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^{14}C produced in Swedish nuclear power reactors – measurements on spent ion exchange resins, various process water systems and ejector off-gas

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This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the authors and do not necessarily coincide with those of the client.

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Summary

This study has been focused on determining the concentration of ^{14}C – in terms of both organic and inorganic ^{14}C content – in spent ion exchange resins from Swedish BWRs and PWRs. The purpose was to offer data that can serve as a basis for reliable estimates of the total ^{14}C inventory in the Swedish waste repository SFR.

The investigation included 3 PWR reactors (Ringhals), 2 BWR-HWC reactors and 4 BWR-NWC reactors. Based on the 22 batch samples analyzed, the total amount of ^{14}C in spent resins, annually produced from the Swedish reactors, was found to be 120 GBq, of which 19% is in organic form. The main contribution originates from the PWR reactors, followed by the BWR-NWC reactors, whereas BWR-HWC-reactors constitute the smallest fraction. For the BWR reactors, nearly 100% of the total ^{14}C inventory in the resins originates from the condensate clean-up resins. The total amount of ^{14}C in spent resins, generated by all energy producing reactors in Sweden during 40 years of operation, has been estimated to be 4.8 TBq. The largest uncertainty related to the derived value concerns the difference in the type of resins used by the reactors, and which strategy the power plants use to immobilize the spent resins.

Measurements of organic and inorganic ^{14}C in various process water systems and in ejector off-gas were also performed. The activity concentration in the reactor water was found to be 28 kBq/kg in PWR-samples – of which 0.3% was in inorganic form – and 1–3 Bq/kg in BWR-samples. Generally, the ^{14}C activity in the water was divided equally between the gas- and water phase. The relative uptake of ^{14}C by the resins, compared to the concentration in the coolant, was found to be lower for PWRs than for BWRs. The analyses of ejector off-gas in a PWR showed that the total amount of ^{14}C released as gas due to steam generator leakages was comparatively low: 1.4‰ of the calculated ^{14}C production in the reactor coolant. Estimated ^{14}C activity released as waterborne discharges was of the same magnitude.

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1 Introduction

Spent ion exchange resins are the dominating type of waste in the Swedish final repository for radioactive operational waste /Ingemansson 2001/. Studies on the consequences of possible leakages from the repository have identified ^{14}C as one of the most important nuclides in the inventory, since it constitutes one of the main dose contributors in future release scenarios /Bergström and Puigdomenech 1987/. The reason for this is its high mobility in the environment, biological availability and long half-life (5730 y). For reliable modeling and predictions of future release and migration of ^{14}C from the repository, knowledge of the ^{14}C inventory in the spent resins deposited is required. Because ^{14}C is a nuclide which is difficult to measure in nuclear waste, this knowledge has so far been limited, and the amount of ^{14}C in the resins has been calculated using somewhat uncertain correlation factors based on ^{60}Co /see e.g. Lundgren et al. 2002/. Direct measurements of ^{14}C in the spent ion exchange resins are therefore of great importance. Moreover, because the chemical form of ^{14}C will affect the mobility of the radionuclide – if released from the repository – a chemical speciation in terms of organic and inorganic ^{14}C present in the waste is also needed.

This study has been focused on determining the concentration of ^{14}C – in terms of both organic and inorganic ^{14}C content – in spent ion exchange resins from Swedish BWRs and PWRs. The purpose was to offer data that can serve as a basis for reliable estimates of the total ^{14}C inventory in the Swedish waste repository SFR. Measurements of organic and inorganic ^{14}C in various process water systems have also been performed for both types of reactors. The results have been used to obtain a rough partial mass balance with respect to ^{14}C produced in the reactor coolant. A few analyses of ejector off-gas are also included in the report, of which a mass balance of steam generator leakages in PWR has been done.

2 Background

The major part of the ^{14}C generated in light water reactors, with a possibility of being found in the reactor water clean-up system, is produced in the reactor coolant /e.g. Vance et al. 1995/. The chemical speciation of ^{14}C compounds found in the ion exchange resins is therefore directly correlated to the prevailing chemical conditions in the coolant, i.e. oxidizing or reducing. In BWRs, where oxidizing conditions prevail (i.e. under Neutral Water Chemistry (NWC) conditions), oxidized forms such as carbon dioxide and bicarbonate (i.e. inorganic carbon compounds) are expected in the coolant /Lundgren et al. 2002/. PWRs are operated under reducing conditions, resulting in the formation of mainly organic carbon species, often generalized to be hydrocarbons (e.g. methane) and CO. The chemical conditions achieved in Swedish BWRs operated with hydrogen injection (i.e. Hydrogen Water Chemistry (HWC) operated) will generally be closer to the NWC operated BWRs rather than the PWRs. Therefore, inorganic carbon species are expected to be the predominant form also in the coolant of HWC operated BWRs /Lundgren et al. 2002/.

Initially it was believed that the ^{14}C in spent ion exchange resins was solely to be in carbonate form /Martin 1986/, since organic carbon species would not be retained by the purification systems /Hesböl et al. 1990, Kunz 1985/, which seems realistic because of the high volatility of hydrocarbons and CO. More recent studies – where a more detailed investigation of organic ^{14}C species believed to be formed in the coolant under reducing conditions have been made – show that the organic compounds not only are limited to hydrocarbons and CO. Possible organic compounds formed include e.g. acetaldehyde, methanol, ethanol, acetone and acetic acid /Matsumoto et al. 1995/; and formaldehyde and formic acid /Lundgren et al. 2002/. Due to their chemical properties, alcohols, aldehydes and ketones are not expected to be bonded to the resins. According to /Lundgren et al. 2002/ the only compound with a possibility to attach to the ion exchange resins is formate. In /Gruhlke et al. 1985/ it is suggested that compounds such as formaldehyde, formic acid and acetic acid may be produced in small amounts in the PWR wastes. However, the attachment mode of the organic species that are absorbed by the resins is unknown (not believed to occur by ion exchange) /Vance et al. 1996/.

Under oxidizing conditions the dominant ^{14}C species retained by the resins is believed to be bicarbonate, HCO_3^- /Lundgren et al. 2002/.

3 Ion exchange resins

3.1 Sampling and sample description

Spent resin samples of 50–2,000 ml were collected from different storage tanks (see Table 3-1) in plastic containers in Ringhals (PWR), Barsebäck (BWR-HWC), Forsmark (BWR-NWC) and Oskarshamn (BWR-HWC). Generally, half of the collected sample volume consisted of excess water. From Ringhals (reactors R2, R3 and R4), 4 batch samples were taken out; from Barsebäck (reactor B2), 3 samples; from Forsmark, 4 samples (collective samples from all three reactors); and Oskarshamn, 1 sample (collective sample from reactors O1 and O2). From each batch sample, 2–8 subsamples were taken out for ¹⁴C-analysis and 1 sample for gamma-analysis. The number of processed subsamples for ¹⁴C-analysis from each reactor type and process system is shown in Table 3-1.

In total, 38 subsamples have been analyzed. The extent of both batch- and subsamples were initially intended to be larger, but has been cut, due to sampling difficulties or lack of sampling possibilities.

The samples from Ringhals were collected in the middle of 2004 and represent different storage tanks; each containing spent ion exchange resins from a variety of cleaning systems, see Table 3-2. Generally, all resins were mixed bed resins, consisting of beads (granular resins), and in operation during the fuel cycle 2002–2003.

The batch samples taken from the BWR-HWC reactors in the end of 2004, generally represent spent ion exchange resins from individual process systems, see Table 3-3. IX 1–3 were mixed bed powder resins, whereas IX 4 consisted of mixed bed granular resins. IX 1 and IX 2 were collected from the same tank but at different levels; consequently IX 2 had a lower water content. Tank T12 in Oskarshamn (represented by IX 3) contained spent fuel pool resins, in operation during 2003 and 2004. The spent resins in T42 (IX 4) covered a production period of up to approximately ten years back.

In Forsmark, the spent resins from the three BWR-NWC reactors are collectively stored in tanks representing different process systems, see Table 3-4. Batch samples from these storage tanks were collected in the beginning of 2005. IX 1 and IX 4 were granular mixed bed resins and IX 2 and IX 3 were mixed powder resins. Tank TC43 and TD44 contained spent resins covering a period of several years back. The resins in tank TC41 covered a production period of up to six years back.

Table 3-1. The number of samples processed (i.e. subsamples) of spent ion exchange resins from each reactor type and process system.

System	332 Condensate cleaning	324 Spent fuel pool cleaning	331 Reactor cleaning	342 Waste processing
BWR-NWC (F1, F2, F3, O1)	2	2	2	2
BWR-HWC (B2, O2)	4	2	2	
PWR (R2, R3, R4)			22	

Table 3-2. Detailed description of batch samples of spent ion exchange resins collected from the three PWR reactors at Ringhals in 2004, including time of operation (fuel cycle). CS = Cleanup system for reactor coolant.

Sample ID	Reactor	Tank	Content/time of operation ¹
IX 1	R3	WPATRS2	CS operation/2002–2003
IX 2	R2	SRST2	2 years of production ³ /2001–2003
IX 3	R4	WPATRS	CS shut down, spent fuel pool, waste processing/2002–2003
IX 4	R3	WPATRS	CS shut down, spent fuel pool, waste processing/2002–2003

¹ Reference: (Aronsson 2005, personal communication).

² WPATRS/SRST = Storage tank for spent resins.

³ Containing spent resins from CS operation, CS shut down, spent fuel pool and waste processing.

Table 3-3. Detailed description of batch samples of spent ion exchange resins collected from BWR-HWC reactors at Barsebäck and Oskarshamn in 2004, including time of operation. RWCU =Reactor water clean-up.

Sample ID	Reactor	System	Tank	Content/time of operation ⁴
IX 1	B2	332	BT 142 (middle)	Condensate cleaning/2004
IX 2	B2	332	BT 142 (top)	Condensate cleaning/2004
IX 3	O1 + O2	324	T12	Spent fuel pool/2003–2004
IX 4	B2	331–342	T42	RWCU+waste processing/?– 2004

Table 3-4. Detailed description of batch samples of spent ion exchange resins collected from BWR-NWC reactors at Forsmark in 2005, including time of operation.

Sample ID	Reactor	System	Tank	Content/time of operation ⁵
IX 1	F1+F2+F3	331	TC43	RWCU, ?–2004
IX 2	F1+F2+F3	3326	TD42	Mostly condensate cleaning, 2004
IX 3	F1+F2+F3	3247	TC41	Mostly spent fuel pool, ?–2004
IX 4	F1+F3	342	TD44	Waste processing, ?–2004

⁵ Reference: (Helmersson 2005, personal communication).

⁶ A minor fraction of the stored resins in TD42 belonged to system 342 and 324.

⁷ A minor fraction of the stored resins in TC41 might have come from TD42; i.e. system 332.

3.2 Method

To separately determine the fractions of inorganic and organic ¹⁴C compounds attached to the ion exchange resins, a method developed by /Magnusson and Stenström 2005/ was used (see Figure 3-1 for relevant system parameters and Figure 3-2 for system setup). The method is based on acid stripping followed by wet oxidation in combination with N₂-purging and magnetic stirring. This allows a complete extraction of inorganic and organic ¹⁴C-species separately.

The technique proved to be very reliable when applied on simulated spent resins, where samples of inactive Amberlite IRN-150L – spiked with ¹⁴C-labelled sodium carbonate, sodium acetate or sodium formate – were treated. The obtained recoveries for the three compounds tested varied from 94% to 97% (median values). Detailed information on the tests conducted (in total, ~ 50 tests) during the optimization of the method can be found in /Magnusson and Stenström 2005/.

3.2.1 Procedure for ^{14}C extraction

From the plastic containers, approximately 50–250 ml (generally 50% wet resin and < 50% excess water) was transferred into smaller glass bottles. From these, subsamples of approximately 1 g ww (wet weight) were taken out by a spoon after removing all excess water and placed in Erlenmeyer flasks. The samples were taken from the bottom of the bottle and visually inspected to assure a representative subsampling. If the chemical treatment was not to be conducted during the same day, a few ml of deionized water was added to the flask to prevent any effects caused by dehydration.

During the first step of the chemical treatment (lasting 30 minutes), 17 ml of 6M H_2SO_4 was added to a resin sample container, containing approximately 1 g of wet resin (see Figure 3-1). The acid stripping extracts the inorganic ^{14}C -species (HCO_3^- and CO_3^{2-}), which were transformed into $^{14}\text{CO}_2$ and absorbed in alkaline gas washing bottles (2×100 ml 2M NaOH). However, up to 2.5% of the simplest organic acids (e.g. formic acid) may also be released during the acid treatment /Magnusson and Stenström 2005/. These are – unlike the volatile hydrocarbons – not possible to collect separately from the carbonates.

During the second step (lasting 60 minutes), 20 ml of a strong oxidant (5% $\text{K}_2\text{S}_2\text{O}_8$) and 2 ml of a catalyst (4% AgNO_3) were added to the container. These chemicals, and subsequent heating ($T_{\text{hot plate}}$ set to $\sim 190^\circ\text{C}$) of the container solution, should assure a complete oxidation of all carbon compounds left in the resin sample container, i.e. organic species. These were collected separately from the inorganic fraction. To oxidize hydrocarbons, released during any time of the treatment, the gas flow passed over a catalyst (platinum on alumina pellets and CuO), placed in a hot oven ($T = 750^\circ\text{C}$).

3.2.2 Analysis

The sample processing and measurements were conducted at Ringhals and Barsebäck. Samples collected from the alkaline gas washing bottles after complete treatment were measured by Liquid Scintillation Counting (LSC) in duplicate to obtain the activities of the inorganic and organic ^{14}C fractions associated with the resin sample. The LSC samples were prepared by adding 3 ml of sample solution (2M NaOH) to 17 ml (Ringhals) or 18 ml (Barsebäck) of scintillation cocktail (OptiPhase Hisafe 3) and then counted for 20 minutes 2–4 times on a Wallac Guardian 1414 (Barsebäck) or a Packard TriCarb 2100TR (Ringhals). The detection limit for samples obtained from a resin sample of 1.0 g ww was about 750 Bq/kg (Barsebäck) or 2,200 Bq/kg (Ringhals). The background was determined by processing blank samples regularly, with the obtained sample solutions measured by LSC simultaneously with the other samples.

Besides the analyses of spent resin samples, a few measurements were also conducted on excess water from the glass bottles, to which a fraction of the collected batch samples were transferred. These analyses were only conducted on excess water from PWR resins. The total number of processed samples – including resins, blanks and samples of excess water – was 50.

Gamma analysis was performed on one separate sample from each batch sample to obtain the activity concentration of ^{60}Co . A quantity of ~ 0.5 g (Barsebäck) or ~ 1.2 g (Ringhals) of wet resin was weighed into appropriate plastic containers and counted using standard geometries. The results were used to calculate the obtained correlation factor between ^{14}C and ^{60}Co in the spent resins analyzed.

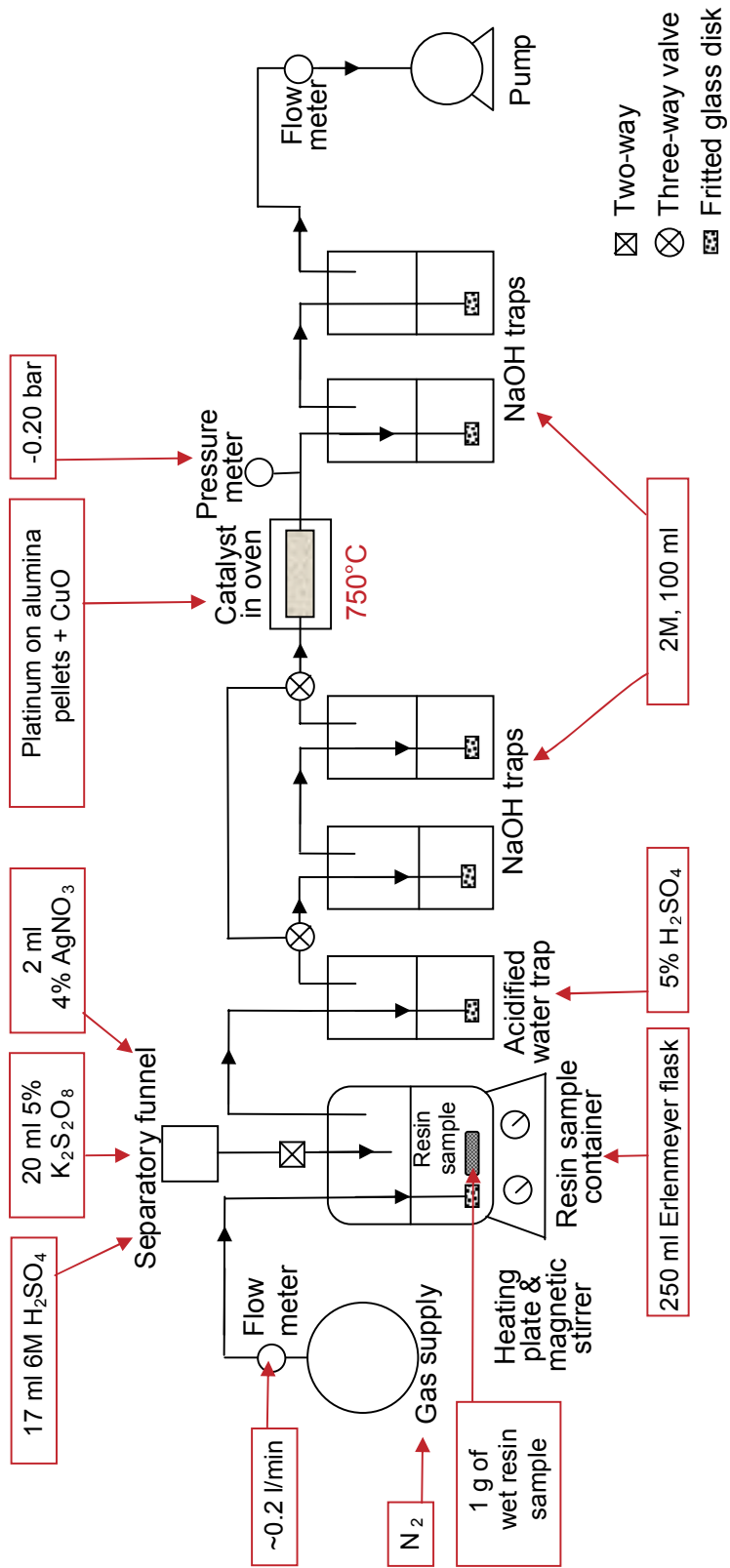


Figure 3-1. Schematic overview of the system used for extracting ¹⁴C from spent resins.



Figure 3-2. System setup for processing samples of spent ion exchange resins. Three-way valves, disconnecting the two bottles used for absorbing inorganic species after completed acid stripping, are missing in the photo.

To determine the fraction of dry solids (D.S.) of each batch sample, generally one additional subsample was taken from the batch. This sample was left to dry in an air stream for approximately one week. The wet weight (ww) was then compared to the dry weight (dw), giving dw/ww values of 0.36–0.44 for the granular resins and 0.35–0.47 for the powder resins.

During the processing of samples, the efficiency of the acid stripping and wet oxidation was again tested. The tests showed that by adding another 8 ml of H_2SO_4 after completed acid stripping, an additional amount of $\leq 3\%$ (number of tests, $n=2$) of the inorganic ^{14}C activity already extracted, was detected. This suggests a stripping efficiency close to 100%. However, the wet oxidation proved to be much more complicated when processing used resins compared to laboratory prepared ^{14}C -spiked resins. Despite repeated wet oxidations (performed on the same sample) the efficiency never reached 100% ($n=20$). The tests showed that by adding another 20–60 ml of oxidant (together with an appropriate amount of catalyst) an additional amount of 13–39% and 6–36% was detected for BWR samples and PWR samples, respectively (values given as organic ^{14}C activity released during additional wet oxidation/organic ^{14}C activity released during ordinary wet oxidation). This means that a fraction of the organic ^{14}C possibly still remains attached to the resins after complete treatment.

In order to determine the initially undetected ^{14}C activity, a few of the treated resin samples were combusted half a year later in Lund. The remaining ^{14}C found was compared to the organic ^{14}C activity detected after the chemical treatment in order to obtain correction factors.

3.3 Results

3.3.1 PWR

The results from the LSC- and ^{60}Co analysis of all subsamples processed from Ringhals are presented in Appendix 1. The data shown are the results and errors obtained after the chemical treatment of the spent resins. A summary, including correction factors for the incomplete extraction of organic ^{14}C , is found in Table 3-5. Calculated correlation factors ($^{14}\text{C}/^{60}\text{Co}$), based on the corrected ^{14}C activities are presented in Table 3-6. The total amount of ^{14}C found in the spent resins after complete treatment was 3.3–46 MBq/kg dw (1.4–20 MBq/kg ww) and the organic fraction 28–35%.

Two of the IX 1 samples (R-1:6 and R-1:8) were processed half a year later than the others (i.e. subsamples were taken from the glass bottle half a year later) to see whether any loss of ^{14}C -activity had taken place during the storage time. No significant difference was found. In IX 1, three different bead fractions could be distinguished visually: white and dark beads (representing anionic and cationic species) and a minor fraction of transparent beads floating on top of the excess water. Two of the IX 1 samples (R-1:5 and R-1:7) were samples from the transparent fraction, i.e. these samples are not representative for the batch sample. After processing, it was clear that the transparent fraction consisted of anionic resin beads (containing 60% more activity than the rest of the samples).

During two tests involving sample IX 1 and IX 2 it was shown that a small part (0.09–1.4%) of the inorganic (or possibly organic compounds soluble in NaOH) ^{14}C fraction easily is driven off by initially purging the resin sample container with N_2 during 30 minutes. This might suggest that the wet resin samples continuously are leaking $^{14}\text{CO}_2$ while stored. Also reduced forms of ^{14}C was emitted by the sample during N_2 -purging; the fraction being 0.2% ($n=1$) of the total organic ^{14}C content. The tests show that there is a possibility that a fraction of the ^{14}C -activity in the spent resins is degassed during storage and handling. However, the tests performed on R-1:6 and R-1:8 did not show any significant activity loss during storage (resins stored in 250 ml glass bottles, containing ~ 40% resins, ~ 30% water and ~ 30% gas volume).

By using the total dry mass of spent ion exchange resins produced during the fuel cycle/cycles of interest and the determined D.S. fraction in the batch sample, the total amount of ^{14}C in the resins used by each reactor unit has been calculated, see Table 3-5. This has been compared to the calculated ^{14}C production in the reactor coolant for the same period.

Batch samples IX 1 and IX 4 together represent one years' production of spent resins from R3, giving a total amount of ^{14}C in the resins used during the fuel cycle 2002/2003 of 27 GBq, corresponding to 9% of the calculated ^{14}C production in the reactor coolant.

Batch sample IX 2 represent two years of production and the total amount of ^{14}C in the spent resins from R2 represents 5.0% of the calculated ^{14}C production in the reactor coolant during two fuel cycles (2001–2003).

Batch sample IX 3 does not represent the full production of spent resins during one year from R4, which makes further calculations uncertain.

The analyses of excess water ($n=5$) showed that the present ^{14}C -species were predominantly organic (87–100%, geometrical mean = 97%). The concentration of ^{14}C in the water was 55.1–399 kBq/liter, estimated to correspond to 0.2–7% (geometrical mean = 2%) of the ^{14}C activity found in the spent resins (assuming 40% resins and 30% water in the bottles, from which the samples of excess water were taken).

Table 3-5. Summary of results obtained from PWR reactors in Ringhals and comparison with calculated ¹⁴C production in the reactor coolant during the fuel cycle/cycles of interest.

Sample ID	Reactor	Content	¹⁴ C activity in IX after chemical treatment (MBq·kg ⁻¹ ww)		Correction factor ¹⁰	Corrected total ¹⁴ C activity (MBq·kg ⁻¹ ww)	Organic fraction	Total mass spent IX ⁸ (kg dw)	Batch sample D.S. content	Total ¹⁴ C activity in spent IX (GBq)	Calculated ¹⁴ C production ⁹ (GBq)	IX/ Production
			Inorganic	Organic								
IX 1	R3	CS operation	13.9	1	1	19.6	29%	440	43%	20.1	314	6.4%
			5.49	1.03								
IX 2	R2	2 years of production	6.00	1	1	8.29	28%	1,620	42%	32.0	626	5.1%
			2.09	1.09								
IX 3	R4	CS shut down, spent fuel pool, waste processing	1.07	1	1	1.64	35%	1,150	43%	4.39	290	1.5%
			0.539	1.06								
IX 4	R3	CS shut down, spent fuel pool, waste processing	0.997	1	1	1.42	30%	2,060	43%	6.80	314	2.2%
			0.400	1.05								

⁸ Reference: (Höglund 2005, personal communication); total mass dw calculated based on a density of the virgin resin of 1.15 kg/liter and a D.S. content of 45% (Avenbrand 2005, personal communication).

⁹ Calculations based on gross electricity production, a thermal efficiency of the plant of 33% and a production rate of 15.6 kBq/MWh_{th} /Lundgren 2005/.

¹⁰ Calculated based on combustion of four subsamples, where the remaining activity in the resins was assumed to be solely of organic origin.

Table 3-6. Calculated correlation factors ¹⁴C/⁶⁰Co for the analyzed spent resins samples from PWR reactors.

Sample ID	Reactor	Content	Corrected total ¹⁴ C activity (MBq·kg ⁻¹ ww)	⁶⁰ Co activity (MBq·kg ⁻¹ ww)	Correlation factor ¹⁴ C/ ⁶⁰ Co
IX 1	R3	CS operation	19.6	21.3	0.92
IX 2	R2	2 years of production	8.29	98.6	0.084
IX 3	R4	CS shut down, spent fuel pool, waste processing	1.64	72.1	0.023
IX 4	R3	CS shut down, spent fuel pool, waste processing	1.42	49.2	0.029

3.3.2 BWR-HWC

The results from the LSC- and ^{60}Co analysis of all subsamples processed from BWR-HWC (reactors B2, O1 and O2) are presented in Appendix 2. A summary with obtained correction factors for the incomplete extraction of organic ^{14}C is found in Table 3-7. Calculated correlation factors ($^{14}\text{C}/^{60}\text{Co}$), based on the corrected ^{14}C activities are presented in Table 3-8. The maximum total amount of ^{14}C was 0.28 MBq/kg dw (0.13 MBq/kg ww); found in condensate clean-up resins. The organic fraction was generally 6–16%.

As can be seen in Appendix 2, the organic content found in the subsamples taken from IX 1 (B-1:1 and B-1:2) differ with a factor of 10. This might be a result of an acid stripping efficiency test conducted on B-1:1, which could have affected the efficiency of the wet oxidation. Combustion of B-1:1 did not reveal the “missing” activity. However, it might have been left in the sample container solution, of which no measurement was performed.

Batch samples IX 1/IX 2 + IX 3 + IX 4 together represent one years’ production of spent resins from one BWR-HWC reactor. These give a total amount of 1.77 GBq, corresponding to 0.5% of the calculated ^{14}C production in the reactor coolant.

3.3.3 BWR-NWC

The results from the LSC- and ^{60}Co analysis of all subsamples processed from Forsmark are found in Appendix 3 with a summary and calculations presented in Table 3-9. Calculated correlation factors ($^{14}\text{C}/^{60}\text{Co}$), based on the corrected ^{14}C activities are presented in Table 3-10. The total amount of ^{14}C found in the spent resins was 0.14–2.3 MBq/kg dw (0.059–0.81 MBq/kg ww). The organic fraction was 4% (condensate clean-up resins) – 9% (RWCU resins).

No detectable forms of ^{14}C were released by solely purging the samples with N_2 prior to acid stripping. Neither were there any reduced forms of ^{14}C detected after the acid stripping was completed, i.e. the fraction of initially present or formed volatile organic ^{14}C compounds was below the detection limit.

By summarizing the total ^{14}C amount for all batch samples (IX 1 – IX 4) collected from Forsmark, this gives an estimate of the total annual production of ^{14}C in spent resins from all three reactors in Forsmark. The summation is straight forward since the spent 342-resins from F2 are incorporated in IX 4 (F1 shares the 342 system with F2 (Aronsson 2005, personal communication)). By adding this activity, the total annual production is 33 GBq, corresponding to 1.9% of the calculated average total ^{14}C production (based on production rates during the years 2002–2004) in the reactor coolant in the three reactors.

Table 3-7. Summary of results obtained from BWR-HWC reactors and comparison with calculated ¹⁴C production in the reactor coolant.

Sample ID	Reactor	System	¹⁴ C activity in IX after chemical treatment (kBq·kg ⁻¹ ww) Inorganic Organic	Correction factor ¹³	Corrected total ¹⁴ C activity (kBq·kg ⁻¹ ww)	Organic fraction	Total mass spent IX ¹¹ (kg dw·a ⁻¹)	Batch sample D.S. content	Total ¹⁴ C activity in spent resins (GBq)	Calculated ¹⁴ C production ¹² (GBq·a ⁻¹)	IX/ Production
IX 1	B2	332	96.5	1.0	105	8%	5,190	37%	1.47	342	0.43%
			6.59	1.36							
IX 2	B2	332	110	1.0	130	16%	5,190	47%	1.44	342	0.42%
			14.5	1.33							
IX 3	O1+O2	324	< 0.842 ¹⁴	1.0	< 2.26 ¹⁵	–	239	37%	< 0.00146	542 ¹⁶	< 0.0003%
			< 1.07 ¹⁴	1.33							
IX 4	B2	331–342	70.3	1.0	75.0	6%	1,500 ¹⁷	36%	0.313	268 ¹⁸	0.12%
			4.44	1.07							

¹¹ Typical annual production rates of spent ion exchange resins, calculated using data from (Arvidsson 2005, personal communication).

¹² Calculations based on gross electricity production, a thermal efficiency of the plant of 33% and a production rate of 24 kBq/MWh_{th}/Lundgren et al. 2002/.

¹³ Calculated based on combustion of two subsamples, where the remaining activity in the resins was assumed to be solely of organic origin.

¹⁴ Detection limit, i.e. the maximum possible concentration of ¹⁴C

¹⁵ Maximum possible concentration of ¹⁴C, theoretical value.

¹⁶ Average value for the total production rate for O1 + O2 over the years 2003 and 2004.

¹⁷ Total mass dw calculated based on a D.S. content of 45%.

¹⁸ Average value over the years 2002–2004.

Table 3-8. Calculated correlation factors ¹⁴C/⁶⁰Co for the analyzed spent resins samples from BWR-HWC reactors.

Sample ID	Reactor	System	Corrected total ¹⁴ C activity (MBq·kg ⁻¹ ww)	⁶⁰ Co activity (MBq·kg ⁻¹ ww)	Correlation factor ¹⁴ C/ ⁶⁰ Co
IX 1	B2	332	0.105	4.68	0.022
IX 2	B2	332	0.130	3.33	0.039
IX 3	O1+O2	324	< 2.26E-03	865	< 2.6E-06
IX 4	B2	331–342	0.0750	103	7.3E-04

Table 3-9. Summary of results obtained from BWR-NWC reactors and comparison with calculated ¹⁴C production in the reactor coolant.

Sample ID	Reactor	System	¹⁴ C activity in IX after chemical treatment (kBq·kg ⁻¹ ww) Inorganic Organic	¹⁴ C activity in IX after chemical treatment (kBq·kg ⁻¹ ww) Correction factor ²¹	Corrected total ¹⁴ C activity (kBq·kg ⁻¹ ww)	Organic fraction	Total mass spent IX ¹⁹ (kg dw·a ⁻¹)	Batch sample D.S. in spent resins content (GBq)	Total ¹⁴ C activity in spent resins (GBq)	Calculated ¹⁴ C IX/ Production ²⁰ (GBq·a ⁻¹)
IX 1	F1+F2+F3	331	53.8 5.00	1 1.12	59.4	9%	3,010 ²²	44%	0.406	1,640 ²³ 0.025%
IX 2	F1+F2+F3	332	782 28.4	1 1.05	812	4%	13,400	35%	31.1	1,690 ²⁴ 1.8%
IX 3	F1+F2+F3	324	416 24.4	1 1.05	442	6%	790	35%	0.998	1,640 0.061%
IX 4	F1+F3	342	68.3 2.92	1 1.12	71.6	5%	3,630 ²²	39%	0.666	1,640 ²⁵ 0.041%

¹⁹ Typical annual production rates of spent ion exchange resins, calculated using data from (Arvidsson 2005, personal communication).

²⁰ Calculations based on gross electricity production, a thermal efficiency of the plant of 34.8% (F1 and F2) and 36.5% (F3); and a production rate of 24 kBq/MWh_{th}/Lundgren et al. 2002).

²¹ Calculated based on combustion of two subsamples, where the remaining activity in the resins was assumed to be solely of organic origin.

²² Total mass dw calculated based on a density of the virgin resin of 1.15 kg/liter and a D.S. content of 45% (Avenbrand 2005, personal communication). Total mass based on F1+F2+F3, since the 342-system in F1 is shared with F2 (Aronsson 2005, personal communication).

²³ Average value of the total production rates (F1+F2+F3) over the years 2002–2004.

²⁴ Total production (F1+F2+F3) during 2004.

²⁵ Average value of the total production rates for F1+F2+F3 during the years 2002–2004.

Table 3-10. Calculated correlation factors ¹⁴C/⁶⁰Co for the analyzed spent resins samples from BWR-NWC reactors.

Sample ID	Reactor	System	Corrected total ¹⁴ C activity (MBq·kg ⁻¹ ww)	⁶⁰ Co activity (MBq·kg ww)	Correlation factor ¹⁴ C/ ⁶⁰ Co
IX 1	F1+F2+F3	331	0.0594	110	5.4E-04
IX 2	F1+F2+F3	332	0.812	7.79	0.10
IX 3	F1+F2+F3	324	0.442	6.0	0.074
IX 4	F1+F3	342	0.0716	111	6.5E-04

3.4 Discussion and conclusions

Based on the correlation factors between ^{14}C activity in spent resins and the calculated ^{14}C production, the total ^{14}C activity in spent resins produced annually from all reactor units in Sweden have been calculated. The calculations were carried out according to following steps: 1) Calculating the average value of annual gross electricity production, based on data over the years 1996–2004 (Aronsson 2005, personal communication), for all reactors operating during this period; 2) Calculating the corresponding thermal energy production using plant-specific thermal efficiencies (Aronsson 2005, personal communication); 3) Calculating the average annual production rate of ^{14}C in the reactor coolant /Lundgren et al. 2002, Lundgren 2005/; 4) Calculating the ^{14}C activity in spent resins from each system (where possible) and reactor unit (including uninvestigated units) using system- and reactor (PWR, BWR-HWC or BWR-NWC) specific correlation factors (from Tables 3-5, 3-7 and 3-9); and multiply with the average annual ^{14}C production. Since the ^{14}C activity in 324-resins from BWR-HWC was below the detection limit, maximum possible values have been calculated, i.e. based on the corrected value found in Table 3-7. The results are presented in Table 3-11 and system specific production rates of ^{14}C in spent resins, displayed in Figure 3-3. Since no system specific correlation factors could be made from the analyzed PWR samples, only the total ^{14}C production in spent resins from PWRs are shown. The calculations do not take into account, the ^{14}C activity found in the excess water, i.e. calculations based on an activity loss of < 10% to the excess water (see Section 3.3.1).

The total amount of ^{14}C in spent resins, annually produced from the Swedish reactors, was found to be 120 GBq, of which 19% is in organic form. The main contribution originates from the PWR reactors, followed by the BWR-NWC reactors, whereas BWR-HWC-reactors constitute the smallest fraction.

Table 3-11. Calculated total ^{14}C activity, and the corresponding normalized ^{14}C activity, in spent resins produced annually from each reactor unit.

Reactor type	Reactor	^{14}C activity in spent resins annually (GBq)	Normalized ^{14}C activity in spent resins (GBq·GWe ⁻¹ ·a ⁻¹)
BWR-HWC	B2	1.5	3.4
	O2	1.6	3.3
	R1	2.1	3.5
BWR-NWC	O1	3.6	12
	O3	11	11
	F1	9.5	12
	F2	9.6	12
	F3	11	11
PWR	R2	16	21
	R3	29	36
	R4	26	33
Total		120	

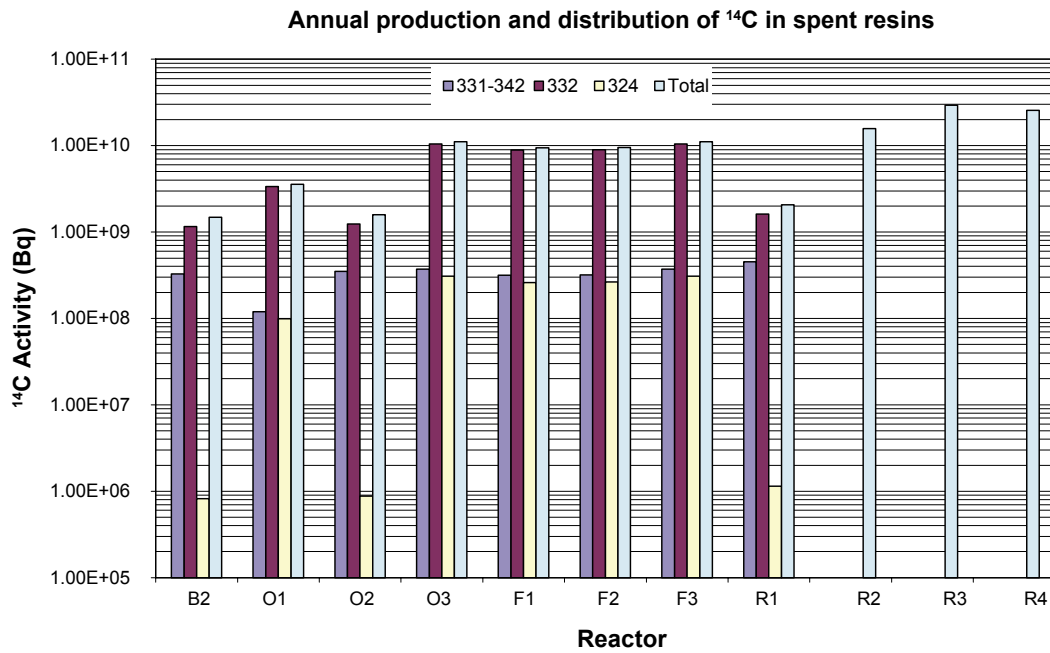


Figure 3-3. Calculated annual ¹⁴C activity in spent resins from system 331–342, 332, 324; and total amount of ¹⁴C activity originating from all operating Swedish reactors (during the period 1996–2004). The activity in BWR-HWC resins from 324 is the maximum possible.

The largest uncertainty related to the derived value concerns the difference in the type of resins used by the reactors, and which strategy the power plants use to immobilize the spent resins. The BWR-NWC reactors F1 and F2 are the only reactors operated using carboxylate resins in their condensate clean-up system (332, which makes up nearly 100% of the total ¹⁴C activity in spent resins from the BWR-NWC reactors). The other waste treatment systems in F1 and F2 – and all other reactors – use sulfonate resins. Due to the difference in chemical characteristics of the two types of resins, the spent carboxylate resins will contain a higher activity of ¹⁴C than the sulfonate resins. Since the ¹⁴C activity in spent resins from O1 and O3 are calculated from data obtained from F1, F2 and F3, these values may have been overestimated. The higher fraction of ¹⁴C on carboxylate resins may be explained as follows: The weakly acidic carboxylate resins have a higher pH than the strong acidic sulfonate resins. The higher pH allows a larger fraction of the acidic species (hydrogen carbonate and simple organic acids) to attach to the resins. Furthermore, the degradation of sulfonate resins by hydrogen peroxide, forming sulfuric acid, may also lower the pH in the sulfonate resins.

Moreover, Forsmark still uses bituminization to immobilize the spent resins, whereas all other Swedish power plant use cement immobilization. Prior to bituminization, the resins are dried using a temperature of 150°C. According to /Salonen and Snellman 1985/, 96–99% of the ¹⁴C activity in spent wet resins might be lost during this heating process. This means that a large fraction of the ¹⁴C is lost to the atmosphere prior to immobilization and that the amount of ¹⁴C in the deposited resins from Forsmark probably is overestimated.

A future project should investigate the difference between the two types of ion exchange resins in more detail- in terms of uptake and loss of ¹⁴C-activity loss during storage and heating.

Based on the data presented in Table 3-11, the total ^{14}C activity in spent resins generated during the life time of the reactors has been calculated. This corresponds to the total theoretical amount of ^{14}C in spent resins that will be – or has been – deposited in the Swedish waste repository SFR. Since the BWR reactor B1 in Barsebäck was shut down 1999, it was not included in Table 3-11, however included in these final calculations. The calculations assume a reactor life time of 40 years (except for B1 and B2, where the actual life time of 24 and 28 years, respectively, has been used). The results are presented in Table 3-12. The total amount of ^{14}C in spent resins, generated by all energy producing reactors in Sweden during their life time, has been estimated to be 4.8 TBq, of which 19% will be in organic form.

Table 3-12. The total- and organic ^{14}C activity in spent resins, generated by all energy producing reactors in Sweden during their life time (assumed to be 40 years).

Reactor type	Reactor	^{14}C activity in spent resins during reactor life time (GBq)	Organic ^{14}C activity (GBq)
BWR-HWC	B1	36 ²⁶	3.9
	B2	42 ²⁷	4.4
	O2	64	6.8
	R1	83	8.8
BWR-NWC	O1	140	6.0
	O3	450	19
	F1	380	16
	F2	380	16
	F3	450	19
PWR	R2	630	180
	R3	1,200	340
	R4	1,000	300
Total		4,800	920 (19%)

²⁶ Based on an actual life time of 24 years.

²⁷ Based on an actual life time of 28 years.

4 Process water

4.1 Sampling and sample description

Water samples from a wide range of process systems were collected at Ringhals (reactor R4), Oskarshamn (reactor O3) and Barsebäck (reactor B2) in May–July 2005. The total number of samples processed was 34 including 9 samples where the ^{14}C concentration in both water- and gas phase was determined (see Table 4-1 for an overview and sample description). Generally, 100 ml of process water was collected in pre-evacuated 100 or 250 ml gas pipettes (the larger volume used for combined gas- and water phase determinations).

From the PWR reactor R4 in Ringhals several samples were taken before (system 313) and after (system 334) the ion exchangers purifying the primary coolant. A few of the samples were taken in duplicates, i.e. at the same time. Process water was also collected before and after the spent fuel pool- (324) and steam generator blow-down (337) ion exchangers. The samples from the steam generator blow-down circuit were collected due to reported leakage from the primary circuit to the secondary circuit through the steam generators. One sample was taken from a waste water tank (342-T103) – collecting waste water from the primary circuit – representing the waterborne ^{14}C releases during June 2005.

From the BWR-HWC reactor B2 in Barsebäck process water was collected before (321; duplicate samples)) and after (331) the reactor circuit purification and the condensate cleaning. Three samples were also taken from the waste water tank (342-T25), representing the waterborne ^{14}C releases during March, April and May, 2005. These samples were initially processed and measured individually, but analyzed as one composite sample, representing the period March–May.

Table 4-1. The number of samples processed from each reactor type and process systems.

Reactor type	System	Description	No. of samples	Processed phase
PWR (R4)	313	Reactor circuit before IX	8	Gas/water
	334	Reactor circuit after IX	6	Gas/water
	324	Spent fuel pool before IX	1	Water
	324	Spent fuel pool after IX	1	Water
	337	Steam generator blow-down before IX	1	Water
	337	Steam generator blow-down after IX	1	Water
	342-T103	Waste water tank, Jun 2005	1	Water
BWR-HWC (B2)	321	Reactor circuit before IX	3	Gas/water
	331	Reactor circuit after IX	1	Water
	332	Condensate cleaning before IX	1	Water
	332	Condensate cleaning after IX	1	Water
	342-T25	Waste water tank, Mar–May 2005	3	Water
BWR-NWC (O3)	321RB902	Reactor circuit before IX	3	Gas/water
	331RB901	Reactor circuit after IX	1	Water
	332RB908	Condensate cleaning before IX	1	Water
	332RB909	Condensate cleaning after IX	1	Water

From the BWR-NWC reactor O3 in Oskarshamn duplicate samples were collected before the ion exchanger in the reactor circuit. Single samples of process water were collected after the reactor circuit purification as well as before and after the condensate cleaning.

4.2 Method

The method used was based on the same principle as described for the ion exchange resins (see Section 3.2) with the chemical treatment optimized for 100 ml of process water. A schematic outline of the system with relevant parameters is seen in Figure 4-1. The method was optimized using ^{14}C -labelled Na_2CO_3 and sodium acetate with obtained recoveries of $> 95\%$ for both compounds. The loss of sodium acetate during acid stripping (resulting in an overestimation of the inorganic ^{14}C content) was determined to be 1.2%.

4.2.1 Procedure for ^{14}C extraction

The gas pipettes were mounted onto the system as shown in Figure 4-1 and the volume of process water transferred to the sample container by applying an overpressure using N_2 at the upper inlet of the pipette. For samples containing gas- and water phase, the sample was purged with N_2 for 30 minutes to collect any volatile compounds before transferring the water volume to the sample container. The inorganic ^{14}C -species were extracted from the water by adding 50 ml of 8M H_2SO_4 and purging the water volume with N_2 for 1 hour. After the acid stripping was completed, the organic species were oxidized by adding 2×50 ml of 5% $\text{K}_2\text{S}_2\text{O}_8$ and 2×5 ml of 4% AgNO_3 in two steps during subsequent heating ($80\text{--}100^\circ\text{C}$) for 1.5 hours. The two ^{14}C -fractions (organic and inorganic) were collected separately in gas washing bottles containing 2M NaOH .

4.2.2 Analysis

All samples were processed at Barsebäck and Ringhals; the samples from Oskarshamn were transported to Ringhals. The procedure of the preparation of LSC samples and equipment used for ^{14}C analysis were identical to the one described in Section 3.2.2. All samples were initially measured 3–5 times by LSC with a detection limit of ~ 20 Bq/kg. Samples below the detection limit were precipitated as BaCO_3 and the precipitate remeasured by LSC giving an improved detection limit of ~ 0.4 Bq/kg.

During the sample processing, the efficiency of the degassing of the water samples prior to water processing was tested to be 100% ($n=1$). The efficiency of the wet oxidation was also 100% (within statistical limits; $n=8$), i.e. no significant difference in the absorbed ^{14}C activity in the wash bottles was seen between the two additions of wet oxidation chemicals. The degree of oxidation by means of the strong oxidant (i.e. the ability of converting the ^{14}C -species to CO_2) was found to be nearly 100%, which means that the use of the catalyst oven had no significance during this part of the treatment.

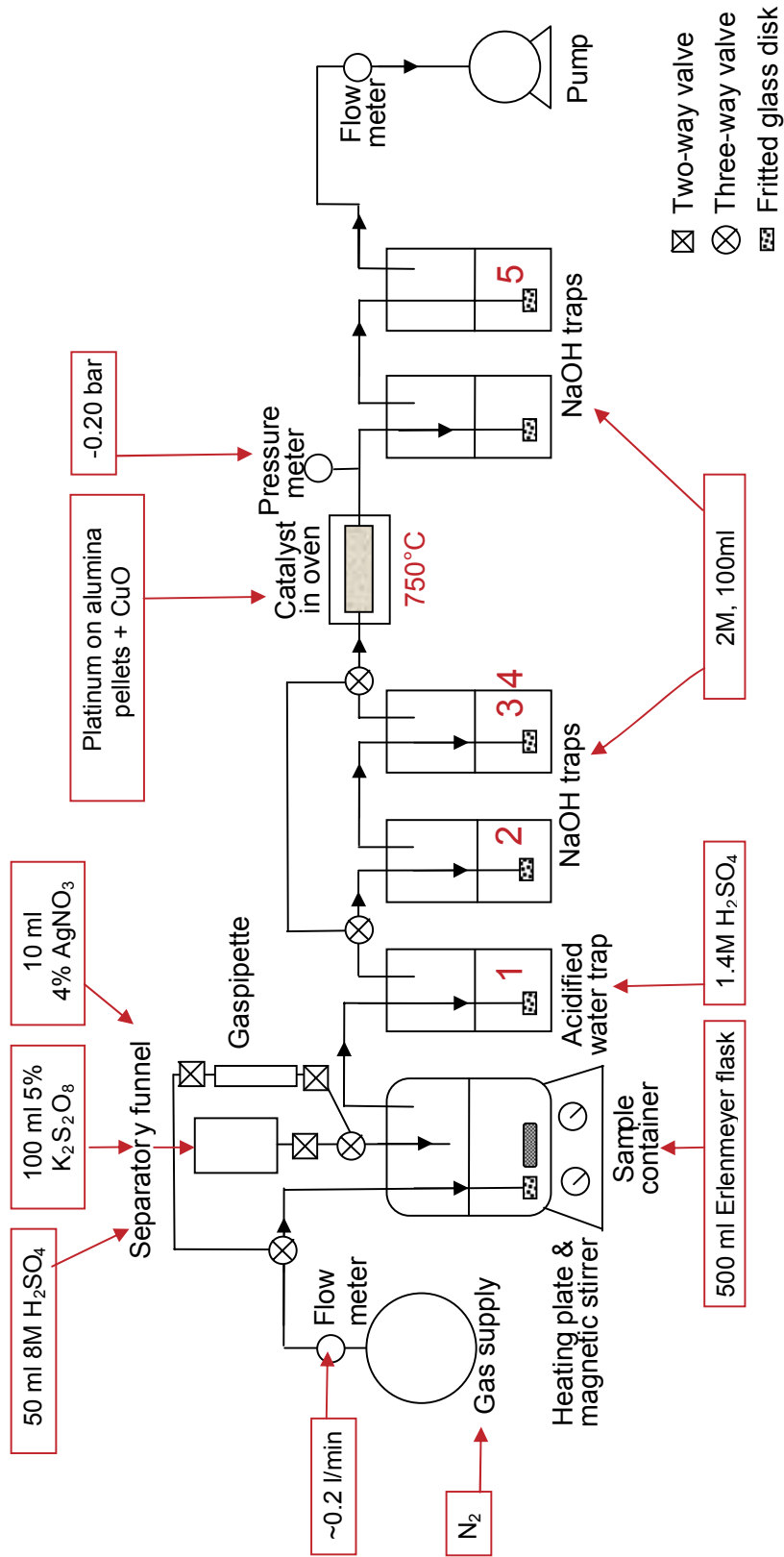


Figure 4-1. Schematic overview of the system used for extracting ¹⁴C from process water.

4.3 Results and discussion

4.3.1 PWR

The results from reactor R4 in Ringhals are presented in Table 4-2. Figure 4-2 displays the ^{14}C concentrations found before (313 samples) and after (334 samples) the coolant purification in the primary circuit. Figure 4-3 displays the ^{14}C concentrations found in various samples collected from the secondary circuit (spent fuel pool, waste water and steam generator blow down).

Sample 313:1 and 313:2 were collected at the same time (27th June) and both processed about 1 week later, showing a good agreement in the ^{14}C concentrations obtained (see Figure 4-2). However, the 334:1 sample collected at the same time has a higher concentration of inorganic ^{14}C than the 313-samples. The samples 313:3 and 334:3 were also collected and processed approximately at the same date (5th July). The lack of inorganic ^{14}C in the 334:3 is therefore unexpected. Sample 313:4 was processed 2 weeks after sampling, which partly might explain the high inorganic ^{14}C concentration found in the gas phase and consequently lower concentration found in the water phase due to degassing. However, a long delay before processing has not affected sample 334:2.

Table 4-2. The obtained results from the ^{14}C -activity measurements of process water from the PWR reactor R4, 2005. Total ^{14}C (Bq/kg) calculated from the measurements resulting in smallest relative error.

Reactor	Sample ID	Sampling date	Date of processing	Phase	Inorganic ^{14}C (Bq/kg)	σ_{relative} (%)	Detection limit (Bq/kg)	Organic ^{14}C (Bq/kg)	σ_{relative} (%)	Detection limit (Bq/kg)	Total ^{14}C (Bq/kg)	Organic fraction (%)
R4	313:1	27-jun	05-jul	Gas	90	10	13	13.4E+03	4.1	13	13.5E+03	99.3
				Water	59	17	18	12.1E+03	4.2	18	12.2E+03	99.5
				Gas	108	9.2	14	12.9E+03	4.1	14	13.0E+03	99.2
	313:2	27-jun	06-jul	Water	37	29	21	12.2E+03	4.1	22	12.2E+03	99.7
					36	6.7	0.74					
	313:3	05-jul	08-jul	Water	114	11	18	18.1E+03	4.1	18	18.2E+03	99.4
					55	23	22	13.5E+03	4.4	22	13.6E+03	99.6
	313:4	07-jul	22-jul	Gas	437	5.5	18	14.6E+03	4.1	18	15.0E+03	97.3
					411	6.5	0.48					
	313:4	07-jul	22-jul	Water	16	6.7	0.54	13.1E+03	4.1	21	13.1E+03	99.9
					177	8.7	21	11.7E+03	4.3	22	11.9E+03	99.5
	334:1	27-jun	07-jul	Gas	165	15	40	11.4E+03	4.2	41	11.6E+03	98.6
					165	6.5	0.95					
					175	15	40					
	334:2	05-jul	22-jul	Water	163	6.5	0.95	12.4E+03	4.3	41	12.5E+03	98.7
					163	6.5	0.95					
	334:3	05-jul	07-jul	Gas	< 1.1		< 1.1	18.1E+03	13	50	18.1E+03	100
					1.7	27	1.1	13.1E+03	13	50	13.1E+03	100
	337 _{before IX}	20-jul	21-jul	Water	< 0.34		< 0.34	4.3	8.5	0.38	< 4.6	> 92.6
	337 _{after IX}	20-jul	21-jul	Water	0.60	51	0.36	2.5	11	0.37	3.1	80.4
	324 _{before IX}	05-jul	06-jul	Water	36	29	21	7.6	7.6	0.85	46	16.4
					39	6.6	0.47					
	324 _{after IX}	05-jul	08-jul	Water	40	29	19	4.4	7.4	0.69	40	10.9
					36	6.5	0.58					
	T103 _{jun05}	30-jun	20-jul	Water	140	10	19	58	20	19	191	29.4
					135	6.5	0.52	56	6.5	0.44		

	LSC measurement liquid.
	LSC measurement precipitate.
	AMS measurement required.

From Figure 4-2 any decontamination by the ion exchange resins can not be seen with respect to neither inorganic nor organic ^{14}C .

The samples below the detection limit in the LSC measurements of precipitates have to be measured by Accelerator Mass Spectrometry (AMS).

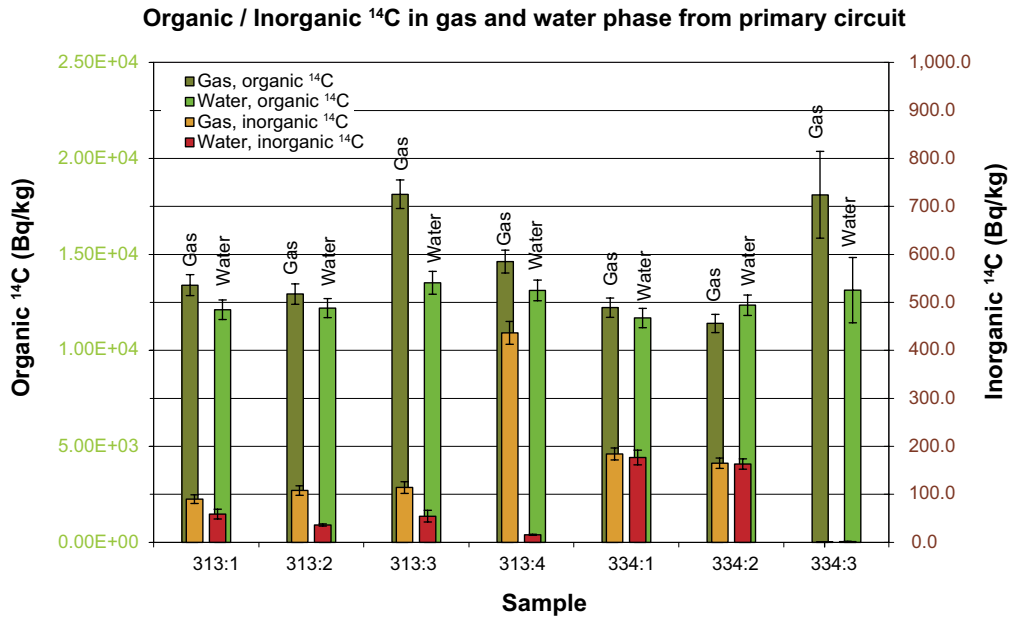


Figure 4-2. The organic and inorganic concentration of ^{14}C found in the gas- and water phase of process water collected before (samples 313) and after (samples 334) the primary coolant purification in the PWR reactor R4, 2005. Note the different y-scales.

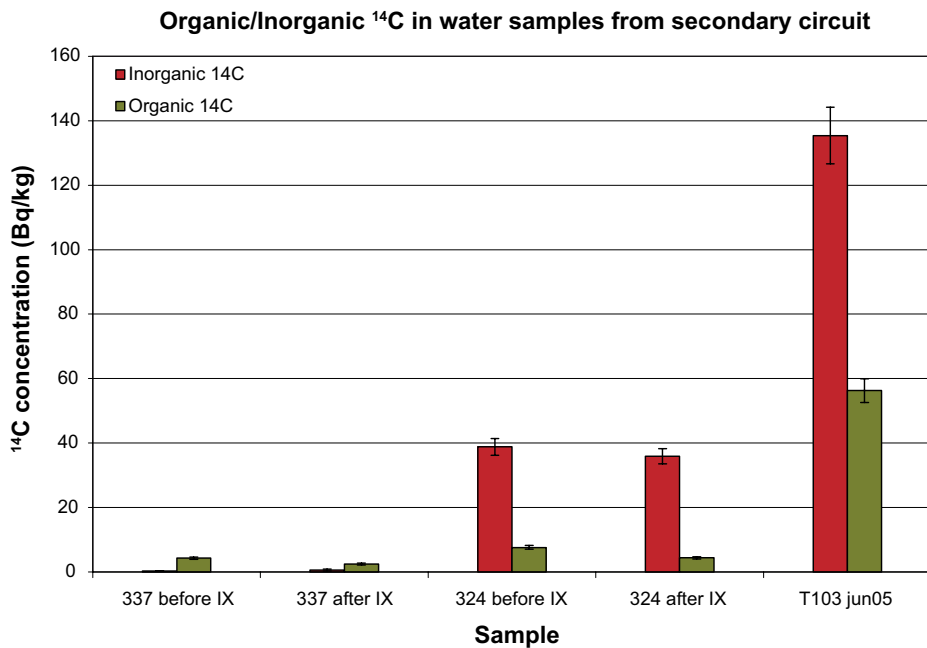


Figure 4-3. The organic and inorganic ^{14}C concentration found in samples of process water collected from various systems in the PWR reactor R4, 2005. The samples were taken before and after the ion exchangers in the steam generator blow-down circuit (337) and spent fuel pool (324); and from the tank of waste water for release (T103).

4.3.1.1 Chemical speciation PWR



About 0.3% of the ^{14}C in the primary coolant was in inorganic form. The organic ^{14}C found in the water samples analyzed, was found NOT to be volatile hydrocarbons or CO. Any hydrocarbons left in the water phase after degassing would have been found after the catalyst (hydrocarbons and CO are not absorbed in NaOH in reduced form) during the 1h of acid stripping. But no activity was ever found after the catalyst (n=6); i.e. the organic ^{14}C fraction in the water remained unaffected until the wet oxidation. If formic and acetic acid is present in the process water, only a small fraction would escape from the sample during the acid treatment (2.2–8.5% and 1.1–2.8%, respectively, according to earlier experiments), but effectively oxidized during the wet oxidation. The presence of these organic acids could therefore partly explain the findings. However, the pH value over the primary coolant resin is > 6.5 , which means that both formic- ($\text{pK}_a = 3.75$) and acetic acid ($\text{pK}_a = 4.75$) should be present as ionic species and readily attached to the resins.

4.3.2 BWR-HWC

The results from reactor B2 in Barsebäck are presented in Table 4-3. Generally, the concentration of organic ^{14}C is higher than the inorganic ^{14}C , which points to the fact that the organic ^{14}C species are less affected by resin uptake than the inorganic species. According to the values obtained, the organic fraction is $> 80\%$ in the 332-system and one third of the organic species in the condensate system is absorbed in the resins. However, this is a contradiction to the analyzed spent condensate resins which had only an organic fraction of $\sim 10\%$.

Table 4-3. The results obtained from the ^{14}C -activity measurements of process water from the BWR-HWC reactor B2, 2005. Total ^{14}C (Bq/kg) calculated from the measurements resulting in smallest relative error.

Reactor	Sample ID	Sampling date	Date of processing	Phase	Inorganic ^{14}C (Bq/kg)	σ_{relative} (%)	Detection limit (Bq/kg)	Organic ^{14}C (Bq/kg)	σ_{relative} (%)	Detection limit (Bq/kg)	Total ^{14}C (Bq/kg)	Organic fraction (%)
B2	321:1	24-May	21-Jun	Water	0.36	46	0.39	1.0	17	0.39	< 1.4	> 73
	321:2	25-May	22-Jun	Gas	< 0.29		0.29	< 0.28		0.28	< 0.57	
				Water	< 0.45		0.45	< 0.45		0.45	< 0.90	
	331	24-May	21-Jun	Water	0.72	30	0.46	1.0	20	0.46	1.7	58
	332 _{before IX}	24-May	20-Jun	Water	< 0.40		0.40	2.4	11	0.40	< 2.8	> 85
	332 _{after IX}	24-May	22-Jun	Water	< 0.38		0.38	1.6	12	0.38	< 2.0	> 80
	T25 _{mar05}	31-Mar	18-Jul	Water								
	T25 _{apr05}	30-Apr	18-Jul	Water	1.5	15	0.40	4.5	7.4	0.4	5.9	76
T25 _{may05}	31-May	18-Jul	Water									

 LSC measurement precipitate.
 AMS measurement required.

4.3.3 BWR-NWC

The results from reactor O3 in Oskarshamn are presented in Table 4-4. The duplicate samples taken from the reactor circuit do not fully agree. About 70% of the inorganic ¹⁴C is found in the gas phase. The sample collected after the condensate purification has a higher ¹⁴C concentration than the sample collected before the ion exchangers.

Table 4-4. The results obtained from the ¹⁴C-activity measurements of process water from the BWR-NWC reactor O3, 2005. Total ¹⁴C (Bq/kg) calculated from the measurements resulting in smallest relative error.

Reactor	Sample ID	Sampling date	Date of processing	Phase	Inorganic ¹⁴ C (Bq/kg)	σ_{relative} (%)	Detection limit (Bq/kg)	Organic ¹⁴ C (Bq/kg)	σ_{relative} (%)	Detection limit (Bq/kg)	Total ¹⁴ C (Bq/kg)	Organic fraction (%)
O3	321RB902:1	11-Jul	20-Jul	Water	0.36	47	0.30	2.7	8.5	0.32	3.1	88
	321RB902:2	11-Jul	20-Jul	Gas	0.79	17	0.23	0.39	29	0.22	1.2	33
				Water	< 0.32		0.32	1.4	14	0.33	< 1.7	> 81
	331RB901	11-Jul	19-Jul	Water	< 0.29		1.1	1.1	17	0.32	< 1.4	> 79
	332RB908	11-Jul	19-Jul	Water	0.68	23	0.34	0.48	41	0.36	< 1.2	41
	332RB909	11-Jul	19-Jul	Water	< 0.33		0.33	2.4	9.2	0.36	< 2.7	> 88

■ LSC measurement precipitate.

■ AMS measurement required.

4.3.3.1 Chemical speciation BWR

Since all of the samples were below the detection limit of the initial LSC measurement (liquid phase), no further information regarding the chemical characterization can be given.

4.4 Partial mass balance

By using the data obtained of total ¹⁴C activity in spent ion exchange resins and the activity concentrations measured in process water, the accumulated fraction (in terms of total and organic ¹⁴C) in the resins have been calculated. This has been done for the PWR reactors R2 and R3 and the BWR-HWC reactor B2. B2 was the only reactor from which both spent resins and process water were analyzed; for R2 and R3 it has been assumed that the ¹⁴C concentration in the reactor coolant is equal to R4. The results are presented in Table 4-5.

Table 4-5. Accumulated fraction of total and organic ¹⁴C in spent resins from R2, R3 (PWRs) and B2 (BWR-HWC), compared to the activity in water (non-gaseous) passing through the resins.

Reactor	Process water system	Chemical form of ¹⁴ C	¹⁴ C activity in H ₂ O (Bq·kg ⁻¹)	¹⁴ C activity in H ₂ O passing through IX ²⁸ (Bq·a ⁻¹)	¹⁴ C activity in spent resins (Bq·a ⁻¹)	¹⁴ C activity in IX / ¹⁴ C activity in H ₂ O
R3	313	Total	13,000	2.6E+12	2.7E+10	1%
		Organic	13,000	2.6E+12	7.8E+09	0.3%
R2	313	Total	13,000	2.6E+12	1.6E+10	0.6%
		Organic	13,000	2.6E+12	9.0E+09	0.2%
B2	321	Total	1.4	6.7E+08	3.1E+08	50%
		Organic	1.0	4.9E+08	1.9E+07	4%
	332	Total	2.8	7.3E+10	1.5E+09	2%
		Organic	2.4	6.2E+10	1.8E+08	0.3%

²⁸ Based on a flow rate of 7kg/s, 17 kg/s and 900 kg/s for the PWRs, B2 331-system and B2-332-system, respectively; and a reactor operation during 335 days (Aronsson 2005, personal communication).

In the PWRs, only a fraction of the ^{14}C in the reactor coolant is attached to the ion exchange resins, whereas the uptake in the BWR resins seems to be significantly higher. However, the calculated total uptake of 50% in system 331 (derived from the water concentration in 321) in B2 is based on the ^{14}C activity found in spent resins purifying both 331- and 342 water (i.e. the activity in the water may have been underestimated). The organic fraction bonded to the resins is generally very small. It should be noted that the activity concentrations in the process water from B2 are based on single measurements.

The samples collected from the waste water tanks in Ringhals and Barsebäck, may be used to estimate the fraction of waterborne discharges, relative to the calculated ^{14}C production in the reactor coolant. If the waste water sample collected from T103 in R4 (PWR) is assumed to be representative over the year, the annual ^{14}C activity released as waterborne discharges from this tank (based on an annual discharge rate of 4,000 m³ (Aronsson 2005, personal communication)) corresponds to 2.2‰ of the calculated production rate in the reactor coolant (average value over the years 2002–2004). Based on the same assumptions, the samples collected from T25 in B2 (BWR), correspond to a total annual release rate (based on an annual discharge of 12,000 m³ (Ernström 2005, personal communication)) of 0.3‰ of the calculated production rate in the reactor coolant.

5 Ejector off-gas samples

5.1 Sampling and sample description

Off-gas samples were collected from the PWR reactor R4 and the BWR-NWC reactor O3 in July 2005, see Table 5-1. From R4 two samples of each 1 liter were collected in Marinelli beakers and from O3, two samples of 10 ml in glass bottles. The two samples from R4 represent ejector off-gas from turbine 1 and 2, where no detectable amounts of ^{14}C would be present during normal reactor operation. However, during 2005, R4 was operated with leaking steam generators, i.e. primary coolant was able to get to the secondary circuit. The two samples from O3 represent off-gas before and after the charcoal columns in the delay system for gas releases.

Table 5-1. Description of off-gas samples collected in 2005 from a PWR and BWR-NWC reactor.

Reactor type	System	Description	No. of samples
PWR (R4)	T41	Ejector off-gas from turbine 1	1
	T42	Ejector off-gas from turbine 2	1
BWR-NWC (O3)	552RB901	Off-gas before charcoal column	1
	552RB903	Off-gas after charcoal column	1

5.2 Method

The system setup resembled the one previously described for spent resins and process water, but with the gas containers directly connected between the N_2 -container and the first gas washing bottle (see e.g. Figure 3-1). The separation of organic and inorganic ^{14}C -compounds was accomplished by the catalyst so that CO_2 would be absorbed in the alkaline washing bottles before the catalyst; and the hydrocarbons and CO in the bottles situated after the catalyst.

5.2.1 Procedure for ^{14}C extraction

The off-gas samples contained in Marinelli beakers were directly connected to two sets of alkaline (2M NaOH) gas washing bottles. During the first part of the gas extraction (~ 5 minutes), a slightly sub-atmospheric pressure was applied over the washing bottles to gently force the gas out of the beaker. The rest of the gas was driven out by purging the beaker with N_2 using an initial flow rate of < 0.2 l/min, which was increased to 0.2 l/min after 15 minutes. Total time of extraction was 1 hour.

5.2.2 Analysis

The processing of samples and LSC measurements were conducted at Ringhals, using the previously described sample preparation and equipment (see Section 4.2.2). All samples were initially measured 3 times by LSC with a detection limit of 1.6 and 190 kBq/m³ for 1 liter and 10 ml samples, respectively. Samples below the detection limit were precipitated as BaCO₃ and the precipitate remeasured by LSC giving an improved detection limit of 0.040 and 5 kBq/m³, respectively.

5.3 Results and discussion

5.3.1 PWR – Mass balance with steam generator leakages

The results obtained from the ejector off-gas samples collected from the PWR reactor R4 in Ringhals are presented in Table 5-2. Approximately 97% of the ¹⁴C is released in reduced form. The ¹⁴C concentration is significantly different in the two samples from turbine 1 (T41) and 2 (T42), respectively. Since the concentration of noble gases in off-gas analyzed from T41 generally is 60% of the concentration in T42 (Aronsson 2005, personal communication), the same relation is expected for ¹⁴C. However, the ¹⁴C concentration in T41 is only 10% of the activity detected in T42. The difference might be explained by a failure during sampling or during the gas extraction.

In Table 5-3 a mass balance over the leaking steam generator tubes is presented. The ejector flow-rates and the steam generator leak rates have been collected from the Process Information System (PIS). Assuming equilibrium between the leakage in the steam generator tubes and the flow-rate in the ejector off-gases, the concentration of gaseous ¹⁴C-species in the reactor coolant has been calculated. The concentration of the organic species lies in the middle of the range of concentrations reported in Table 4-2 (see sample ID 313:X). The calculated concentration of inorganic, gaseous ¹⁴C-species is in the upper end of the range reported in Table 4-2. However, it should be noted that the water samples (including gas phase) from the primary circuit (system 313) were collected 2–3 weeks before the off-gas sample.

By assuming constant ¹⁴C concentration, leak- and flow rates throughout the year, the total amount of ¹⁴C released as gas due to the leakage through the steam generators is 0.47 GBq/a. This corresponds to 2.3‰ of the reported (Arvidsson 2005, personal communication) average airborne ¹⁴C releases during 2002–2004 and 1.4‰ of the calculated ¹⁴C production in the reactor coolant.

Table 5-2. The concentration of ¹⁴C in ejector off-gas samples collected from turbine 1 (T41) and 2 (T42) in the PWR reactor R4, 2005. Total ¹⁴C (Bq/m³) calculated from the measurement resulting in smallest relative error.

Reactor	Sample ID	Sampling date	Date of processing	Inorganic ¹⁴ C (Bq/m ³)	σ _{relative} (%)	Detection limit (Bq/m ³)	Organic ¹⁴ C (Bq/m ³)	σ _{relative} (%)	Detection limit (Bq/m ³)	Total ¹⁴ C (Bq/m ³)	Organic fraction (%)
R4	4-T41	20-Jul	21-Jul	< 34		34	445	8.9	36	< 479	> 93.0
	4-T42	20-Jul	21-Jul	125	14	34	3.68E+03	23	1.6E+03	4.30E+03	97.1

	LSC measurement liquid.
	LSC measurement precipitate.
	AMS measurement required.

Table 5-3. Concentration of gaseous ¹⁴C in the reactor coolant, calculated from analyses in the ejector off-gas samples from T42, recalculated to the total steam generator leak rate in the PWR reactor R4, 2005. Raw data taken from PIS (Process Information System).

T41 Leak rate (kg/h)	T42 Leak rate (kg/h)	T42 Ejector flow (m ³ /h)		T42 Concentration (Bq/m ³)	T42 Activity flow (Bq/s)	T41+T42 Activity flow (Bq/s)	RC Concentration (Bq/kg)	Annual release (Bq)
1.54	2.21	8.2	Inorganic	1.25E+02	2.85E-01	4.83E-01	4.64E+02	1.37E+07
1.54	2.21	8.2	Organic	4.17E+03	9.50E+00	1.61E+01	1.55E+04	4.57E+08
			Sum	4.30E+03	9.78E+00	1.66E+01	1.59E+04	4.70E+08

The concentration of ¹⁴C in the water phase (i.e. non-gaseous species) in the reactor coolant have been calculated using data obtained for the 337-samples (steam generator blow-down), presented in Table 4-2. Since the inorganic fraction in the samples was below the detection limit, the calculation has only been done for the organic fraction of ¹⁴C. Assuming equilibrium between the leak and the uptake on the blow-down ion exchange resins, the concentration of organic ¹⁴C in the reactor coolant may be calculated from the leakage- and flow rates and uptake in the 337 resins. The result of this calculation is shown in Table 5-4.

Table 5-4. Concentration of non-gaseous ¹⁴C in the reactor coolant, calculated from analyses in the blow-down system 337 in the PWR reactor R4, 2005. The blow-down flow rate was 5.6 kg/s (PIS data base).

	Steam generator Total leak-rate (kg/h)	Before 337 IX Concentration (Bq/kg)	After 337 IX Concentration (Bq/kg)	Uptake on 337 IX Concentration (Bq/kg)	RC Concentration (Bq/kg)
Organic	3.75	4.3	2.5	1.8	9.68E+03

The concentration obtained is about 80% of the values reported for the organic fraction in the water phase of the 313-samples in Table 4-2. This lower value may be explained by another phenomenon called hide-out (enrichment on the heat transfer surfaces) which acts as an additional clean-up flow.

5.3.2 BWR-NWC

The results obtained from the off-gas samples collected before (552RB901) and after (552RB903) the charcoal columns in the BWR-NWC reactor O3 in Oskarshamn are presented in Table 5-5. Approximately 98% of the ¹⁴C after the columns is in oxidized form. The concentration of ¹⁴CO₂ is 85% lower after the charcoal column, whereas the reduced forms are adsorbed to some 40%. The high retention indicates that there has been no breakthrough of ¹⁴CO₂ in the columns.

Since the columns are operated in a mode with regular back-flushings, it is difficult to compare the outflow from the columns with the releases measured in the stack. However, in Table 5-6 below, a comparison with the calculated production rate /Lundgren et al. 2002/ is shown, which seems to be in good agreement with the calculated in-flow to the columns. This indicates that there was no back-flushing of another column going on at the time of the sample collections, since the concentration in 552RB901 in that case might have been significantly higher than the production rate. To characterize the dynamics of adsorption of ¹⁴C on the charcoal columns, further measurements are needed.

Table 5-5. The concentration of ^{14}C in off-gas samples collected before (552RB901) and after (552RB903) the charcoal columns in the BWR-NWC reactor O3, 2005. Total ^{14}C (Bq/m^3) calculated from the measurements resulting in smallest relative error.

Reactor	Sample ID	Sampling date	Date of processing	Inorganic ^{14}C (Bq/m^3)	σ_{relative} (%)	Detection limit (Bq/m^3)	Organic ^{14}C (Bq/m^3)	σ_{relative} (%)	Detection limit (Bq/m^3)	Total ^{14}C (Bq/m^3)	Organic fraction (%)
O3	552RB901	11-Jul	22-Jul	3.37E+06	6.5	1.9E+05	1.90E+04	12	4.1E+03	3.21E+06	0.6
				3.19E+06	6.7	5.1E+03					
	552RB903	11-Jul	21-Jul	5.4E+05	22	1.9E+05	1.90E+04	20	3.7E+03	4.85E+05	2.2
				4.74E+05	6.8	5.1E+03					

LSC measurement liquid.
 LSC measurement precipitate.

Table 5-6. In- and out-flow of ^{14}C to the charcoal columns, compared with calculated production rate. Concentration values (Bq/m^3) taken from the measurements resulting in the smallest relative error. Flow rate through columns (m^3/s) from (Hjelm 2005, personal communication).

	Calculated production rate (Bq/s)	552RB901 (Bq/s)	552RB903 (Bq/s)	Retention (%)
Inorganic		2.06E+04	2.90E+03	86
Organic		1.16E+02	6.66E+01	43
Sum	2.20E+04	2.07E+04	2.96E+03	86

6 References

Bergström U, Puigdomenech I, 1987. “Radiological consequences to man due to leakage from a final repository for reactor waste (SFR)” SKB Progress Report SFR-87-12. Svensk Kärnbränslehantering AB.

Gruhlke J M, Neiheisel J, Battist L, 1986. ”Estimates on the quantities, form, and transport of carbon-14 in low-level radioactive waste” EPA-520/1-86-019.

Hesböl R, Puigdomenech I, Evans S, 1990. “Source terms, isolation and radiological consequences of carbon-14 waste in the Swedish SFR repository” SKB TR 90-02. Svensk Kärnbränslehantering AB.

Ingemansson T, 2001. “Nuklidinventariet i SFR-1” SSI Report 2001:23.

Kunz C, 1985. “Carbon-14 discharge at three light-water reactors” Health Physics 49:1, 25–350.

Lundgren K, 2005. “Ringhals 4 – FSAR Topical Report – Neutron activation of reactor coolant and containment atmosphere” Ringhals doc. id 1881412, preliminary report.

Lundgren K, Ingemansson T, Widmark G, 2002. “Carbon-14 in Nordic BWRs- Production and chemical forms” SSI P 1294.01.

Magnusson Å, Stenström K, 2005. “Determination of organic and inorganic ¹⁴C on ion exchange resins – method description”, Internal Report LUNFD6/(NFFR-3097), Lund University.

Martin J E, 1986. “Carbon-14 in low-level radioactive wastes from two nuclear power plants” Health Phys. 50, 57–64.

Matsumoto J, Banda T, Muraoka S, 1995. “Adsorption of carbon-14 on mortar” Scientific basis for nuclear waste management XVIII, Eds T. Murakami and R.C. Ewing, Materials research society symposium proceedings 353, p 1029–1035.

Salonen L, Snellman M, 1985. “Carbon-14 releases from Finnish nuclear power plants” Final report of research agreement no. 3065/R2/CF. Part of the IAEA coordinated program “On carbon-14 from nuclear facilities”.

Vance J N, Cline J E, Robertson D E, 1995. “Characterization of Carbon-14 generated by the nuclear power industry”, EPRI TR-105715, EPRI, Palo Alto.

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PWR

Concentration of ^{14}C and ^{60}Co in spent resin samples- originating from a variety of cleaning systems – from PWR reactors in Ringhals. All ^{14}C -results presented are from the LSC measurements after the chemical treatment, i.e. not corrected for incomplete extraction of organic ^{14}C .

IX 1: R3 WPATRS

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
R-1:1	1,09E+07	3,8%	1,83E+03	4,84E+06	4,0%	1,90E+03	31%	1,57E+07	3,67E+07	
R-1:2	1,56E+07	3,6%	1,35E+03	5,67E+06	3,6%	1,41E+03	27%	2,12E+07	4,95E+07	
R-1:3	1,77E+07	3,6%	1,94E+03	6,56E+06	3,6%	2,01E+03	27%	2,43E+07	5,66E+07	2,13E+07 ±
R-1:4	1,32E+07	3,6%	2,09E+03	5,77E+06	3,6%	2,17E+03	30%	1,90E+07	4,42E+07	1E+05
R-1:6*	1,18E+07	3,6%	6,00E+02	4,58E+06	3,8%	6,36E+02	28%	1,64E+07	3,82E+07	
R-1:8*	1,44E+07	3,8%	5,83E+02	5,51E+06	3,8%	6,17E+02	28%	1,99E+07	4,65E+07	
Average	1,39E+07			5,49E+06			28%	1,94E+07	4,53E+07	
Std deviation	18%			13%			2%	16%	16%	
R-1:5**	2,19E+07	3,6%	2,24E+03	8,64E+06	3,6%	2,33E+03	28%	3,06E+07	7,13E+07	
R-1:7***	2,34E+07	3,6%	1,30E+03	8,04E+06	3,7%	1,33E+03	26%	3,15E+07	7,34E+07	

IX 2: R2 SRST

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
R-2:1	5,00E+06	3,6%	1,37E+03	1,79E+06	3,8%	1,43E+03	26%	6,79E+06	1,58E+07	
R-2:2	6,97E+06	3,6%	2,18E+03	2,17E+06	3,7%	2,26E+03	24%	9,13E+06	2,13E+07	
R-2:3	7,16E+06	3,6%	1,41E+03	2,12E+06	3,7%	1,47E+03	23%	9,28E+06	2,16E+07	9,86E+07 ±
R-2:4	7,25E+06	3,6%	2,00E+03	2,04E+06	3,7%	2,61E+03	22%	9,30E+06	2,17E+07	3E+05
R-2:5	6,57E+06	3,6%	1,75E+03	2,59E+06	3,7%	1,82E+03	28%	9,17E+06	2,14E+07	
R-2:6	3,06E+06	3,8%	2,13E+03	1,85E+06	4,1%	2,21E+03	38%	4,91E+06	1,14E+07	
Average	6,00E+06			2,09E+06			27%	8,10E+06	1,89E+07	
Std deviation	28%			14%			6%	23%	23%	

IX 3: R4 WPATRS

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
R-3:1	1,11E+06	3,7%	1,95E+03	4,37E+05	3,8%	2,02E+03	28%	1,55E+06	3,60E+06	
R-3:2	1,20E+06	3,7%	2,02E+03	6,34E+05	3,8%	2,10E+03	35%	1,83E+06	4,27E+06	7,21E+07 ±
R-3:3	1,04E+06	3,8%	2,16E+03	5,68E+05	3,8%	2,24E+03	35%	1,61E+06	3,75E+06	7E+05
R-3:4	9,37E+05	3,9%	1,73E+03	5,19E+05	3,8%	1,80E+03	36%	1,46E+06	3,39E+06	
Average	1,07E+06			5,39E+05			33%	1,61E+06	3,75E+06	
Std deviation	10%			15%			4%	10%	10%	

IX 4: R3 WPATRS

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
R-4:1	8,59E+05	3,7%	1,36E+03	3,80E+05	3,8%	1,42E+03	31%	1,24E+06	2,89E+06	
R-4:2	9,99E+05	3,7%	1,15E+03	4,14E+05	3,7%	1,20E+03	29%	1,41E+06	3,29E+06	4,92E+07 ±
R-4:3	1,20E+06	3,7%	1,23E+03	4,69E+05	3,7%	1,27E+03	28%	1,67E+06	3,90E+06	3E+05
R-4:4	9,30E+05	3,7%	1,16E+03	3,36E+05	3,8%	1,20E+03	27%	1,27E+06	2,95E+06	
Average	9,97E+05			4,00E+05			29%	1,40E+06	3,26E+06	
Std deviation	15%			14%			2%	14%	14%	

* Subsample processed half a year later than the others

** Subsample contained approximately 50% transparent beads, 25% anionic beads (white) and 25% cationic beads (dark)

*** Subsample contained nearly 100% transparent beads

BWR-HWC

Concentration of ^{14}C and ^{60}Co in spent resin samples from BWR-HWC reactors in Barsebäck and Oskarshamn. Batch samples IX 1 and IX 2 are spent resins from the condensate purification system (332) in reactor B2; IX 3 from the spent fuel pool (324), representing O1 and O2; and IX 4 from the reactor water clean-up (331) and waste processing (342) in B2. All ^{14}C -results presented are from the LSC measurements after the chemical treatment, i.e. not corrected for incomplete extraction of organic ^{14}C .

IX 1: 332

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
B-1:1	1,00E+05	4,0%	4,14E+02	1,29E+03	19%	4,68E+02	1,3%	1,01E+05	2,71E+05	4,68E+06 ± 1E+04
B-1:2	9,30E+04	3,9%	5,08E+02	1,19E+04	5,9%	6,64E+02	11%	1,05E+05	2,80E+05	
Average	9,65E+04			6,59E+03			6,3%	1,03E+05	2,76E+05	
Std deviation	5,1%			100%			7%	2,5%	2,5%	

IX 2: 332

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
B-2:1	1,42E+05	3,8%	5,76E+02	1,47E+04	5,7%	7,54E+02	9,4%	1,57E+05	3,37E+05	3,33E+06 ± 1E+04
B-2:2	7,84E+04	4,1%	6,45E+02	1,42E+04	7,4%	8,44E+02	15%	9,26E+04	1,99E+05	
Average	1,10E+05			1,45E+04			12%	1,25E+05	2,68E+05	
Std deviation	4,1%			2,60%			4%	36%	36%	

IX 3: Oskarshamn 324

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
B-3:1	< 8,42E+02	—	8,42E+02	<1,07E+03	—	1,07E+03	—	<1,91E+03	<5,11E+03	8,65E+08 ± 2E+06
B-3:2	<3,72E+02	—	3,72E+02	9,47E+02	32%	4,81E+02	>72%	<1,32E+03	<3,53E+03	

IX 4: 331-342

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
B-4:1	6,88E+04	4,0%	5,07E+02	6,78E+03	7,9%	6,55E+02	9,0%	7,56E+04	2,08E+05	1,025E+08 ± 1E+05
B-4:2	7,18E+04	4,1%	6,56E+02	2,10E+03	22%	8,32E+02	2,8%	7,39E+04	2,03E+05	
Average	7,03E+04			4,44E+03			5,9%	7,47E+04	2,06E+05	
Std deviation	3,0%			75%			4%	1,6%	1,6%	

BWR-NWC

Concentration of ^{14}C and ^{60}Co in spent resin samples from BWR-NWC reactors in Forsmark. Batch sample IX 1 represent spent resins from the reactor water clean-up (331); IX 2 from the condensate purification system (332); IX 3 from the spent fuel pool; and IX 4 from waste processing. All ^{14}C -results presented are from the LSC measurements after the chemical treatment, i.e. not corrected for incomplete extraction of organic ^{14}C .

IX 1: 331

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
F-1:1	5,28E+04	4,4%	7,31E+02	7,16E+03	9,6%	7,58E+02	12%	6,00E+04	1,36E+05	1,102E+08
F-1:2	5,48E+04	4,2%	7,99E+02	2,84E+03	16,4%	8,29E+02	4,9%	5,76E+04	1,31E+05	$\pm 4\text{E}+05$
Average	5,38E+04			5,00E+03			8,4%	5,88E+04	1,34E+05	
Std deviation	2,6%			61%			5%	2,8%	2,8%	

IX 2: 332

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
F-2:1	7,63E+05	3,6%	4,70E+02	3,21E+04	4,5%	4,88E+02	4,0%	7,95E+05	2,30E+06	7,79E+06 \pm
F-2:2	8,01E+05	3,7%	5,49E+02	2,46E+04	4,9%	5,31E+02	3,0%	8,26E+05	2,39E+06	5E+04
Average	7,82E+05			2,84E+04			3,5%	8,11E+05	2,34E+06	
Std deviation	3,4%			19%			1%	2,7%	2,7%	

IX 3: 324

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
F-3:1	4,34E+05	3,7%	6,75E+02	2,41E+04	4,9%	7,01E+02	5,3%	4,59E+05	1,33E+06	6,0E+06 \pm
F-3:2	3,98E+05	3,7%	6,02E+02	2,46E+04	5,1%	6,01E+02	5,8%	4,23E+05	1,22E+06	1E+05
Average	4,16E+05			2,44E+04			5,5%	4,41E+05	1,27E+06	
Std deviation	6,2%			1,3%			0,4%	5,7%	5,7%	

IX 4: 342

Subsample	Inorganic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic ^{14}C (Bq/kg ww)	σ_{relative}	Detection limit (Bq/kg ww)	Organic fraction	Total ^{14}C (Bq/kg ww)	Total ^{14}C (Bq/kg dw)	Co-60 (Bq/kg ww)
F-4:1	8,18E+04	4,0%	5,54E+02	2,69E+03	14,0%	4,05E+02	3,2%	8,45E+04	2,16E+05	1,107E+08
F-4:2	5,48E+04	4,1%	5,50E+02	3,15E+03	12,0%	5,71E+02	5,4%	5,79E+04	1,48E+05	$\pm 4\text{E}+05$
Average	6,83E+04			2,92E+03			4,3%	7,12E+04	1,82E+05	
Std deviation	28%			11%			2%	26%	26%	