Determination of Transition Metals in the Primary Water of Pressurized Water Reactors by Inductively Coupled Plasma Mass Spectrometry

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A method for the determination of the transition metals Fe, Mn, Ni and Co in the primary water of pressurized water reactors (PWRs) is described and applied to the analysis of water at the Loviisa power plants. Samples are collected *via* the standard sampling line or an isokinetic line. There is no difference in the results obtained. The water is filtered through a 0.45 μm membrane filter under oxygen-free conditions to separate the soluble and insoluble fractions. The transition metals are concentrated and separated from boric acid using cation chromatography. A quantitative yield is obtained and concentration factors of up to 500 are used. The samples are analysed using inductively coupled plasma mass spectrometry. The detection limits obtained are Fe 2, Mn 0.2, Ni 0.2 and Co 0.02 ng l^{-1} when a 1 l sample is used.

Keywords: *Inductively coupled plasma mass spectrometry; transition metals; primary water; pressurized water reactor*

The radiation field build-up on the primary circuit components in nuclear power plants is of concern because of the exposure of the workers to radiation. Most of the radiation is caused by activation products from transition metals, mostly ^{60}Co from Co, ^{58}Co from Ni and ^{54}Mn from Fe. In order to study the physico-chemical mechanisms of the production and transport of the radionuclides, it is necessary to know the concentrations of the source metals in the primary water. The concentration of Fe must also be known in order to understand the corrosion and corrosion product transport phenomena. Usually the soluble and insoluble metals are determined separately in order to obtain information on the solubilities. Two different stages of reactor operations are normally studied. During full power, the transition metal concentrations are very low and the variation moderate. Therefore the sampling frequency can be one sample per month, or less. During shutdown the solubilities increase and fast transients in the transition metal concentrations can be observed. Therefore shutdowns, and sometimes the return to full power, are followed with high sampling frequency, up to one sample per hour over 3–4 days.

The analysis is not without problems. The sampling is complicated by the need to reduce the pressure and temperature, the latter because the pH changes, which affects the solubility.[1] The samples are transported through tens of metres of steel pipes. The problems concerning representative sampling have been extensively investigated and discussed in a number of papers.[1–6] It has been shown that absorption and desorption phenomena affect the transport of radionuclides and transition metals in the pipelines in a way which is not yet fully understood. However, it has also been shown that under appropriate conditions, analytical results behave logically in a way that enables a comparison between different reactors and different years of the same reactor to be made. Bridle and co-workers[1,7] introduced a so-called isokinetic sampling system in which the pressure drop is effected with a 50 m long capillary tube instead of the normal valves and heat exchangers. They indicate that the isokinetic sampling line gives a more representative sample than the standard sampling line because the water flow is smooth without variations in flow rate. However, experimental data verifying this statement have not been presented. Svoboda *et al.*[2] compared an isokinetic sampling line with a standard one. They did not observe any differences in the concentrations of Fe, Ni and Co.

The samples should be filtered before the water has been in contact with air. This prevents the oxidation of Fe, which would drastically decrease its solubility. Usually 0.45 μm membrane filters are used, although part of the insoluble material seems to be in colloidal form with particle sizes much below that. The lowest Co concentrations found in the insoluble fraction during full power are below 1 ng l^{-1}.[8] Therefore several tens or even hundreds of litres of water might be filtered for a sample.

The lowest soluble Ni concentrations during full power are a few tens of ng l^{-1}, the lowest Co concentrations a few ng l^{-1}.[8] Therefore most analytical techniques in use require a preconcentration step. Two methods have been used. Ion-exchange filters, such as Acropor SA 6404, Whatman SA2 and Sumimoto Chemical Expapier F-2, have been used.[2,9,10] These have the benefits that fairly high sampling rates can be maintained, 50–150 ml min^{-1}, and that they can be used in combination with a particle filter and an anion filter. The reported capacities can be 0.5 mmol per filter, although experiment has shown the practical capacity to be one tenth of this.[2] In combination with ion chromatographs, preconcentration columns can be conveniently used.[1,7] They have capacities of up to 0.45 mequiv. One drawback is the low sampling speed, 1–2 ml min^{-1}. Two different types have been used in this work. They are the Dionex MetPac CC-1 and Waters Guard Pak.

A variety of analytical techniques have been used for the determination of the transition metals. The only technique capable of determining Co during full-power conditions in most reactors without preconcentration is voltammetry.[11–13] The reported detection limits are 1–2 ng l^{-1} for Co and 8 ng l^{-1} for Ni. All other methods require a degree of preconcentration if low concentrations are to be detected. Ion chromatography has been widely applied in European power plants, mainly by the Winfrith Group in the UK.[1,7,14–17] Using preconcentration of a 1 l sample, a detection limit of 0.5 ng l^{-1} of Co has been reported.[1] Atomic absorption spectrometry can, in principle, be used after preconcentration from a very large sample.[2] The detection limit could be significantly improved by using electrothermal vaporization, but no actual results from determinations of low levels of Co have been reported. A few determinations using X-ray fluorescence[2,9] and flow injection techniques[18,19] have also been reported, the latter without any details on the method. Neutron activation analyses have also been used, but

the detection limits are low, requiring a considerable preconcentration.[2] Inductively coupled plasma mass spectrometry (ICP-MS) is a very sensitive method for analysis of water but to date very few applications on the analysis of nuclear power plant (NPP) waters have been reported. There are a few references to work on loop tests and boiling water reactor (BWR) analysis, but these contain very few details.[20–22]

The present paper describes the development of methods for sampling, preconcentration and ICP-MS determination of soluble and insoluble Fe, Mn, Ni and Co in the primary water of a pressurized water reactor (PWR), results for the determination of these metals in the primary water of the Loviisa 1 power plant during the refuelling shut-down in 1992, and Loviisa 1 and Loviisa 2 during full power in 1993 are presented. Only a few results are presented to show the applicability of the method. The full results together with a discussion will be published later.

Experimental

Sampling

In old reactors the sampling lines are fixed and cannot be changed. In the Loviisa 1 NPP an isokinetic sampling by-line had also been installed and this was compared with the standard line.

The basic principle of sampling is to have a steady continuous flow through the sampling line in order to avoid transients which change the physico-chemical parameters. Another principle is to prevent the oxidation of Fe before the filtering and the preconcentration steps. In the present work a 0.45 μm membrane filter was used, mainly in order to facilitate a comparison with literature data. It was realized, however, that a smaller pore size filter should be used in order to separate all the smaller particles. This will be investigated in future work.

During the shut-down of the Loviisa 1 plant (18–21 July, 1992) 0.5–1 l samples were filtered in closed Nalgene filter holders equipped with 1 l receiver containers. Nitrogen gas was passed through the system to prevent contact with air. Part of each of the filtered samples was poured into 100 ml acid washed polyethylene bottles, which were immediately frozen. The frozen samples were stored for up to 3 months before analysis. Most of the samples were taken from the purification circuit before the ion-exchange columns. This sampling line had an isokinetic by-line installed in 1989.[7] Samples were taken both from the standard outlet and the isokinetic line. The isokinetic line was opened 9 d before the actual sampling started. The flow rate was 35 ml min^{-1}. The standard sampling line was opened 5 d before the sampling started. The flow rate was about 1 l min^{-1}.

During the shut-down of the Loviisa 2 plant (11–15 September, 1992), sampling was made from the corresponding purification circuit, but only using the standard sampling line, which was opened 3 d before the sampling started. A Millipore filter holder was connected directly to the sampling line to prevent the access of air. A 10 l bottle was used for measurement of the volume of the filtered water. From 10 to 20 l samples were filtered. The water was preconcentrated immediately after filtering, but the preconcentration was made from water that had been poured from one container into another. Therefore it had been in contact with air.

The above experiences resulted in the final sampling arrangement shown in Fig. 1. Both the filtering and the preconcentration are performed in a closed system which prevents the access of air. This system was used for the analysis of the water of the Loviisa 2 reactor during full power in February 1993. The continuous sampling flow was 1.6 l min^{-1}. About 30 l of water were filtered.

Preconcentration

The reason for the preconcentration is two-fold. Firstly, the samples have to be concentrated in order to improve the
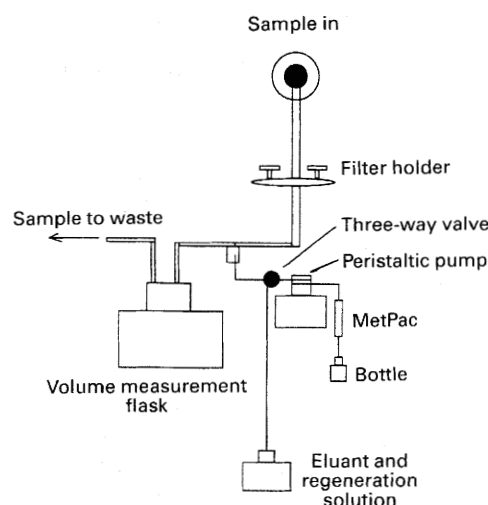

Fig. 1 Principles of the arrangement for sampling primary water of a PWR

detection limits and thus the accuracy of the analysis. Secondly, the high boric acid concentrations, up to 12 g l^{-1}, prevent the direct injection of the water into the ICP-MS system, which is equipped with a normal nebulizer. Therefore the boric acid has to be removed. Two different preconcentration columns were tested. These were the Waters Guard Pak, which is packed with a porous polystyrene gel with sulfonate functional groups, and the Dionex MetPac CC-1, which contains a macroporous iminodiacetate chelating resin. The preconcentration procedures, shown in Table 1, are those used, although not necessarily the optimal ones. The reason for choosing these schedules was partly to be able to run the two preconcentration columns in parallel.

The Waters column was tested in the laboratory using simulated reactor water containing 12 g l^{-1} of boric acid, 6 mg l^{-1} of NH$_3$, 2 mg l^{-1} of KOH, 50 ng ml^{-1} of Fe, 13 ng ml^{-1} of Mn, 106 ng ml^{-1} of Ni and 0.3 ng ml^{-1} of Co. Different amounts of the water were run through the column, which was eluted according to Table 1. The metal concentrations in the 10 ml elution fraction were determined by ICP-MS to establish the yield. The capacity was determined by running 500 ml of the water through the column in order to exceed the capacity significantly. The column was then eluted and the concentrations determined by ICP-MS.

Because only preconcentration with the Waters Guard Pak was tested in the laboratory the behaviour of the Dionex MetPac CC-1 was tested by comparing it with the Waters Guard Pak. This was done by preconcentrating every second sample of the Loviisa 1 samples with each column. The Loviisa 1 samples were left to thaw in the refrigerator, after which they were preconcentrated according to the above scheme. In the Loviisa 2 sampling, a number of parallel samples were preconcentrated with each column immediately after sampling.

The blank of the MetPac CC-1 column and the reagents was measured by regenerating the column with 20 ml of the

Table 1 Preconcentration scheme for the Waters Guard Pak and the Dionex MetPac CC-1. During the shut-downs 50–100 ml samples were preconcentrated, while during full power 1–2 l samples were used

Step	Dionex MetPac CC-1	Waters Guard Pak
Adsorption	100–2000 ml water	100 ml water
Elution	2 ml 1 mol l^{-1} HNO$_3$	10 ml 0.1 mol l^{-1} HNO$_3$
Regeneration	8 ml 1 mol l^{-1} HNO$_3$ 10 ml buffer 2 mol l^{-1} NH$_4$Ac, pH 5.5	10 ml 0.1 mol l^{-1} HNO$_3$

buffer solution, cleaning the column with 10 ml of de-ionized water and eluting with 2 ml of 1 mol l^{-1} HNO$_3$.

Determination of the Transition Metals by ICP-MS

The samples were analysed with a VG PlasmaQuad PQ2 + ICP-MS. The operating parameters are shown in Table 2. The samples are radioactive, although the activity is low being mostly at a level permissible for free handling. In order to avoid unneccessary releases of radioactive material, the gaseous effluents were filtered with an absolute filter and the liquid effluents collected and stored as radioactive waste.

The samples taken during shut-down were diluted with water to 5–10 ml, and run with the sample changer with the parameters shown in Table 2 using a 30 s dual mode counting time. The scanning mode was used. The samples taken during full power were diluted to 5 ml. The samples were run using the sample changer by injecting a 250 µl sample followed by a 1% HNO$_3$ washing solution. After a 45 s delay, when the sample reached the analyser, a 10 s measurement in the peak jumping pulse counting mode was started. The best masses to be used proved to be ^{55}Mn, ^{56}Fe, ^{58}Ni and ^{59}Co.

Calibration curves were produced from solutions of 1.0–50 ng ml^{-1} prepared by dissolving pure metals in HNO$_3$. The measurements were checked by analysing suitably diluted SPEX multi-element plasma standard ICP-MS solutions. Blanks were prepared from the corresponding elution solutions.

When using the normal Ni sampling cone, the blanks caused by contamination from the analyser and the argon ions varied somewhat, but were of the order of Fe 180, Ni 3–10, Mn 0.3 and Co 0.04–0.1 ng ml^{-1}. Through using a sampler made from Al at the work-shop of the Reactor Laboratory, the Ni blank was lowered to 0.5 ng ml^{-1}. Also the Fe blank was decreased to 35 ng ml^{-1}. In future the skimmer will also be made of Al and the solutions and other parts of the analyser checked once more to decrease the blanks further. The Al sampler seems to be much more resistant to the plasma than the Ni sampler, probably because of its excellent heat and electrical conductivity.

The column was shown to give a considerable blank when left standing for a few hours both after the regeneration and elution. Therefore the column has to be cleaned by elution with 1 mol l^{-1} HNO$_3$ before it is taken into use. The purity of the reagents must be controlled.

The similarity of the Ni and Co results and the great difference in concentration caused a suspicion of a Ni interference on the Co peak. Therefore pure Ni solutions were analysed for Co in order to detect any overlapping.

The reproducibility of the ICP-MS measurements were investigated by analysing the same samples several times at different time intervals. The long-term stability of the concentrated samples, which were stored in a refrigerator, and the long-term stability of the ICP-MS instrumentation was investigated by analysing a set of samples twice with an interval of 3 months. The first time was in October 1992 and the second time in January 1993.

The samples on the filters were first dissolved in HNO$_3$, but the dissolution proved to be incomplete. Later a procedure was adopted where the filter was soaked in 1 ml of 1+1 HCl with gentle heating. The sample was then diluted with water for the ICP-MS measurements.

Table 2 Operating parameters for ICP-MS

R.f. power	1.35 kW
Cooling gas	14 l min^{-1}
Aerosol carrier gas	0.8 l min^{-1}
Sample introduction	1 ml min^{-1}
Measurement time	10–30 s per pulse counting mode
Dwell time	0.64 ms
Range	50–65 m/z

Results and Discussion

Sampling and Storage of Samples

A comparison of the soluble metal concentrations obtained from samples taken through the standard sampling device (A) and the isokinetic sampling line (B) is shown in Fig. 2. Although there is some variation in the results, there is no indication that there would be a systematic difference between the two sampling lines. This confirms the results of Svoboda et al.[2] that the isokinetic sampling line is no better than the normal one. The results also indicate that no loss or contamination occurred during the 3 months' storage of the samples.

Preconcentration

The results of the recovery tests are shown in Table 3. The recoveries are satisfactory provided the concentrations are well below the capacity of the column. The results for Fe are poor, probably because it occurs in colloidal form or is hydrolysed. In the real preconcentration situation, the non-filterable Fe is kept in solution because of the lack of oxygen, which would oxidize it.

The results of the comparison of the two different column types are shown in Fig. 3. It can be seen that in general the results for all elements agree well, although the Dionex results are systematically somewhat higher. The reason for this is not known. Although the Waters column contains metal parts, there is no sign of contamination. The low concentrations also agree well. In practical use, the Dionex columns have been
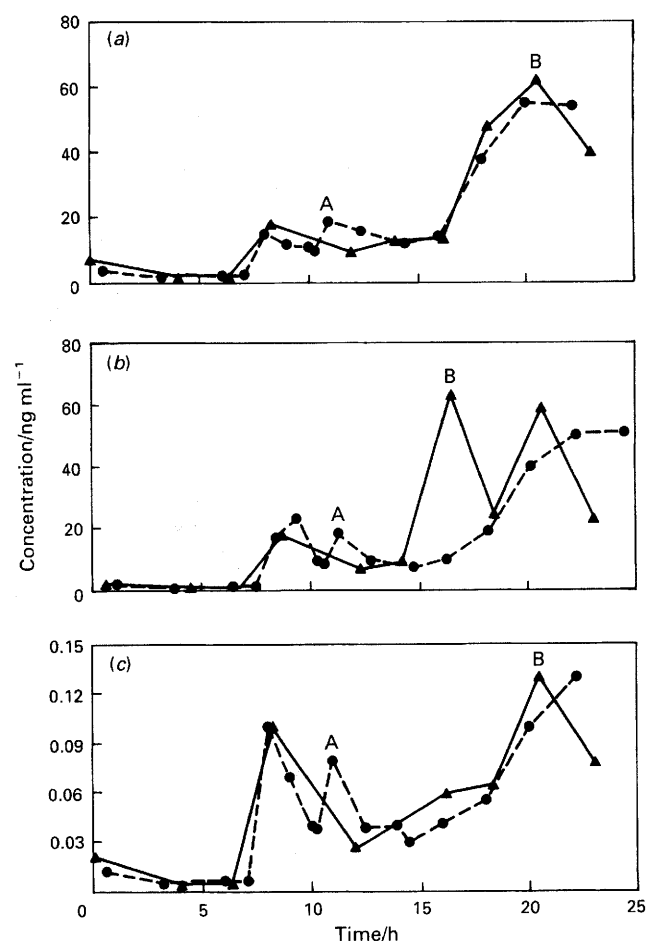

Fig. 2 Metal concentrations of samples obtained from the Loviisa 1 shut-down using A, standard and B, isokinetic sampling lines in parallel for: (a) Mn; (b) Ni; and (c) Co. Time scale in hours from a reference time

Table 3 Results of the concentrations tests with a Waters Guard Pak column: the yield of the elements and the total capacity of the column

Parameter	Fe	Mn	Ni	Co
Yield from 10 ml of water (%)	44	100	99	100
Yield from 20 ml of water (%)	34	100	100	100
Yield from 50 ml of water (%)	7	89	97	97
Capacity/ng	560	1210	9800	27
Corresponds to x ml of water	10	94	92	90

more reliable, the capillary connections of the Waters columns tending to be clogged at times.

ICP-MS Determinations

The results of the interference of Ni on Co when an area of 0.6 m/z is used for integration are shown in Table 4. This means that 1 ng of Ni corresponds to 7×10^{-5} ng of Co. A Ni to Co ratio of 14 000 would cause a positive error of 10% in the Co result. In the results presented, the greatest Ni to Co ratio is 400. When a 0.4 m/z integration area is used, no effect can be detected.

The reproducibility of the ICP-MS results is shown in Table 5. The results indicate that the long-term stability of the concentrated samples is good.

The detection limits for the different metals depend mainly on the variation of the blanks. Using the aluminium sampler and the 10 s measurement technique, detection limits can be as good as 1 ng ml^{-1} for Fe, 0.1 ng ml^{-1} for Mn, 0.1 ng ml^{-1} for Ni and 0.010 ng ml^{-1} for Co. With concentration of a 1 l sample to 2 ml, the sample primary water detection limits are correspondingly: Fe 2, Mn 0.2, Ni 0.2 and Co 0.02 ng l^{-1}. Naturally, accurate results can only be obtained with concentrations at ten times these levels, but even then the limits are clearly below what is normally found in these samples.

The concentrations of the soluble metal ions during the refuelling shut-downs of Loviisa 1 in 1992 are shown in Fig. 4. Some values of the concentrations in the primary water of the Loviisa 1 and Loviisa 2 power plants in 1993 during full power are shown in Table 6. The concentrations of the elements show similar behaviour to results reported from other reactors.[1,7,15,17] The TV04 samples the water in the purification circuit before the ion exchangers and TVO8 after the water has passed the ion exchangers. The results for soluble Ni and

Table 4 Effects of Ni on the concentration of Co

| Ni/ng ml^{-1}	Co/ng ml^{-1}
100	0.008
200	0.011
400	0.025

Table 5 Transition metal concentrations (ng ml^{-1}) of samples taken during the Loviisa 2 (LO2) shut-down. Parallel measurements made with an interval of 3 months

Sample	Fe	Fe	Mn	Mn	Ni	Ni	Co	Co
LO2/6	—	—	0.91	0.89	0.13	0.13	0.0045	0.004
LO2/10	0.11	0.21	2	2.2	0.36	0.43	0.007	0.006
LO2/13	1.7	1.5	2.6	3	0.71	0.87	0.009	0.011
LO2/16	4.5	4	3.6	3.7	1.3	1.4	0.017	0.017
LO2/19	1.2	0.45	2.3	2.2	0.75	0.83	0.01	0.008
LO2/41	11.5	13.6	3.9	3.7	3.6	3.8	0.019	0.018
LO2/44	14	13.7	4.3	3.7	3.5	3.2	0.02	0.017
LO2/47	29	28	7.7	5.4	7.8	6.2	0.032	0.024
LO2/50	18.5	12	11.1	7.5	7.5	6	0.023	0.014
LO2/53	32	44	30	18	27	25	0.069	0.048
LO2/56	35	22	28	18	43	52	0.11	0.074
LO2/60	30	19	23	14	41	49	0.11	0.076

Co are of the same order of magnitude as the full power values for other reactors as reported in the literature.[1,7,8,15,16,17] The range of results is as follows: Ni 10–300 ng l^{-1} with a few values above 500 ng l^{-1}; and Co 0.4–10 µg l^{-1} with a few values considerably above these. The work is continuing with further development of the sampling and analysis methods. More results together with a discussion of the significance of the results will be published later.

Conclusions

A new method has been developed that enables the sampling and determination of Fe, Mn, Ni and Co in the primary water of PWRs using ICP-MS. The soluble and insoluble fractions are analysed separately. The fast transients in the concentrations of Mn, Ni and Co occuring during shut-down can be followed. This fact, together with the detected decrease of the transition metal concentrations in the water going through the purification system, seems to indicate that the sampling and
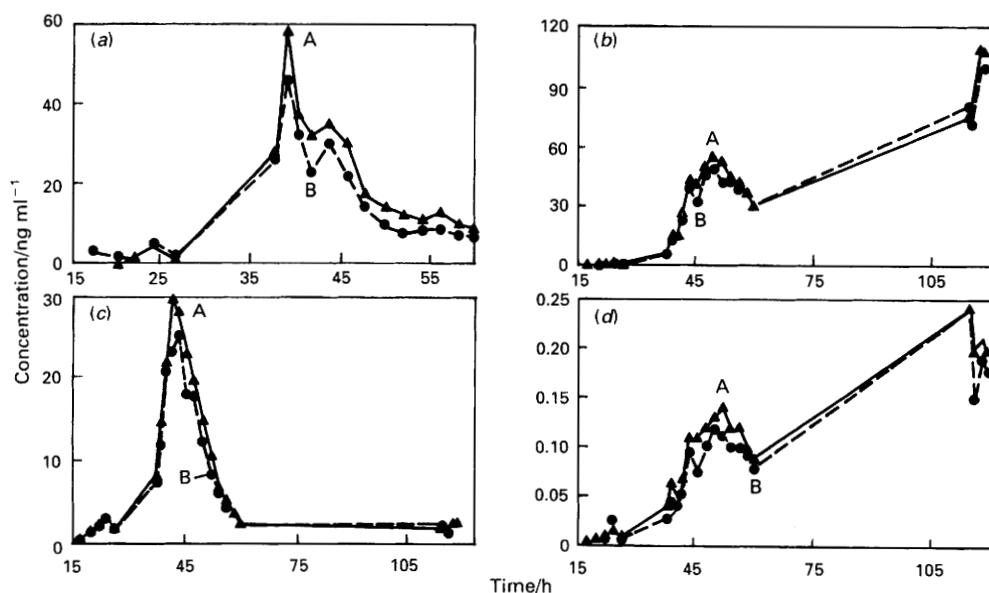
Fig. 3 Comparison of the results obtained by preconcentration of parallel samples during the Loviisa 2 shut-down using the columns A, Dionex MetPac CC-1 and B, Waters Guard Pak for (a) Fe; (b) Ni; (c) Mn; and (d) Co
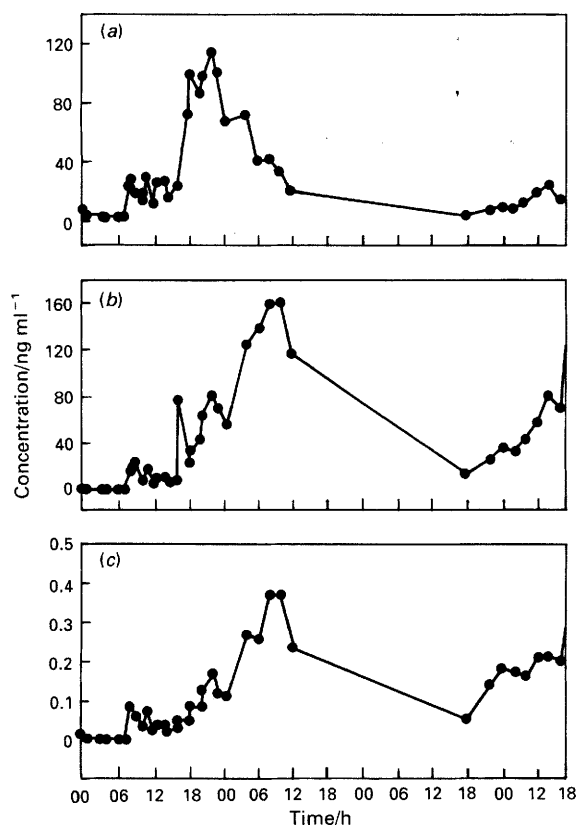
Fig. 4 Soluble transition metal concentrations (a) Mn, (b) Ni and (c) Co in the primary water of the Loviisa 1 power plant during the 1992 re-fuelling shut-down. Time in hours from noon the day before shut-down (July 18, 1992) to July 21, 1992. The reactor reached sub-criticality on July 18, 1993 at 6.46 hours

Table 6 Soluble transition metals (in ng l^{-1}) in the Loviisa 1 and Loviisa 2 primary water during full power in 1993

Sampling date	Fe	Mn	Ni	Co
Loviisa 1, TVO4:				
22–23 June, 1993	460	54	17	0.9
23–24 June, 1993	270	48	8	0.8
Loviisa 2, TVO4:				
3–4 February, 1993	2300	112	171	7.8
4–5 February, 1993	2400	103	171	3.8
11–12 February, 1993	2000	101	169	3.7
16–17 February, 1993	1700	71	110	2.7
28–29 April, 1993	1260	81	117	3.4
Loviisa 2 TVO8:				
29–30 April, 1993	73	10	22	0.55

analysis is representative, enabling the follow-up of concentration gradients occuring in different parts of the primary circuit. The detection limits are low enough to enable the determination of the elements during full power in all reactors.

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