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Abstract Radiochemical procedures were developed for the analysis of plutonium in primary cooling water and in ion-exchange resin. Analyses of cooling-water samples from unit 2 of the Ringhals nuclear power station demonstrated that a major fraction of the plutonium found in the cooling water is associated with particles. The amounts of plutonium isotopes retained annually in the ion-exchange cleaning system are estimated at 3 MBq of ^{238}Pu and 2 MBq of $^{239+240}Pu$ and in a mechanical filter the amounts are estimated at 2 MBq of ^{238}Pu and 0.6 MBq of $^{239+240}Pu$.

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Contents

1 Introduction

The presence of transuranic radionuclides in the primary cooling water of nuclear power reactors may occur due to leaking fuel elements and so-called tramp uranium on the external surfaces of new fuel elements. These radionuclides are collected in the cleaning systems of the primary cooling circuits, and the longlived transuranic radionuclides may be of concern in connection with the ultimate disposal of radioactive waste from these systems.

This report contains the results of a project made for SKB (Swedish Nuclear Fuel and Waste Management Co.) with the objective of developing methods to determine transuranic radionuclides in the primary cooling water of a nuclear power reactor and to estimate the efficiencies of the ion-exchange system and a mechanical filter to remove these radionuclides from the water and to estimate the annual inventories of transuranics in these devices. Unit 2 (PWR) of the Ringhals power plant was investigated in this study which was carried out in the period 1 July to 31st December 1991.

The present investigation deals with analyses of the alpha-particle emitting plutonium isotopes ^{238}Pu, ^{239}Pu and ^{240}Pu. Due to the inability through alpha spectrometry to distinguish between ^{239}Pu and ^{240}Pu, the sum of the activities of these two isotopes $^{239+240}$Pu is determined. These two isotopes occur in the fuel of light-water power reactors at a burn-up of 18 MWd (kg U)-1 in approximately equal amounts of activity (becquerels) and in about one third of the amount of ^{238}Pu activity (Hedemann Jensen et al. 1977).

2 Analytical Procedures

The project plan envisaged the analysis of plutonium in samples from the primary cooling water collected before and after the ion-exchange system and before and after the mechanical filter as well as direct determinations of plutonium in samples from the ion-exchange system.

The development of radiochemical procedures for the determination of plutonium in samples of cooling water and ion-exchange resin were based on previous experience with analysis of plutonium in environmental samples (Chen et al. 1991).

2.1 Radiochemical Procedures

Radiochemical procedures were developed for the analysis of plutonium in cooling-water samples and in samples of ion-exchange resin. The procedures are based on the principle of controlled valence of the plutonium ions. Tests of the procedures have yielded chemical recoveries in the range from 64% to 100%. The radiochemical procedures are outlined in detail in the appendix.

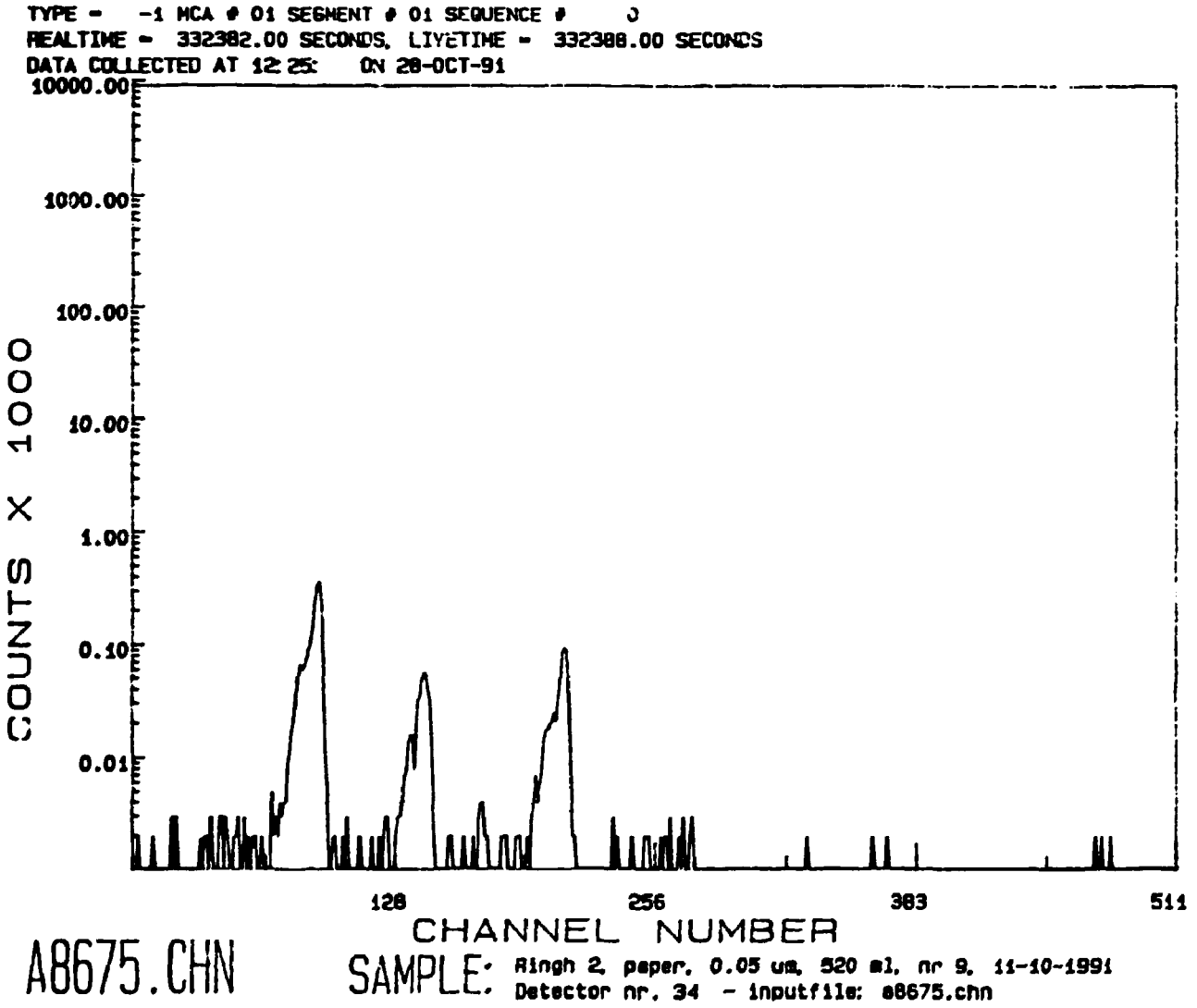
2.2 Alpha-Spectrometry Procedures

The samples are electroplated on stainless steel discs with a diameter of 19 mm; the active area has a diameter of 16 mm. The samples are mounted in vacuum chambers under silicon detectors (surface barrier or ion implanted types) with active areas corresponding to a diameter of 12 mm. The source-detector distance is approximately 3 mm. The detectors are calibrated with sources made from standard solutions purchased from recognised vendors. The sources are prepared according to a technique outlined by Chen et al. (1989). The detector efficiencies range from 29% to 39%. Intercomparison of results for plutonium analyses with other laboratories (Harwell in Great Britain and Physikalisch-Technische Bundesanstalt in Germany) have yielded good agreement.

The samples are counted for 72 hours or more, and background counts made on blanks are recorded for similar time periods. The alpha spectra are evaluated by observing the total counts in the energy intervals for the relevant isotopes

which cover the chemical-yield tracer (^{242}Pu or ^{236}Pu), $^{239+240}$Pu and ^{238}Pu. All alpha spectra are inspected visually to check for the proper selection of the energy intervals according to conditions of energy resolution and possible gain drift over the counting period. A typical alpha spectrum is shown in Figure 1. The results are evaluated on personal computers where the data are entered for a spread-sheet calculation. Detection limits are calculated from the background counts in the appropriate counting intervals by using a confidence factor of 4.65 to the standard deviation of the background count to obtain a 95% confidence level (Currie 1968).

Figure 1. Example of alpha spectrum showing three peaks of (from left to right) the yield determinant ^{242}Pu, $^{239+240}$Pu and ^{238}Pu.



3 Results of Pu Analyses

3.1 Sampling Programme

Samples of cooling water from the primary circuit of unit 2 of the Ringhals power plant were collected before and after the ion-exchange system to obtain data on the plutonium levels at these points. The sampling point before the ion-exchange system is referenced 'RC-2', and the sampling point after the ion-exchange system is referenced as '2-CVCSOUT'. From these points sampling of cooling water was carried out from 17 June to 2 December. The sampling volumes started out by 0.1 litre but were later increased to 1 litre in order to obtain a higher sensitivity.

During the project it turned out that it would not be possible to obtain samples of ion-exchange resin due to prohibitive high radiation doses to staff performing the collection. For that reason it was decided to increase the number of cooling-water samples to improve the data collection for an indirect assessment of the plutonium inventory in the ion-exchange system.

The preliminary results of plutonium in the cooling water indicated that a significant proportion of the plutonium activity is associated with particles and for that reason it was decided to include in the project an evaluation of the efficiency of a mechanical filter to remove plutonium from the cooling water. It was not possible to obtain cooling-water samples collected after the mechanical filter. Instead an indirect evaluation of the efficiency of the mechanical filter was planned by simulating in the laboratory the filtration of cooling water through membrane filters of approximately the same pore size (5 μm) as that of the mechanical filter (6 μm) and analyzing the two plutonium fractions: that on the filter and that in the filtrate solution.

In order to obtain additional information on the size distribution of the particles associated with plutonium, the cooling-water samples were filtrated through membrane filters of varying pore sizes.

A total of 26 samples were collected at sampling point RC-2 before the ion-exchange system and 29 samples were collected at 2-CVCSOUT after the ion-exchange system. Of these 55 samples 38 were filtrated and thus analyzed both for filter fraction and filtrate solution. The results of the analyses are given in full detail in the appendix.

3.2 Results of Pu in Cooling-Water Samples

The resulting concentrations of ^{238}Pu and $^{239+240}Pu$ in the cooling-water samples are given in the Tables 1 and 2 for the two sampling points RC-2 and 2-CVCSOUT, respectively. The tables give the dates of collection of the samples, the sample volumes, and the resulting concentrations of plutonium in millibecquerels per litre. The tables include the arithmetic mean values for the plutonium concentrations. The counting uncertainties are given in terms of relative standard deviations. The results are also shown in graphical form in Figures 2 and 3.

Table 1. Results of plutonium concentrations in cooling water (millibecquerels per litre) sampled before the ion-exchange system (RC-2) at Ringhals unit 2.

Date	Volume (ml)	Pu-238 (mBq/l)	SD (%)	Pu-239+240 (mBq/l)	SD (%)
17 Jun	100	128.00	6	34.00	7
18 Jun	100	12.00	8	4.00	11
19 Jun	100	21.00	7	5.60	10
24 Jun	100	45.00	7	22.00	8
26 Jun	150	20.00	7	7.10	7
26 Jun	100	401.00	6	300.00	6
28 Jun	100	2.60	19	0.70	32
02 Jul	100	< 1.3		< 0.9	
02 Jul	100	< 1.0		0.70	41
03 Jul	50	7.40	23	15.00	14
03 Jul	47	< 3.4		3.20	25
03 Jul	100	2.60	17	0.60	31
25 Sep	375	27.00	9	12.19	12
26 Sep	390	17.58	12	5.30	20
30 Sep	500	7.17	18	3.66	23
02 Oct	550	9.02	8	3.64	11
07 Oct	500	4.83	14	1.81	22
08 Oct	550	8.14	9	3.87	11
22 Oct	520	2.51	11	1.08	16
23 Oct	520	2.94	11	1.13	15
24 Oct	530	8.43	9	3.76	13
25 Oct	545	8.41	9	3.13	15
28 Oct	555	3.86	12	1.09	23
29 Oct	520	9.77	11	4.31	15
30 Oct	530	11.92	9	5.04	12
01 Nov	530	6.83	8	5.75	8
Mean		33.4		17.9	
Median		8.3		4.2	

Table 2. Results of plutonium concentrations in cooling water (millibecquerels per litre) sampled after the ion-exchange system (2-CVCSOUT) at Ringhals unit 2.

Date	Volume (ml)	Pu-238 (mBq/l)	SD (%)	Pu-239+240 (mBq/l)	SD (%)
24 Jun	100	7.20	10	3.30	13
26 Jun	100	1.70	25	1.20	25
28 Jun	100	4.20	13	1.30	24
02 Jul	100	8.20	11	4.90	13
03 Jul	100	75.00	6	24.00	8
09 Oct	550	3.19	13	1.97	16
10 Oct	550	14.60	7	6.09	9
11 Oct	520	11.65	7	7.47	7
14 Oct	550	277.85	6	43.56	6
15 Oct	520	1.74	24	< 1.3	
16 Oct	550	2.66	10	1.16	14
31 Oct	530	5.82	13	2.59	20
04 Nov	580	3.25	10	0.87	18
05 Nov	520	1.60	18	0.63	29
06 Nov	580	1.18	20	0.44	29
07 Nov	580	10.55	8	3.97	10
08 Nov	540	8.40	9	4.15	12
11 Nov	590	3.39	11	0.99	21
12 Nov	545	6.12	9	2.09	14
19 Nov	1025	6.33	8	2.39	11
20 Nov	1080	2.73	11	0.99	18
21 Nov	1125	1.03	18	0.30	34
22 Nov	1100	1.06	19	0.30	37
25 Nov	1100	0.81	19	0.14	41
26 Nov	1125	2.70	10	0.97	16
27 Nov	1120	2.50	11	1.42	14
28 Nov	1130	2.00	13	0.96	23
29 Nov	1100	1.55	11	0.65	19
02 Dec	1115	19.70	6	6.27	8
Mean		16.9		4.45	
Median		3.25		1.30	

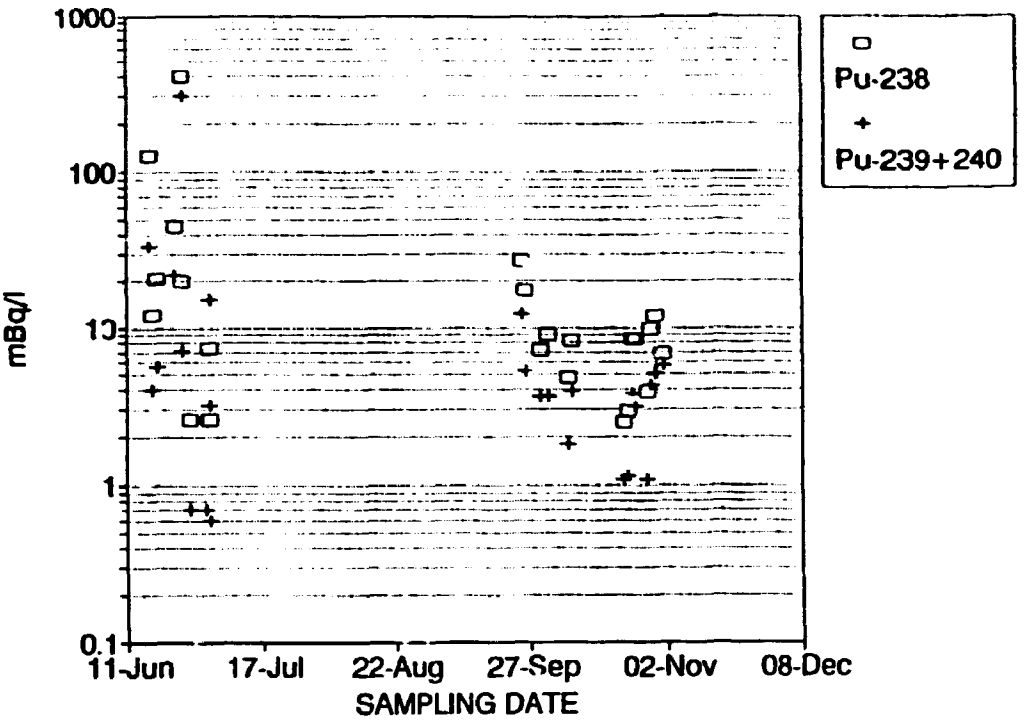
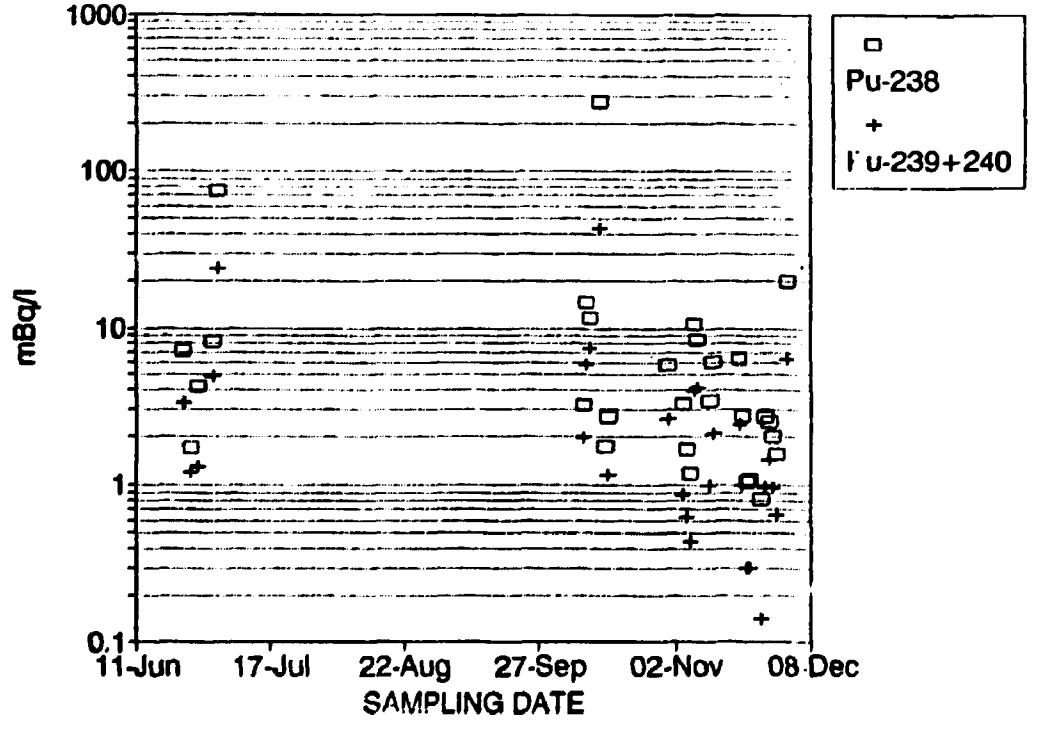


Figure 2. Plot of plutonium concentrations in cooling water sampled before the ion-exchange system (RC-2) shown as a function of time of sampling.

Figure 3. Plot of plutonium concentrations in cooling water sampled after the ion-exchange system (3-CVCSOUT) shown as a function of time of sampling.



3.3 Results of Pu in Filtered Samples of Cooling Water

In Table 3 we summarize the results of the data used to derive the filter efficiency used for the mechanical filter. Due to the small difference between the pore size (5 μm) of the filter used compared to the pore size (6 μm) of the mechanical filter and since all the cooling-water samples considered for this filter size were collected after the ion-exchange system (2-CVCSOUT) which is where the mechanical filter is located, the resulting average efficiency of 63% is considered representative for the mechanical filter.

Table 3. Results of analyses of samples (from 2-CVCSOUT) filtrated through a microfilter (pore size 5 micrometre). The results show the activities of the plutonium isotopes found on the filter and in the filtrate and give the calculated filter efficiencies. Pu-239 is used in the table for the sum of Pu-239 and Pu-240.

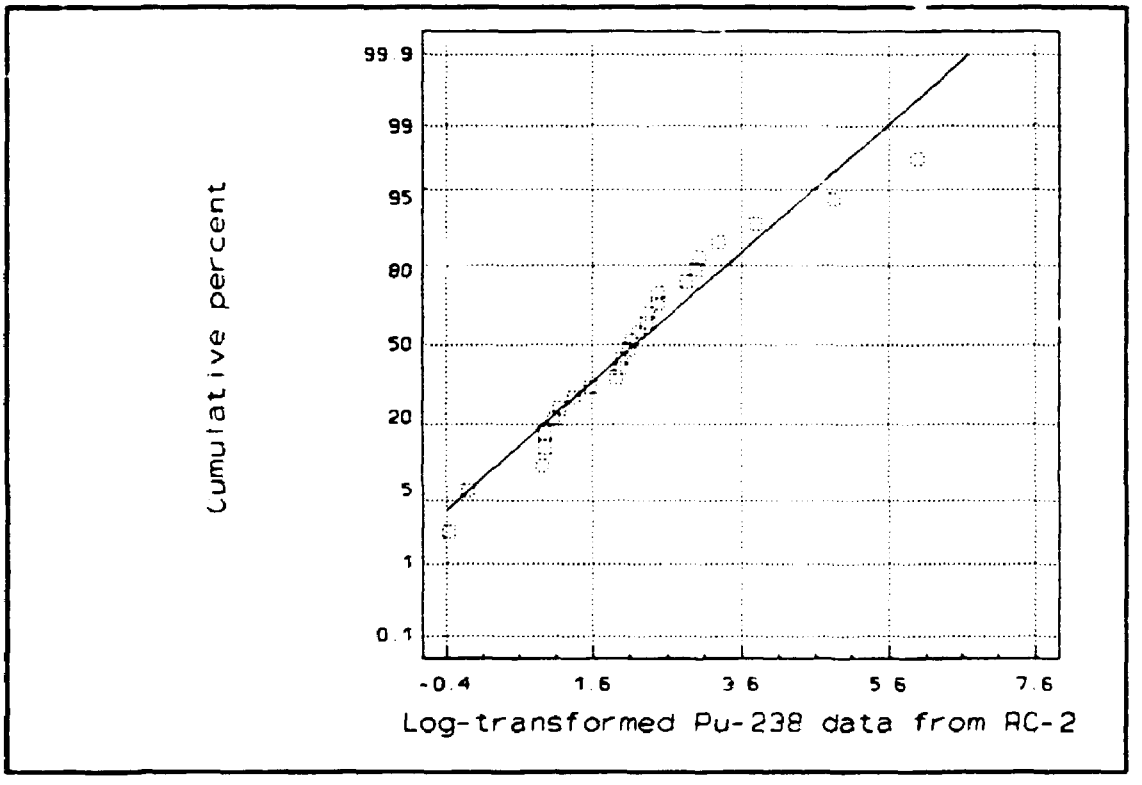
		Filter				Solution				Filter efficiency	
Date	Volume (ml)	Pu-238 (mBq)	SD (%)	Pu-239 (mBq)	SD (%)	Pu-238 (mBq)	SD (%)	Pu-239 (mBq)	SD (%)	Pu-238 (%)	Pu-239 (%)
19 Nov	1025	3.93	10	1.85	12	2.[illegible]	12	0.60	22	60.5	75.3
20 Nov	1080	1.63	14	0.79	20	1.[illegible]	16	0.28	39	55.3	73.7
21 Nov	1125	0.70	25	0.34	34	0.46	25	<0.18		60.2	
22 Nov	1100	0.45	29	0.33	37	0.72	25	<0.24		38.7	
25 Nov	1100	0.51	24	0.15	41	0.39	31	<0.14		56.8	
26 Nov	1125	1.85	13	0.64	20	1.19	17	0.45	26	60.7	58.8
27 Nov	1120	1.86	14	1.05	17	0.94	18	0.54	23	66.4	66.2
28 Nov	1130	1.29	17	0.41	48	0.97	21	0.68	23	57.0	37.5
29 Nov	1100	1.03	14	0.[illegible]	23	0.67	17	0.22	35	60.6	69.[illegible]
02 Dec	1115	19.01	7	5.6[illegible]	9	3.[illegible]	13	1.33	19	86.3	81.0
									Mean	60.3	66.0
									SD (%)	19	22

4 Assessment of Pu in the Reactor Cooling System

4.1 Estimate of Pu in the Ion-Exchange Resin

The results shown in Tables 1 and 2 and in Figures 2 and 3 indicate that the data do not follow simple normal distributions. The data are in fact better described by log-normal distributions as is shown in Figures 4, 5, 6 and 7 where the log-transformed data for the two groups of plutonium isotopes are shown for each of the two sampling points in cumulative normal-probability plots.

Figure 4. Normal probability plot of the log-transformed ^{238}Pu data ($mBq\ l^{-1}$) from the cooling water samples collected before the ion-exchange system (RC-2).



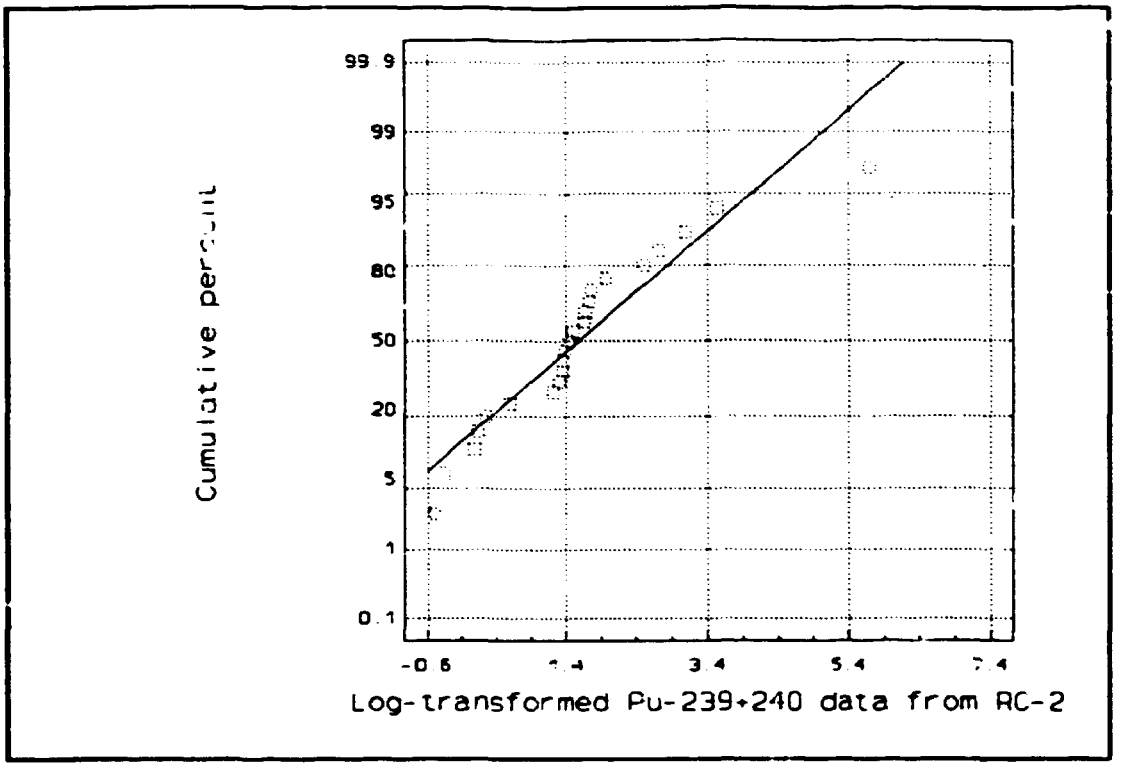
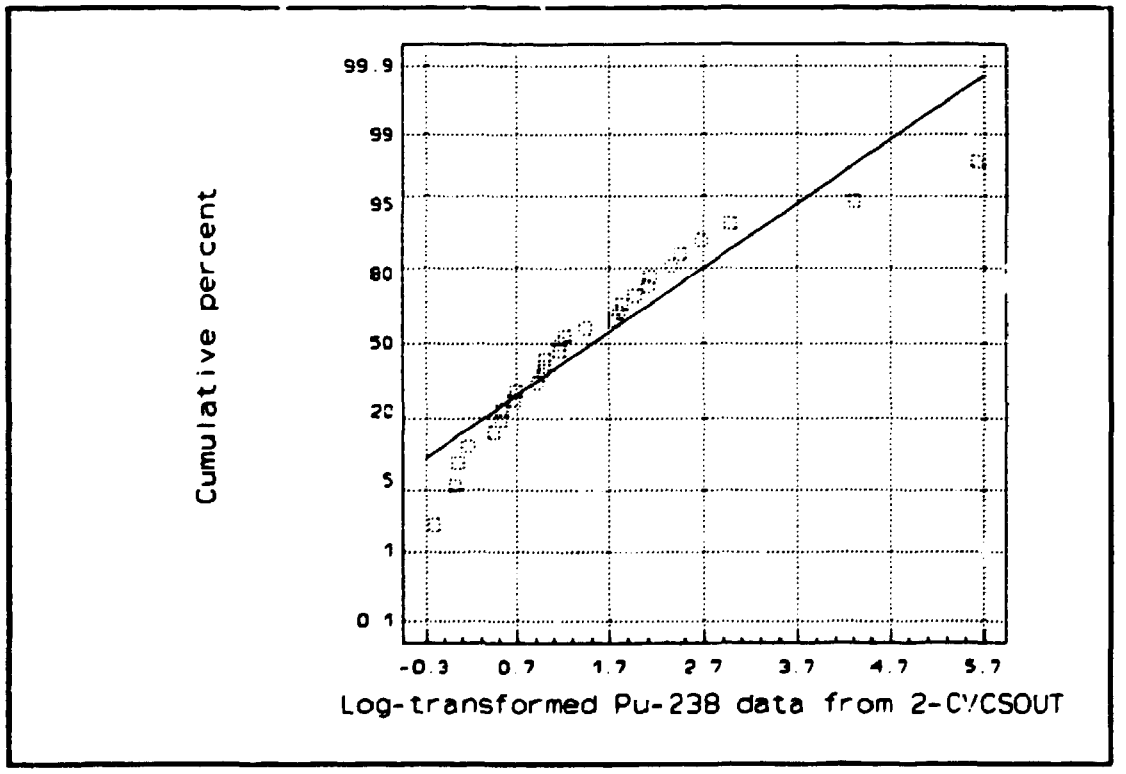


Figure 5. Normal probability plot of the log-transformed $^{239+240}Pu$ data (mBq l^{-1}) from the cooling water samples collected before the ion-exchange system (RC-2).

Figure 6. Normal probability plot of the log-transformed ^{238}Pu data (mBq l^{-1}) from the cooling water samples collected after the ion-exchange system (2-CVCSOUT).



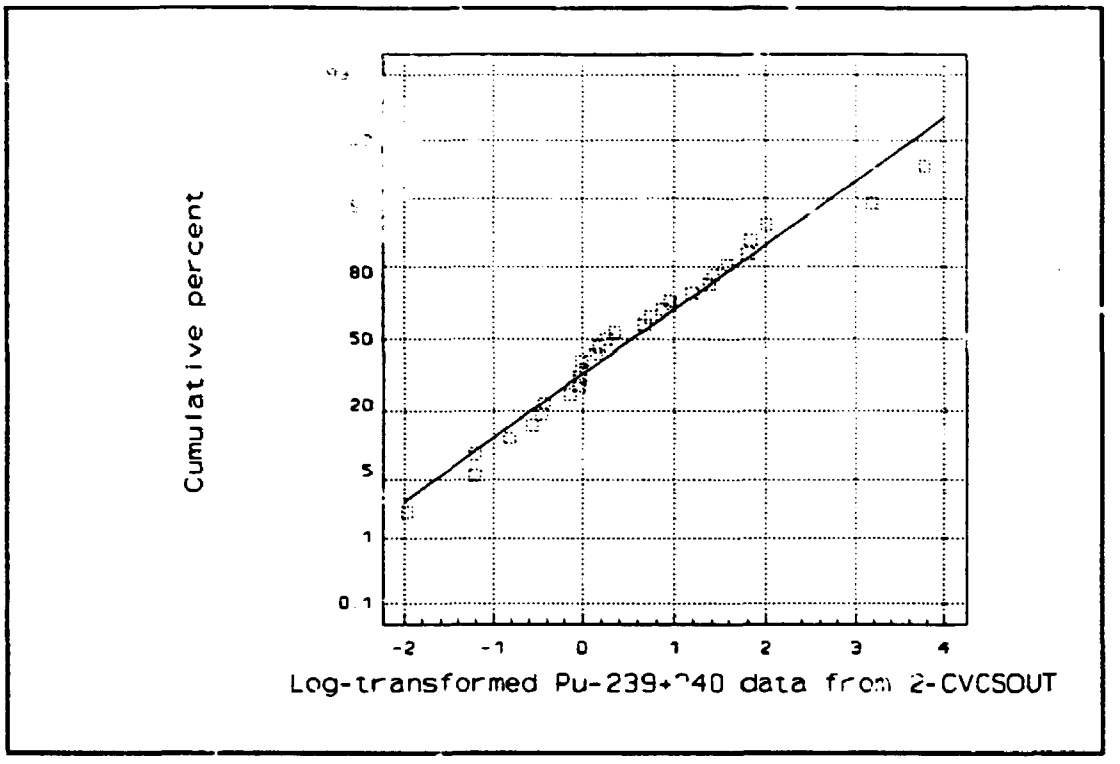


Figure 7. Normal probability plot of the log-transformed $^{239+240}Pu$ data (mBq l^{-1}) from the cooling water samples collected after the ion-exchange system (2-CVCSOUT).

Since the procedure to be used for estimating the efficiency of the ion-exchange system for removal of plutonium in the primary cooling water is based on assumptions of lower concentrations of plutonium in the cooling water after the ion-exchange system than before, the data have been analyzed to see if they can support this assumption. The data do show higher levels before than after the ion-exchange system, and one-tailed Students t-tests have been carried out on the log-transformed data to see if the differences are significantly larger than zero. For the ^{238}Pu data the t-test shows that the difference found is significant at the 5% level (t = 1.83, 53 degrees of freedom), and for the $^{239+240}Pu$ data the difference is significant at the 1% level (t = 2.80, 50 df). If we compare the plutonium data for all three isotopes together by adding the concentrations for ^{238}Pu and $^{239+240}Pu$ we find that the difference is significant at the 1% level (t = 2.69, 50 df).

The efficiency of the ion-exchange system for cleaning the cooling water may thus be calculated by comparing the arithmetic mean concentrations of plutonium before and after the ion- exchange system. The results are summarised in Table 4. In addition to the arithmetic mean values calculated from the measured data (Tables 1 and 2) Table 4 includes the arithmetic mean values obtained from the log-normal distributions fitted to the observed data and shown in Figures 4-7 as straight lines. While the arithmetic mean values from the observations depend strongly on the few large observations in each group, this is not the case for the fitted distributions which provide better representations of all the data in the group.

The data from Table 4 show ratios between the levels after to those before the ion-exchange system varying from 0.25 for $^{239+240}Pu$ to 0.50 for ^{238}Pu, and for the sum of the three plutonium isotopes the ratio is 0.42. For the data from the fitted distributions the ratios are nearly the same, although the difference between the ratios for ^{238}Pu and $^{239+240}Pu$ is less pronounced. We may thus conclude that the mean cleaning efficiency of the ion-exchange system for plutonium is about 60% corresponding to a ratio of the levels after-to-before the system of 0.4.

From this we can estimate the inventories of plutonium in the ion-exchange system after one years operation. We assume identical amounts of activity of ^{239}Pu and ^{240}Pu, a mean flow of cooling water from the primary system through the ion-exchange system and the mechanical filter of

Table 4. Arithmetic mean values of plutonium isotopes in primary cooling water collected before (RC-2) and after (2-CVCS) the ion-exchange system, the differences and the ratios between the two. The values are used to estimate the cleaning efficiency of the ion-exchange system to plutonium in the primary cooling water

	Data from measurement			Data from fitted distributions		
	^{238}Pu mBq l^{-1}	$^{239-240}Pu$ mBq l^{-1}	$^{238-239-240}Pu$ mBq l^{-1}	^{238}Pu mBq l^{-1}	$^{239-240}Pu$ mBq l^{-1}	$^{238-239-240}Pu$ mBq l^{-1}
Before (RC-2)	33.52	17.31	50.83	22.57	12.15	36.31
After (2-CVCS)	16.86	4.33	21.19	10.34	3.86	14.26
Difference	16.66	13.00	29.64	12.23	8.29	22.05
After/Before	0.50	0.25	0.42	0.46	0.32	0.39

Table 5. Inventories and concentrations of ^{238}Pu, ^{239}Pu and ^{240}Pu in the ion-exchange system after one years operation estimated from measured data directly and from data fitted to the observations

	Data from measurement			Data from fitted distributions		
	^{238}Pu	^{239}Pu	^{240}Pu	^{238}Pu	^{239}Pu	^{240}Pu
Inventory (MBq y^{-1})	4.2	1.1	1.1	2.8	0.8	0.8
Concentration (kBq kg^{-1} y^{-1})	4.4	1.1	1.1	2.9	0.8	0.8

6.6 l s^{-1} and a total weight of the ion-exchange resin in the system of 961 kg. The annual inventories and concentrations of the three plutonium isotopes in the ion-exchange system are given in Table 5, where the results are calculated from the plutonium levels before the ion- exchange system both from the observed data and the fitted data and a cleaning efficiency of the system of 60%. The overall results are that the annual inventories in the ion-exchange system may be estimated at approximately 1 MBq for both ^{239}Pu and ^{240}Pu and at 3 MBq for ^{238}Pu.

4.2 Estimate of Pu in the Mechanical Filter

The annual inventory of plutonium isotopes in the mechanical filter may be estimated in a similar fashion as in the preceding section. The mean cleaning efficiency of the filter is estimated at 63% from Table 3. When we apply the same assumptions as in the previous section we get the inventories shown in Table 6.

The concentrations of plutonium in the primary cooling water after the mechanical filter may be estimated from the measured concentrations at 2-CVCSOUT after a 63% reduction at 6.2 mBq ^{238}Pu l^{-1} and 1.6 mBq $^{239+240}Pu$ l^{-1}.

Table 6. Inventories and concentrations of ^{238}Pu, ^{239}Pu and ^{240}Pu in the mechanical filter after one years operation estimated from measured data directly and from data fitted to the observations

	Data from measurement			Data from fitted distributions		
	^{238}Pu	^{239}Pu	^{240}Pu	^{238}Pu	^{239}Pu	^{240}Pu
Inventory (MBq y^{-1})	2.2	0.3	0.3	1.4	0.3	0.3

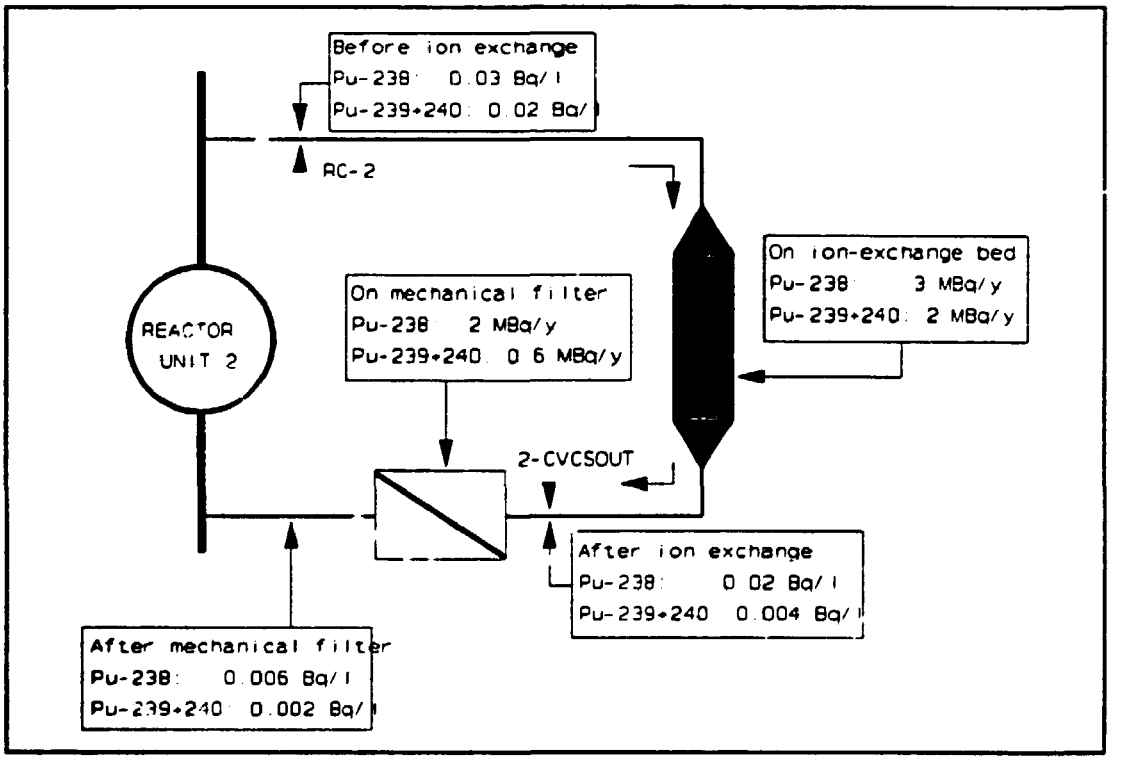
5 Conclusion

The results of the project may be summarised as shown in Figure 8 which outlines the flow through the ion-exchange system and the mechanical filter from the primary cooling water for unit 2 of the Ringhals power station. The concentrations of plutonium isotopes in the cooling water are indicated at various points as well as the annual inventories in the ion-exchange bed and in the mechanical filter.

Recommendations for future research comprise the following items:

- determination of plutonium in other reactor types,
- determinaion of plutonium in samples of ion-exchange resin,
- development of radiochemical procedures for the analysis of americium and curium in samples of reactor cooling water,
- determination of americium and curium in reactor cooling water,
- determination of transuranic elements in cooling water from spent-fuel storage ponds.

Figure 8. Schematic diagram showing the flow of the cooling water from the primary circuit of unit 2 of the Ringhals nuclear power station through the ion-exchange system and the mechanical filter.



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References

Chen, Q., A. Aarkrog, S.P. Nielsen, H. Dahlgaard, H. Nies, Y. Yixuan and K. Mandrup (1991). Determination of Plutonium in Environmental Samples by Controlled Valence in Anion Exchange. Risø-M-2856.

Chen, Q., S.P. Nielsen and A. Aarkrog (1989). Preparation of thin alpha sources by electrospraying for efficiency calibration purposes. *J. Radioanal. Nucl. Chem., Letters* 135 /2/, p. 117-123.

Currie, L.A. (1968). Limits for Qualitative Detection and Quantitative Determination. *Anal. Chem.* 40, p. 586.

Hedemann Jensen, P., E. Lundtang Petersen, S. Thykier-Nielsen and F. Heikel Vinther (1977). Calculation of the Individual and Population Doses on Danish Territory Resulting from Hypothetical Core-Melt Accidents at the Barsebäck Reactor. Risø-R-356.

Appendix

A.1 Radiochemical Procedure for Cooling-Water Samples

The following description outlines the radiochemical procedure for the analysis of alpha-emitting plutonium isotopes in cooling- water samples by controlled valence.

a. Add to 500 ml reactor cooling water: ^{242}Pu spike, 20 ml conc. HNO_3 and 200 mg $NaNO_3$. Evaporate slowly to dryness.
b. Add 20 ml conc. HNO_3 and 3 ml conc. HCl. Cover with glass disc, heat for 1 h to dissolve the particles. Evaporate to dryness.
c. Dissolve the sample in 25 ml 0.5 N HNO_3. Add 800 mg $NaSO_3$. Let stay for 20 min to reduce the Pu to Pu^{+3}. Add 800 mg $NaNO_2$. Let stay for 10 min to get Pu^{+4}. Acidify the solution to 8 N HNO_3.
d. Pass the solution through a column (11x1.5 cm, AG 1-X_4, 100-200 mesh) which is equilibrated with 20 ml 8 N HNO_3 + 0.4 g $K_2S_2O_8$ and 40 ml 8 N HNO_3 + 0.4 g $NaNO_2$. The flow rate through the column is about 1 ml/min.
e. Wash the column with 150 ml 8 N HNO_3 + 0.8 g $NaNO_2$ (fresh) to decontaminate for uranium. Wash the column with 15 ml conc. HCl + a few milligrams $NaNO_2$. Wash the column with 80 ml conc. HCl to decontaminate for thorium.
f. Strip with 15 ml H_2O, 15 ml 0.4 N NaOH, 25 ml 2 N HNO_3 and 75 ml 0.5 N HNO_3 + 0.04 M $NaNO_3$. Add 0.35 ml 18 M H_2SO_4, 5 ml conc. HNO_3 and 5 ml conc. HCl. Evaporate to dryness.
g. Dissolve the sample in 3 ml 0.5 N H_2SO_4. Wash the beaker with 2 ml 0.5 N H_2SO_4 and two times with 5 ml electrolyte. Transfer the two 5-ml parts one by one to the electrodeposition cell. The current is 0.6 A/cm^2 for 2-3 h. Cool with water. Electrolyte: 30 g $(NH_4)_2C_2O_4 . H_2O$ + 51.4 g NH_4Cl + 0.9 g $NH_2OH.HCl$ + 3.3 ml diethylenetriaminepentaacetic acid/NH_3 (25% NH_3) in 1 litre.

A.2 Radiochemical Procedure for Samples of Ion-Exchange Resin

The following description outlines the radiochemical procedure for the analysis of alpha-emitting plutonium isotopes in samples of ion-exchange resin by controlled valence.

a. Add 1 g (1-6 g) ion-exchange resin, 1 ml 18 M H_2SO_4 (1-2 ml), 2 ml conc. HNO_3 (2-10 ml) and ^{242}Pu spike to a 150 ml beaker. Leach the sample over night. Evaporate to dryness.
b. Heat the sample at 650°C over night. Add 9 ml conc. HNO_3 + 3 ml conc. HCl to the sample. Cover with a glass disc, heat and boil for 1 h. Evaporate to dryness.
c. Dissolve the sample in 20 ml 0.5 N HNO_3. Add 800 mg $NaSO_3$. Let stay for 20 min to reduce the Pu to Pu^{+3}. Add 800 mg $NaNO_2$. Let stay for 10 min to get Pu^{+4}. Acidify the solution to 8 N HNO_3.
d. Pass the solution through a column (11x1.5 cm, AG 1-X_4, 100-200 mesh) which is equilibrated with 20 ml 8 N HNO_3 + 0.4 g $K_2S_2O_8$ and 40 ml 8 N HNO_3 + 0.4 g $NaNO_2$. The flow rate through the column is about 1 ml/min.
e. Wash the column with 150 ml 8 N HNO_3 + 0.8 g $NaNO_2$ (fresh) to decontaminate for uranium. Wash the column with 15 ml conc. HCl + a few milligrams $NaNO_2$. Wash the column with 80 ml conc. HCl to decontaminate for thorium.
f. Strip with 15 ml H_2O, 15 ml 0.4 N NaOH, 25 ml 2 N HNO_3 and 75 ml 0.5 N HNO_3 + 0.04 M $NaNO_3$. Add 0.35 ml 18 M H_2SO_4, 5 ml conc. HNO_3 and 5 ml conc. HCl. Evaporate to dryness.
g. Dissolve the sample in 3 ml 0.5 N H_2SO_4. Wash the beaker with 2 ml 0.5 N H_2SO_4 and two times with 5 ml electrolyte. Transfer the two 5-ml parts one by one to the electrodeposition cell. The current is 0.6 A/cm^2 for 2-3 h. Cool with water. Electrolyte: 30 g $(NH_4)_2C_2O_4 . H_2O$ + 51.4 g NH_4Cl + 0.9 g $NH_2OH.HCl$ + 3.3 ml diethylenetriaminepentaacetic acid/NH_3 (25% NH_3) in 1 litre.

A.3 Detailed Analytical Results

The following two tables give the detailed results of the plutonium analyses including the radiochemical yield for each sample. Table A1 gives the results for the samples collected before the ion-exchange system at RC-2, and Table A2 gives the results for the samples collected after the ion-exchange system at 2-CVCSOUT.

The tables give for each sample the collection date, the sample volume, the filter size, the chemical yield determined from the spike and the results for ^{238}Pu and $^{239+240}Pu$ for the filter fraction and the filtrate fraction and the total. In the tables ^{239}Pu refers to the sum of ^{239}Pu and ^{240}Pu.

Table A1. Detailed results of samples collected at RC-2 before the ion-exchange system

			Results from filter					Results from solution					Total result				
Date	Vol.	Filt	Y.	^{238}Pu	SD	^{239}Pu	SD	Y.	^{238}Pu	SD	^{239}Pu	SD	Y.	^{238}Pu	SD	^{239}Pu	SD
	ml	µm	%	mBq/l	%	mBq/l	%	%	mBq/l	%	mBq/l	%	%	mBq/l	%	mBq/l	%
17 Jun	100												90	128	6	34.0	7
18 Jun	100												88	12.0	8	4.0	11
19 Jun	100												95	21.0	7	5.6	10
24 Jun	100												84	45.0	7	22.0	8
26 Jun	150	GF/F	65	20.0	7	6.7	10	68	< 0.6		0.4	40		20.0	7	7.1	7
26 Jun	100												78	401	6	300	6
28 Jun	100												101	2.6	19	0.7	32
02 Jul	100												64	< 1.3		< 0.9	
02 Jul	100												86	< 1.0		0.7	41
03 Jul	50												88	7.4	23	15.0	14
03 Jul	47												93	< 3.4		3.2	25
03 Jul	100												94	2.6	17	0.6	31
25 Sep	375	0.45	32	23.7	10	10.3	13	37	3.3	22	1.9	26		27.0	9	12.2	12
26 Sep	390	0.15	27	6.0	15	1.6	26	22	11.6	17	3.7	28		17.6	12	5.3	21
30 Sep	500	0.15	18	1.3	38	0.9	40	31	5.9	20	2.8	28		7.2	18	3.7	23
02 Oct	550	0.15	62	9.0	8	3.6	11	58	< 1.7		< 1.4			9.0	8	3.6	11
07 Oct	500	0.15	73	3.1	15	0.9	27	57	1.7	28	0.9	34		4.8	14	1.8	22
08 Oct	550	0.15	82	6.6	9	3.0	11	70	1.6	27	0.9	32		8.1	9	3.9	11
22 Oct	520	0.05	65	2.1	12	1.1	16	88	0.4	27	< 0.2			2.5	11	1.1	16
23 Oct	520	0.05	73	2.6	11	1.1	15	57	0.4	37	< 0.6			2.9	11	1.1	15
24 Oct	530	0.05	65	8.4	9	3.8	13	33	< 1.0		< 0.7			8.4	9	3.8	13
25 Oct	545	0.05	52	7.6	10	3.1	15	76	0.9	25	< 0.3			8.4	9	3.1	15
28 Oct	555	0.05	71	3.4	13	1.1	23	60	0.5	32	< 0.5			3.9	12	1.1	23
29 Oc	520	0.05	43	9.8	11	4.3	15	58	< 0.6		< 0.4			9.8	11	4.3	15
30 Oct	530	0.05	43	11.0	9	5.0	12	50	0.9	28	< 0.4			11.9	9	5.0	12
01 Nov	530	0.05	73	3.4	12	2.1	15	72	3.4	10	3.7	10		6.8	8	5.8	8

Table A2. Detailed results of samples collected at 2-CVCSOUT after the ion-exchange system

			Results from filter					Results from solution					Total result				
Date	Vol.	Filt.	Y.	^{238}Pu	SD	^{239}Pu	SD	Y.	^{238}Pu	SD	^{239}Pu	SD	Y.	^{238}Pu	SD	^{239}Pu	SD
	ml	µm	%	mBq/l	%	mBq/l	%	%	mBq/l	%	mBq/l	%	%	mBq/l	%	mBq/l	%
24 Jun	100												86	7.2	10	3.3	13
26 Jun	100												83	1.7	25	1.2	25
28 Jun	100												80	4.2	13	1.3	24
02 Jul	100												98	8.2	11	4.9	13
03 Jul	100												85	75.0	6	24.0	8
09 Oct	550	0.15	96	2.1	12	0.9	17	90	1.1	29	1.1	25		3.2	13	2.0	16
10 Oct	550	0.15	69	13.2	7	5.8	9	88	1.4	37	< 1.0			14.6	7	5.8	9
11 Oct	520	0.05	84	10.5	7	7.2	7	98	1.2	23	0.2	53		11.7	7	7.5	7
14 Oct	550	0.05	52	277.3	6	43.4	6	66	0.6	25	< 0.15			278	6	43.4	6
15 Oct	520	0.05	40	1.7	24	< 0.78		63	< 0.79		< 0.56			1.7	24		
16 Oct	550	0.05	76	2.1	11	1.0	15	71	0.5	25	0.2	41		2.7	10	1.2	14
31 Oct	530	0.05	50	4.0	17	1.7	24	39	1.9	21	0.9	34		5.8	13	2.6	20
04 Nov	580	0.05	89	2.3	12	0.9	18	59	1.0	17	< 0.48			3.3	10	0.9	18
05 Nov	520	0.05	69	0.9	26	0.5	33	69	0.7	23	0.1	57		1.7	18	0.6	29
06 Nov	580	0.05	67	0.4	34	0.2	38	61	0.8	25	0.2	44		1.2	20	0.4	29
07 Nov	580	0.05	70	9.9	8	3.8	10	72	0.7	23	0.2	46		10.6	8	4.0	10
08 Nov	540	0.05	60	7.5	10	3.8	13	64	0.9	27	0.4	41		8.4	9	4.2	12
11 Nov	590	0.05	69	2.8	11	1.0	21	71	0.6	34	< 0.35			3.4	11	1.0	21
12 Nov	545	0.05	58	4.9	10	2.1	14	76	1.2	18	< 0.24			6.1	9	2.1	14
19 Nov	1025	5.00	64	3.8	10	1.8	12	57	2.5	12	0.6	22		6.3	8	2.4	11
20 Nov	1080	5.00	56	1.5	14	0.7	20	49	1.2	16	0.3	38		2.7	11	1.0	18
21 Nov	1125	5.00	42	0.6	25	0.3	34	53	0.4	25	< 0.16			1.0	18	0.3	34
22 Nov	1100	5.00	55	0.4	29	0.3	37	45	0.7	25	< 0.22			1.1	19	0.3	37
25 Nov	1100	5.00	50	0.5	24	0.1	41	46	0.4	31	< 0.13			0.8	19	0.1	41
26 Nov	1125	5.00	48	1.6	13	0.6	20	42	1.1	17	0.4	26		2.7	10	1.0	16
27 Nov	1120	5.00	39	1.7	14	0.9	17	50	0.8	18	0.5	23		2.5	11	1.4	14
28 Nov	1130	5.00	29	1.1	17	0.4	48	44	0.9	21	0.6	23		2.0	13	1.0	23
29 Nov	1100	5.00	48	0.9	14	0.5	23	59	0.6	17	0.2	35		1.6	11	0.7	19
02 Dec	1115	5.00	41	17.1	7	5.1	9	28	2.7	13	1.2	19		19.8	6	6.3	8

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An Assessment of ^{238}Pu and $^{239+240}Pu$ in the Primary Cooling Water of a PWR

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Abstract (Max. 2000 characters)

Radiochemical procedures were developed for the analysis of plutonium in primary cooling water and in ion-exchange resin. Analyses of cooling-water samples from unit 2 of the Ringhals nuclear power station demonstrated that a major fraction of the plutonium found in the cooling water is associated with particles. The amounts of plutonium isotopes retained annually in the ion-exchange cleaning system are estimated at 3 MBq of ^{238}Pu and 2 MBq of $^{239+240}Pu$ and in a mechanical filter the amounts are estimated at 2 MBq of ^{238}Pu and 0.6 MBq of $^{239+240}Pu$.

Descriptors INIS/EDB

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