorg	Org
F3-12	Dried	130906	1.6%	0.17%	26%	88%	30%	33%	54%
F3-13	Dried	130908	2.3%	0.17%	37%	92%	9.5%	47%	43%
F3-14	Dried	130908	2.2%	0.19%	36%	91%	7.4%	30%	43%
F3-15	Dried	130912	1.8%	0.059%	30%	96%	8.1%	98%	43%
Mean	Dried		2.0%	0.15%	32%	93%	15%	43%	45%

Table A21-3. Results from dried resin samples from F3. The reference samples were mixed, since each sample was too small for a separate analysis. (F12-9, sheet F3dry).

*Table A21-3* shows a summary of the results for dried samples of sulfonate powder resins from F3. The reference sample F3-11 already has the ordinary combustion correction for wet samples. The upper part of the table shows the uncorrected values. The middle part shows the values corrected by combustion. The lower part shows the residual activity as a percentage of the reference sample. It can be seen that the inorganic fraction is removed more extensively than the organic; the total remaining activity is 2.0 %. The remaining concentration is some 2 to 3 times higher than for the carboxylate samples from F12<sup>104</sup>.

# A21.4. Summary of Tests with Dried Resins

*Table A21-4* shows the concentrations in the dried resins, compared with the mean concentrations from the mass balance calculations for units F1+F2 and F3, respectively. The remaining total activity is between 0.2 % and 2 % with an organic fraction of about 90 %. The lower residual concentrations in the resins from F1+F2 compared to the resins from F3 might depend on the longer drying time (See footnote in section A21.1).

These results are based on fairly few samples, but they give a strong indication that the dried resins have released the main part of their C-14 activity to air. This conclusion is supported by measurements of C-14 in the ventilation system in the F3 WHB (See below and *Appendix 18*).

	F12: 2 dried samples, Bq/kg dw	F12: Mass Balance Mean, Bq/kg dw	F12: Mean residual	F3: 4 dried samples	F3: Mass Balance Mean	F3: Mean residual after drying
Total, Bq/kg dw	7.94E+03	1.44E+06	0.55%	2.10E+04	2.85E+06	0.74%
Inorg, Bq/kg dw	7.25E+02	1.42E+06	0.051%	1.48E+03	2.80E+06	0.053%
Org, Bq/kg dw	7.22E+03	2.49E+04	29%	1.96E+04	4.94E+04	40%
Org fraction	91%	1.7%		93%	1.7%	
Tot. Std.dev.	18%	13%		13%	5%	
Inorg std.dev	109%	13%		43%	5%	
Org. std.dev	15%	14%		14%	7%	

Table A21-4. Summary of results from dried resin samples from F12 and F3. (F12-9, sheet Comp).

# A21.5. Comparison with Dryer Release Measurements from the F3 WHB

FKA has measured the releases from the F3 WHB resin dryer (*Ref.* 8). *Table A21-6B* shows data for the moulds (kokiller) manufactured during the measurement period, March 2013-April 2014.

<sup>&</sup>lt;sup>104</sup> The higher concentration is probably a result of less efficient drying than for sulfonate resins. Carboxylate resins temporarily stored in tanks TC56 and TC58 could not be dewatered as efficiently as the sulfonate resins from the normal storage tank TD42, prior to drying. Annelie Jansson, FKA, personal communication, November 2015.



*Table A21-6A* summarizes the measured release from resin drying. *Table A21-6C* summarizes the amounts of CCU resins solidified from tanks TD42 and TC56.

*Table A21-5* shows the calculated mean concentration in the solidified resins, assuming that all C-14 released comes from the dried resins<sup>105</sup>. The mean concentration is 6.68E6 Bq/kg dw, and is higher than the average concentrations of 1.44E6 and 2.85E6 Bq/kg/dw from the mass balances in *Appendix 1* and 2 for F12 and F3, respectively<sup>106</sup>.

		Resin, kg dw	Activity, Bq	Mean conc, Bq/kg dw	Max. conc., Bq/kg dw	Conc. Back- flushing, Bq/kg/dw	Note	
Drying & stack release		12684	8.48E+10	6.68E+06			TD42+TC56	
Wet analysis	F12	34169	4.94E+10	1.44E+06	4.40E+06	1.16E+07	*	
Wet analysis	F3	22555	6.42E+10	2.85E+06	4.74E+06		*	
Wet analysis	F12+F3	56724	1.14E+11	2.00E+06				
* Some samples probably had a too low concentration, due to inadvertent handling								

Table A21-5. Comparison of releases from resin drying and analysis of wet resins.(F3-3, sheet Summary).

The maximum concentrations measured in wet resins are some 30 % lower than the concentration calculated from the releases. The highest concentration in F12 resins have been measured in samples from the back-flushing tank in the unit, 1.16E7 Bq/kg. There are no analyses available for F12 CCU resins in the handling chain from F12 to the F3 WHB.

The reference samples for the dried samples (*Tables A21-2 and -3*) had concentrations 3.55E6 and 1.07E6 Bq/kg dw. The average concentration calculated from the releases from the dryer is significantly higher. The only measured concentration equal or greater than the calculated average concentration is the sample from the back-flushing tank in F1. However, it is unlikely that such a high concentration would reach the storage tanks in the F3 WHB. Most likely, there is an overestimation of the average concentration from the release measurement. The flow measurement in the vent line from the dryer may be the reason for this.

The analyses of dried samples demonstrate that the drying reduces the concentration of C-14 in the CCU resins. The concentration calculated from the releases seems to be too high, due to some uncertainties in the sampling or the air flow measurements.

Contributions from other sources such as degassing in the storage tanks, when resins are back-flushed from F3 and when the transports from F12 are transferred to the tanks have contributed only in March 2013-July 2013 when the stack air was sampled. Thereafter, only the vent from the dryer was sampled. See *Table A21-6A* for details.

<sup>&</sup>lt;sup>105</sup> When the sampling was done in WHB stack (March-July 2013) some releases from storage tanks may also have been measured.

<sup>&</sup>lt;sup>106</sup> Some samples for the mass balances were left in the sampling position for several days, where they turned almost dry. Most likely, they also lost some C-14.

	Releases of	f C-14 from F3	WHB								
					Kokill id	Date	TD42	TC56	TC43	TC41	т
		Sampling	C-14OX	C-140X			6876	5808	1661	604	38
Start	End	point	(Bq/s)	(Bq)	14-1703	2014-04-24	0	0	0	0	20
2013-03-13	2013-03-15	WHB stack**	1.56E+02	2.45E+07	14-170	2014-04-24	679	0	0	0	(
2013-03-15	2013-03-27	WHB stack**	1.38E+03	1.45E+09	14-170	2014-04-24	0	0	0	0	(
2013-03-27	2013-04-22	WHB stack*	1.77E+03	3.98E+09	14-180	2014-04-29	0	0	0	0	
2013-04-22	2013-05-02	WHB stack*	5.46E+03	4.64E+09	14-180	2014-04-29	0	0	637	0	
2013-05-02	2013-05-08	WHB stack*	7.76E+03	4.03E+09	14-180	2014-04-29	0	0	0	0	(
2013-05-08	2013-05-16	WHB stack*	4.43E+02	3.07E+08	14-180	2014-04-14	0	0	482	0	(
2013-05-16	2013-06-02	WHB stack*	5.88E+03	8.63E+09	14-180	2014-04-14	0	0	0	0	(
2013-06-02	2013-06-12	WHB stack*	8.45E±03	7 /2E±00	14-170	2014-01-31	0	0	0	0	24
2013-06-02	2013-06-20	WHB stack*	2 885+03	1.955+00	14-170	2014-01-31	0	705	0	0	(
2013-00-12	2013-00-20		2.00E+03	1.9500+09	14-170	2014-01-31	0	0	0	0	(
2013-00-20	2013-07-10		1.01E+03	1.21E+10	13-171	2014-01-07	0	708	0	0	2
2013-07-10	2013-07-16	WHB stack*	1.17E+04	6.29E+09	13-171	2014-01-07	0	0	0	0	(
2013-07-16	2013-07-29	WHB stack*	5.00E+03	5.53E+09	13-171	8 2013-12-21	0	0	0	0	23
2013-07-29	2013-08-05	Dryer***	1.00E+03	6.13E+08	13-171	8 2013-12-21	690	0	0	0	(
2013-08-05	2013-09-03	Dryer***	5.41E+02	1.35E+09	13-1718	8 2013-12-21	0	0	0	0	0
2013-09-03	2013-09-18	Dryer***	2.44E+03	3.16E+09	13-171	2013-12-14	0	0	0	0	33
2013-09-18	2013-09-25	Dryer***	3.06E+03	1.86E+09	13-171	2013-12-14	660	0	0	0	0
2013-09-25	2013-09-30	Drver***	7.16E+02	3.12E+08	13-1/1	2013-12-14	0	0	0	0	2/
2013-09-30	2013-10-16	Dryer***	/ 11E-01	5.69E±05	13-171	2013-11-28	0	671	0	0	24
2013-09-30	2013-10-10	Dryer***	4.112-01	J.03L+0J	13-171	2013-11-28	0	0	0	0	0
2013-10-16	2013-11-04	Dryer	9.55E+02	1.57E+09	13-171	2013-11-19	0	0	0	0	27
2013-11-04	2013-11-19	Dryer***	1.74E+03	2.25E+09	13-171	2013-11-19	0	644	0	0	0
2013-11-19	2013-11-26	Dryer***	9.10E+02	5.52E+08	13-171	2013-11-19	0	0	0	0	0
2013-11-26	2013-12-10	Dryer***	3.72E+03	4.51E+09	13-1714	2013-09-19	0	0	0	0	23
2013-12-10	2014-01-08	Dryer***	1.56E+03	3.90E+09	13-1714	2013-09-19	677	0	0	0	(
2014-01-08	2014-02-05	Dryer***	1.53E+03	3.70E+09	13-1714	2013-09-19	0	0	0	0	
2014-02-05	2014-02-17	Drver***	5.74E+02	5.95E+08	13-180	2013-08-29	0	0	0	0	
2014-02-17	2014-04-11	Drver***	7.87E+02	3.59E+09	13-171	2013-00-23	0	0	0	0	24
2014-04-11	2014-04-24	Dryer***	2 88E±02	3.22E±08	13-171	2013-07-18	0	636	0	0	0
2014 04 11	2014 04 24	Dryor***	2.002+02	1.21E+00	13-171	2013-07-18	0	0	0	0	0
2014-04-24	2014-04-30	Dryer	2.200 +02	1.21E+00	13-1710	2013-07-10	0	0	0	0	26
iviean/Sum	-		2.38E+03	8.48E+10	13-1710	2013-07-10	0	694	0	0	0
* 30743RC901	L				13-1710	2013-07-10	0	0	0	0	0
** 30743RC90	)3				13-170	2013-06-15	0	612	0	0	25
***30743tork					13-170	2013-00-15	0	012	0	0	( (
Tahle	A21-6A (	F3-3 show	t Rolon	505)	13-170	2013-07-04	0	0	0	0	16
I doie	.121 0/1. (	1 <i>J J</i> , <i>snee</i>	a nereu	5057.	13-1708	8 2013-07-04	0	768	0	0	0
					13-170	8 2013-07-04	0	0	0	0	0
	Sum al	l Sum			13-170	2013-05-18	0	144	0	0	C
					13-170	2013-05-18	724	0	0	0	0
	tanks,	1D42+FC	56,		13-170	2013-05-18	0	0	0	0	(
	kg dw	kg dw			13-170	2013-05-03	605	0	0	0	1
	1979/	1769/			13-170	2013-05-03	0	0	0	0	(
	10/04	12004			13-170	2013-04-23	719	0	0	0	(
Table	A21-6C.	(F3-3, she	et Moul	ds).	13-170	2013-04-23	0	0	0	0	24
					13-170	2013-04-23	0	0	0	0	(
					13-1704	2013-03-25	0	0	0	0	23
					13-1704	2013-03-25	768	0	0	0	0
					13-170	2013-03-25	0	0	0	0	(
					13-170	2013-03-14	U	0	U	U	1 Z

 Table A21-6. A: Releases of C-14 from resin drying. B Moulds (kokiller) produced during the measurement period. C: Amounts of CCU resins solidified.

# A21.6. Summary of Dried Resins from F3

*Table A21-7* shows a summary of the remaining accumulation of C-14 in dried resins from F3, based on the four samples analyzed, together with the results from the mass balance for wet resins (see *Appendix 2*).

	<b>D</b>		
	Dried resins*		
	Mass	Mass	
	balance	balance	
	TD42	TD42	Ref. 4 +
	Sept 2009-	Sept 2009-	BFF + ds-
	Dec 2012	Dec 2012	corr.
Equivalent Full Power Hours (EFPH)	31800	31800	
Equivalent Full Power Years (EFPY)	3.63	3.63	
Body-Feed Factor, Average (BFF)	2.08	2.08	2.08
Dry Solids Correction Factor in Sample			1.16
Residual after drying:			
Organic fraction	93%	1.7%	24%
Total accumulation in CCU	0.031%	2.6%	1.7%
Organic accumulation in CCU	0.029%	0.044%	0.40%
Resin, kg dw		22555	
Total accumulation, Bq/MWh <sub>th</sub>	7.45E+00	6.12E+02	4.05E+02
Inorganic accumulation, Bq/MWh <sub>th</sub>	5.25E-01	6.01E+02	3.08E+02
Organic accumulation, Bq/MWh <sub>th</sub>	6.92E+00	1.06E+01	9.72E+01
Total Std. Dev.	15%	4.9%	
Inorganic Std. Dev.	43%	4.9%	
Organic Std. Dev.	17%	7.2%	
* Based on four samples			

Table A21-7. Comparison of accumulation of C-14 in CCU resins at F3 from this study. (F3-1, sheet Summary dried).

## A21.7. Summary of dried resins from F1+F2

*Table A21-8* shows a summary of the remaining accumulation of C-14 in dried resins, based on the two samples analyzed. It presents the results from two different calculations; one for wet resins (see *Appendix 1*) and one for dried resins.

- 0. Recalculates the mass balance from Model 1 to the concentration of dried resin samples, using the mean concentration from the mass balance.
- 1. Mass Balance over T42, T43 and TC56, using all samples from TC56 (in 2012 also TC58)



Calculation Model	0	1
	Recalc. to	Best
Classification	dried resins	estimate
	Dried Mass	
	balance	Mass
	TC56+	balance
	T43+T42	TC56+
	****	T43+T42
Equivalent Full Power Hours, F1	40057	40057
Equivalent Full Power Hours, F2	33356	33356
Equivalent Full Power Years, F1	4.57	4.57
Equivalent Full Power Years, F2	3.81	3.81
Body-Feed Factor, Average (BFF)	1.00	1.00
Dry Solids Correction Factor in Sample	e	
Organic fraction	91%	1.7%
Total accumulation in CCU	0.0068%	0.93%
Organic accumulation in CCU	0.0062%	0.016%
Total accumulation, Bq/MWh <sub>th</sub>	1.67E+00	2.30E+02
Inorganic accumulation, Bq/MWh <sub>th</sub>	1.53E-01	2.26E+02
Organic accumulation, Bq/MWh <sub>th</sub>	1.52E+00	3.97E+00
Total Std. Dev.	22%	13%
Inorganic Std. Dev.	110%	13%
Organic Std. Dev.	24%	14%
**** Based on two samples		

Table A21-8. Comparison of accumulation of C-14 in CCU resins at F1+F2 from this study.The leftmost column with results is based on dried resins.(F12-1, sheet Summary dried).



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### APPENDIX 22 A Correlation for the Accumulation of C-14 in CCU Resins

### A22.1. R1

In unit R1 in the cycle 2013-2014, the accumulation of C-14 (as Bq/MWh<sub>th</sub>) dropped to less than half of the value for the previous cycles; see *Appendix 4* for details. The most obvious factor was the decrease in CCU resin mass used compared to earlier cycles. Another possible factor is that the unit was running its first cycle with complete Forward Pumping (FP) of the hot drains. This operational mode reduces the amount of condensate to be cleaned to some 70 % of its earlier value. Furthermore, only three out of four filters per turbine train were used<sup>107 108</sup>. The temperature of the water was also reduced, compared to earlier cycles.

The length of the cycles for R1 has been varying, so it seems reasonable to normalize the use of CCU resins to the number of EFPH produced in each cycle<sup>109</sup>. Furthermore, it is assumed that C-14 is absorbed only by the anion part of the CCU resin. This is reasonable, since C-14 only exists as anionic species in the CCU-system; Se Figure A20-7, right part. Accordingly, the normalization should be made using the amount of anion equivalents, yielding the parameter  $x_a$ ,<sup>110</sup> having the unit anion equivalents/h. For the full discussion on the amount of anion equivalents in the CCU resin, see *Appendix 34* in *Part 2* of this report.



*Figure A22-1a* shows large variations in the parameter  $x_a$  (the lowest value is the cycle 2013-2014) and the total accumulation as Bq/MWh<sub>th</sub>. The highest accumulation was in the very short cycle in 2009 (some two months).

<sup>&</sup>lt;sup>107</sup> R. Hellström, RAB, personal communication May 2014.

<sup>&</sup>lt;sup>108</sup> Before the cycle 2013-2014 a filter was used for 42 days. During the 2013-2014 cycle, the average operating time was 60 days.

<sup>&</sup>lt;sup>109</sup> The production of C-14 is proportional to EFPH. For a given configuration and operational mode of a plant, the volume (and mass) of condensate cleaned is also proportional to EFPH.

<sup>&</sup>lt;sup>110</sup> Number of anion equivalents divided by EFPH.



In Figure A22-1a, a regression line fits the accumulation factor to the data points. The correlation function for the accumulation factor is<sup>111</sup>

where

	$f = ax_a$	Eq. (A22-1)
f	accumulation factor of C-14, Bq/MWhth	
$n_a$	number of anion equivalents used	
Ν	number of Equivalent Full Power Hours, EFPY	
$x_a = n_a / N$	anion equivalents/h	
a	coefficient (slope) for $x_a$ in the regression,	
	(Bq/anion equivalents)/ MW <sub>th</sub>	

The regression coefficient,  $r^2=0.91$ , indicates a good fit. The standard deviation in the coefficient a ("slope" in Figure A22-1a) is 13 %. A similar fit has been made for the inorganic accumulation with  $r^2 = 0.84^{112}$ . Figure A22-1b shows the regression for the organic fraction. Here the regression coefficient is lower;  $r^2=0.82$ , but the fit is still fairly good. A summary of the regression results are given in Table A22-2.

#### A22.2. 01 + 02

f  $n_a$ Ν

How do other units look? Let us first have a look at O1+O2, where the accumulation has been calculated for each tank contents. Figure A22-2a shows a large scatter for  $x_a$  as well as the accumulation. The regression coefficient is mediocre;  $r^2=0.40$ . The organic fraction has a poor regression coefficient;  $r^2=0.13$ ; see Figure A22-2b.



<sup>&</sup>lt;sup>111</sup> The justification for choosing Eq. A22-1 is the following: If you use no CCU resin at all  $(x_a=0)$  there will be no accumulation of C-14.

<sup>&</sup>lt;sup>112</sup> The regression coefficient for the total and the inorganic accumulations are almost equal, since the inorganic fraction constitutes 94-99 % of the total accumulation. This applies to all the regressions presented in this Appendix.



However, if the regression analysis is made for the mean accumulation for each sampling campaign, the regression is good for the total accumulation,  $r^2=0.93$ ; see *Figure A22-3a*. The organic regression coefficient is lower,  $r^2=0.76$ , but still fairly good; see *Figure A22-3b*.

The better fit obtained with the campaign mean values may be explained by the fact that several individual samples lacked detected values. The value assigned to these samples, 25 % of the detection limit, may have contributed to the scatter among the individual samples. In the campaign mean values these values do not contribute significantly.



#### A22.3. O3





Unit O3 has sampled the resins prior to transfer to concrete tanks; see *Chapter A11.7*. The  $x_a$ -values for O3 are lower than for R1, but in the same range as for O1+O2. The fit for the total accumulation is also good;  $r^2$ =0.90; see *Figure A22-4a*. The fit for the organic accumulation is less;  $r^2$ =0.69; see *Figure A22-4b*.

### A22.4. F1+F2 and F3

No regression analysis is possible for the units F1+F2 and F3, since the analysis and calculation models used only yield one single  $x_a$ -value and one accumulation value from the mass balance.

## A22.5. Regression Analysis of Data from R1, O3 and F3, with CCU Resin Use Normalized to EFPH

*Figure A22-5a* shows regression for all values from the units O3, F3 and R1. The regression coefficient is  $r^2$ =0.90, indicating a very good fit. The reason for choosing these units is the absence of an efficient air bubbling of the spent CCU resins. The only data point from F3 lies above the regression line, indicating that the regression may underestimate the accumulation in F3. The organic fit  $r^2$ =0.79 is lower, but still good; See *Figure A22-5b*.



### A22.6. Regression Analysis of Data from O1+O2 and F1+F2, with CCU Resin Use Normalized to EFPH

It was observed that the regression line total accumulation for O1+O2 also included the data point for F1+F2; See *Figure A22-6a*. The fit  $r^2$ =0.96, is very good. However, the organic accumulation shows a substantially lower fit,  $r^2$ =0.59, compared to  $r^2$ =0.76 for O1+O2. This is probably due to the low organic accumulation for F1+F2; the data point lies well below the regression line in *Figure A22-6b*.





# A22.7. Regression Analysis of Data from R1, O3 and F3, with CCU Anion Resin Use Normalized to Mass of Condensate Cleaned

				x,kg	x <sub>a</sub> ,	$x_{c_i}$ anion		m <sup>3</sup> con-
	Steam	CCU	Anion	CCU	anion	equiv./		densate
	flowrate,	flowrate,	equiv./	resin	equiv./	kg con-		/anion
Unit	kg/s	kg/s	kg DW	/EFPH	EFPH	densate	Bq/MWh <sub>th</sub>	equiv.
03	2105	1215	1.11	0.39	0.43	1.12E-07	4.15E+02	8.96E+03
R1 2 FP	1248	942	1.33	1.09	1.46	2.51E-07	1.16E+03	3.99E+03
F3	1818	1180	1.18	0.27	0.32	7.55E-08	6.12E+02	1.32E+04
R11FP	1248	1041	1.33	0.91	1.21	3.23E-07	1.09E+03	3.10E+03
R1 No FP	1248	1248	1.33	0.64	0.85	3.24E-07	6.18E+02	3.08E+03

Table A22-1. Summary of condensate flow-rates, and various ways to express the parameter x to plot the accumulation of C-14 in CCU resins. The x-values are from the sampling periods. (A-2, sheet CCUupt CV).

The regression results Shown in the previous sections indicate that there is a saturation of the CCU resins with respect to C-14. The concentrations in the resins from O3, F3 and R1 are all in the range 1-4E6 Bq/kg dw; see *Figures A13-2, A2-4 and A4-52*.

*Table A22-1* shows a summary for condensate flow-rates and CCU resin use per EFPH. Since C-14 is expected to occur as anionic species, the parameter anion equivalents per EFPH and per kg condensate cleaned has also been given. For O3 and F3 one summary value is given, while for R1 three sets of values are given: without FP, FP for one turbine train and FP for both turbine trains.

*Figure A22-7* shows the accumulation factor as a function of the parameter  $x_c$ , CCU anion equivalents/kg cleaned condensate. The diagram shows more data points than shown in Table

A22-1; actually the same points as in *Figure A22-5*. The regression coefficient,  $r^2$ =0.89 is 0.01 units lower than for the regression vs. the  $x_a$ -value, and still good.



Figure A22-7. Regression of the accumulation of C-14 in BWR CCU resins as a function of the parameter x<sub>c</sub> anion equivalents/kg condensate cleaned. (A-2, sheet D3 f).

#### A22.8. Formulas for the Calculation of Historic Accumulation from the Correlation to EFPH Normalization

The correlation for the accumulation factor is

$$f = ax_a$$
 Eq. (A22-1)  
The correlation for the accumulation of C-14 to the energy production may be written as

$$Q = Eax_a \qquad \qquad Eq. (A22-2)$$

which may be simplified to

$$Q = an_a P \qquad \qquad Eq. (A22-3)$$

where

Index

f	accumulation factor of C-14, Bq/MWh <sub>th</sub>
Q	accumulation of C-14, Bq
Ε	energy production, MWh <sub>th</sub>
$n_a$	number of anion equivalents used
Р	thermal power of the unit, MW <sub>th</sub>
Ν	<i>E/P</i> , number of EFPY
$x_a = n_a / N$	anion equivalents/h
a	coefficient (slope) for $x_a$ in the regression,
	(Bq/anion equivalents)/ MW <sub>th</sub>
a	anion



This means that the installed thermal power and the amount of anion equivalents used are the two main factors dominating the accumulation of C-14 in CCU resins. The slope coefficient a may be interpreted as a correction for losses in the process and the handling of the resins.

All equations for calculating the historical accumulation of C-14 in resins will be presented in *Appendix 23*.

*Table A22-2* shows a summary of all the regression analyses made in this *Appendix*. The coefficients *a*, to calculate the accumulation factor from the parameter  $x_a$  are given together with their regression coefficients  $r^2$  and their standard deviations.

# A22.9. Summary of Results from Regression Analysis and Selection of Parameters for Calculation

	<b>Regression from</b>	Coefficient <i>a</i> , (Bq/anion equiv.)/MW <sub>th</sub>			Org.	St	andard devia	viation	
Nr	Unit	Total	Inorganic	Organic	fract.	Total	Inorganic	Organic	
1	03	8.84E+02	8.71E+02	1.30E+01	1.5%	8.8%	8.8%	18%	
	r <sup>2</sup>	0.90	0.90	0.69					
2	R1	7.89E+02	7.71E+02	1.83E+01	2.3%	13%	13%	18%	
	r <sup>2</sup>	0.91	0.90	0.84					
3	O3 F3 R1	8.23E+02	8.06E+02	1.69E+01	2.1%	7.2%	7.4%	11%	
	r2	0.90	0.89	0.79					
4	O1+O2 batch means	2.94E+02	2.72E+02	2.23E+01	7.6%	16%	17%	33%	
	r <sup>2</sup>	0.93	0.92	0.76					
6	01+02 F1+F2	3.17E+02	2.72E+02	2.23E+01	7.1%	10%	11%	41%	
	r2	0.96	0.95	0.59					
7	O1+O2 single samples	1.97E+02	1.74E+02	2.27E+01	12%	18%	17%	37%	
	r <sup>2</sup>	0.40	0.41	0.13					
8	01+02 F1+F2	3.17E+02	2.72E+02	5.47E+00	1.7%	10%	11%	41%	
	$\mathbf{r}^2$	0.96	0.95	0.59					

Table A22-2. Summary of regression results to calculate the accumulation factors from the<br/>regression analyses. (X-1, sheet Table).

O3 and F3 shall use regression  $(1)^{113}$ . For F3 this regression may underestimate the accumulation, so some other methods should also be used for F3.

R1 shall use regression (2). B1+B2, not air bubbling their resins, shall use the regression (3). O1+O2 before the introduction of air bubbling in 2001 shall use the same set of parameters; regression (3). O1+O2 shall use their own set of parameters, regression (4), for resins from 2001 onwards. F1+F2 shall use regression (8), based on O1+O2 and F1+F2 air bubbled resins, but with an adjusted, lower organic accumulation<sup>114</sup>; see *Table A22-3* for adopted values.

The regressions<sup>115</sup> (6) and (7) are not used for calculations; they are included as a comparison.

<sup>&</sup>lt;sup>113</sup> The number in parenthesis refers to the correlation number in the first column of *Table A22-2*.

<sup>&</sup>lt;sup>114</sup> Since the organic fraction in regression (7) is appreciably higher than the mean value obtained in sample analyses for F1+F2, the organic coefficient has been adjusted to 1.7 % of the total.

<sup>&</sup>lt;sup>115</sup> Regression (5) used all data points, just as a test. It is not included in the table.

Figure A22-8 shows the results from the regressions shown in *Table A22-2*. Table A22-3 and Figure A22-9 show the regression parameters to be used for calculations of the historical and future accumulations.



Figure A22-8. Summary of regression results to calculate the accumulation factors. (X-1, sheet D fact).

	Regression from	Coefficient	Org.	Sta	andard devia	tion	Used for		
Nr	Unit	Total	Inorganic	Organic	fract.	Total	Inorganic	Organic	
1	03	8.84E+02	8.71E+02	1.30E+01	1.5%	8.8%	8.8%	18%	O3, F3
2	R1	7.89E+02	7.71E+02	1.83E+01	2.3%	13%	13%	18%	R1
3	O3 F3 R1	8.23E+02	8.06E+02	1.69E+01	2.1%	7.2%	7.4%	11%	B1+B2; O1+O2 before 2001
4	01+02	2.94E+02	2.72E+02	2.23E+01	7.6%	16%	17%	33%	O1+O2 from 2001 onwards
8	01+02 F1+F2	3.17E+02	2.72E+02	5.47E+00	1.7%	10%	11%	41%	F1+F2 with adj. org. fract.

Table A22-3. Summary of regression results used to calculate the accumulation factors fromthe regression analyses. (X-1, sheet Table)



Figure A22-9. Summary of regression results used to calculate the accumulation factors. (X-1, sheet D fact x u).