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Supplement 5 to Annual Report STUK-A89

M. Puhakainen and T. Rahola

SÄTEILYTURVAKESKUS
Strålsäkerhetscentralen

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ABSTRACT

Sludge samples from wastewater treatment plants were studied by the Finnish Centre for Radiation and Nuclear Safety from 1979 onwards. Sampling of sludge was extended to include more sewage treatment plants after the accident at the Chernobyl nuclear power station. The study was continued in some of the wastewater treatment plants in order to continuously follow the level of and changes in the fallout radioactivity. Sludge samples were also taken from treatment plants in communities close to the nuclear power stations at Loviisa and Olkiluoto.

For a long time the most frequently detected nuclide in sewage sludge was ^{137}Cs originating from Chernobyl. The ^{137}Cs activity concentration in sludge varied in 1988 from 68 to 750, in 1989 from 16 to 480 and in 1990 from 11 to 300 Bq kg^{-1} dry weight. The activation products in sludge originating from nuclear power stations in Finland were some becquerels per kilo, at the most about twenty becquerels per kilo dry weight. The most frequently detected medical radionuclide was ^{131}I, frequently detected in almost all wastewater treatment plants.

CONTENTS

1 INTRODUCTION

Sludge samples from wastewater treatment plants were studied for radionuclide content by the Finnish Centre for Radiation and Nuclear Safety from 1979 onwards. At first samples were collected from cities, where hospitals use large amounts of radionuclides and from communities located in the vicinity of Finnish nuclear power stations.[4-5] Sampling of sludge was extended to include more sewage treatment plants after the accident at the Chernobyl nuclear power station. In 1986 samples were received from 25 communities. The highest concentration of ^{137}Cs measured in sludge was 12000 Bq kg^{-1} dry weight.[6] In 1987 the number of samples tested decreased and the highest ^{137}Cs activity measured was 1800 Bq kg^{-1} dry weight.[7]

From 1988 on the study was continued in some of the wastewater treatment plants in order to continuously monitor the level of and changes in the fallout radioactivity.

Sludge samples were also taken from treatment plants in communities close to the nuclear power stations at Loviisa and Olkiluoto to detect radionuclides originating from these stations. Such samples were taken and analysed from 1984 onwards.

The Finnish Centre for Radiation and Nuclear Safety has also studied the transfer to wastewater treatment plants of ^{131}I used in hospitals. In that study the samples were taken primarily from wastewater arriving at the plant. The results of this wastewater sampling will be published in a separate report later. The results of the measurements of sludge samples taken at the same time as the wastewater samples are, however, given in this report.

2 MATERIAL AND METHODS

In most of the wastewater treatment plants studied, sewage is treated using a biological–chemical simultaneous precipitation method in which phosphorus compounds are precipitated by adding compounds of iron, aluminium or calcium.[10] The most commonly added compound is ferrosulphate. Almost all the plants studied use mechanical dewatering which allows the concentration of dry material in the sludge to reach levels of 15–25%. In the plants in Helsinki, Espoo and Kuopio a digestion process, involving a 20–30 day residence time for the sludge, is used before drying.

The sludge samples were take from 10 wastewater treatment plants. The sampling locations are shown in Figure 1; and data on treatment methods, daily discharges and amounts of sludge produced are given in Table I. The treatment plant at Loviisa is situated about 10 km from the nuclear power plants, at Loviisa and those at Rauma and Eurajoki about 10 km from the nuclear power plants at Olkiluoto. Some samples were dried at a temperature of 105 °C, homogenised and measured gammaspectrometrically using a cylindrical geometry for approximately 1000 min to detect even minute amounts of radionuclides. Other samples were packed without pretreatment in Marinelli beakers of about half a litre and measured with lithium-drifted or high-purity Ge detectors. The measurement time for these samples was 60–400 minutes.

The calculated activities were corrected for physical decay back to the midpoint of the collecting period. The percentage of dry material in all the samples was determined after drying the sludge at 105 °C.

There is a residential area for maintenance workers close to the Loviisa nuclear power plant. The sewage from these houses flows into the local wastewater treatment plant. The sewage is treated chemically using $AlSO_4$ – precipitation. The sludge formed in the septic tank has a dry weight of about 4 per cent and was transported to a dumping site once a week. As the tank is not always pumped complety empty, older sludge may be mixed with more recent matter, and the samples taken may consist of a mixture of both old and new sludge. Samples were taken from the septic tank during the maintenance and refuelling periods at the nuclear power plant in 1988–90.

3 RESULTS

The activity concentrations of gamma-emitting radionuclides in the dewatered sewage sludge samples taken at different treatment plants in 1988-1990 are shown in Tables II-IV. The ^{137}Cs activity concentration varied in 1988 from 68 to 750, in 1989 from 16 to 480, and in 1990 from 11 to 300 Bq kg^{-1} dry weight. The variations in ^{137}Cs activity concentrations between 1986 and 1990 at the wastewater treatment plants in Rauma (Maanpääniemi), Loviisa (Vårdö) and Helsinki (Kyläsaari) are shown in Figures 2 and 3. In 1988 radionuclides other than cesium also detected in sludge were ^{106}Ru, ^{125}Sb and ^{144}Ce originating from the Chernobyl fallout. After 1988, of the fallout nuclides, only ^{134}Cs and ^{137}Cs were detected. In 1990 ^{125}Sb was detected in one sample. In July 1990 the ^{137}Cs activity concentration in sludge from Helsinki (Kyläsaari) was about 40% of the corresponding concentration in 1988. The ratio of the ^{134}Cs and ^{137}Cs activity concentrations decreased from 0.28 in 1988 to 0.16 in 1990.

Between 1988 and 1990 small amounts of ^{51}Cr, ^{125}Sb, ^{54}Mn, ^{58}Co and ^{60}Co were detected in the Eurajoki wastewater treatment plant during the annual maintenance shutdown periods of the nuclear power station at Olkiluoto. At Rauma only ^{54}Mn and ^{60}Co of the above mentioned nuclides were detected. At Loviisa ^{110m}Ag was detected during every maintenance shutdown period. In 1988 ^{54}Mn, ^{58}Co and ^{60}Co were also detected. The activity concentrations in sludge from the septic tank for the residential area for maintenance workers near the Loviisa nuclear power station are given in Table V. The activation products ^{51}Cr, ^{54}Mn, ^{58}Co, ^{60}Co, ^{59}Fe, ^{110m}Ag and ^{124}Sb were detected during the annual maintenance shutdown period. Fission products ^{95}Zr, ^{106}Ru, ^{131}I, ^{141}Ce and ^{144}Ce were detected in 1989 and 1990.

Of the gamma-emitting radionuclides used in medicine, ^{51}Cr, ^{57}Co, ^{59}Fe and ^{131}I were detected in sewage sludge. Of the short-lived radionuclides used in medicine ^{111}In ($T_{1/2}$ = 2.8 d), ^{67}Ga ($T_{1/2}$ = 3.3 d) and ^{201}Tl ($T_{1/2}$ = 3.1 d) were occasionally detected in treatment plants. The most frequently detected medical radionuclide was ^{131}I, which was found in almost all wastewater treatment plants. The greatest concentration of ^{131}I 7900 Bq kg^{-1} dry weight was found on March 15, 1989 in Lappeenranta. At a Lappeenranta hospital patients were given ^{131}I for therapy on March 9 and 10, 1989. The variations in ^{131}I activity concentration in sludge samples from Helsinki (Kyläsaari) are shown in Figure 4. Concentrations varied from 31 to 220 Bq kg^{-1} dry weight.

4 DISCUSSION

The amounts of fallout radionuclides in sewage sludge are influenced by the fallout level in the sewer area, the amounts of rain and runoff, the type of runoff area and the treatment method used on the sewer water. Also the activity of tapwater and of foodstuffs influence the activity concentration as do the Al-precipitation produced in waterworks and transported to wasterwater treatment plants and the sludge from industrial prosesses.

The inhabitants of the Helsinki area get their tap water partly from different fallout regions. In the Helsinki area there are two wastewater treatment plants, at Kyläsaari and Suomenoja. The sewer area served by those plants belongs to the same fallout region. Both treatment plants use the same treatment method, digestion and mechanical dewatering of the sludge. The difference between them is that rain and runoff flow to the Kyläsaari treatment plant from a very large area whereas at Suomenoja no rain water enters the treatment process. A special source of activity in the Kyläsaari treatment plant is the Al-precipitation produced in waterworks using raw water from a lake in the highest fallout region. [6-7] The amounts of sludge at the Kyläsaari plant are double those at the Suomenoja plant.[10] For a long time the two most frequently detected nuclides in sewage sludge were ^{134}Cs and ^{137}Cs orginating from Chernobyl. The ^{137}Cs concentrations at the Suomenoja plant were approximately one tenth of those at the Kyläsaari plant during the 1986–1989 period. Further investigations will give information on the importance of the different factors influencing radionuclide concentrations of sewage sludge in the Helsinki area.

In Sweden ^{137}Cs levels in sludge samples taken in 1989–90 from a wastewater treatment plant at Lund were studied by Isaksson et al.[3] They estimated that about 35 per cent of the total ^{137}Cs activity in the treatment plant originated in excreta from the inhabitants of the Lund area and the remainder came mostly from runoff.

The activation products in sludge originating from nuclear power stations in Finland amounted to a few becquerels per kilo, at the most about twenty becquerels per kilo dry weight. Sludge was produced at the treatment plant in Rauma at a rate of about 14–30 m^3, at Eurajoki about 9 m^3 and at Loviisa 3–4 m^3 per day. This usually corresponded to less than 10 kBq/d during the maintenance and refuelling period. Many workers at the Loviisa power station live in Loviisa, and many workers at the Olkiluoto power station live in Eurajoki or Rauma. The control sites used were Kotka, within a c. 35 km radius of the Loviisa nuclear power station, and Pori, within a c. 35 km radius of the Olkiluoto nuclear power station. During the maintenance, refuelling and shutdown period at the nuclear power stations, of the artificial radionuclides only ^{131}I and further nuclides originating in Chernobyl was detected in the wastewater treatment plants in Kotka and Pori.

The greatest activity concentration in sludge came from medically used ^{131}I. In Lappeenranta and Kuopio the sampling was scheduled at times when hospitals used large amounts of ^{131}I for therapy. The Kuopio treatment plant uses a digestion process, involving a 25 day residence time for sludge. This caused a decrease in the activity concentration before sampling. The decrease in the ^{131}I concentration at Helsinki (Kyläsaari) after 1987 was partly caused by the University hospital in Helsinki beginning to use ^{123}I instead of ^{131}I for therapy.[2] The greatest amount of ^{51}Cr collected daily in sludge was 7500 kBq. This was calculated assuming the daily sludge production to be about 130 m^3. With a residence time of about 25 d, 14 MBq/d was collected in the sludge.

Annually 1 100 000 m^3 dewatered sewage sludge is produced in Finland. Of this 75% is utilized; 50% for fertilizing agricultural fields and 25% for landscaping. We studied the uptake of ^{137}Cs and observed that only a very small amount was transferred from soil supplemented with sludge to cereal.[8] The rules on the use of sewage sludge on fields were changed recently. The maximum acceptable amount of sludge is one metric ton of dry matter per hectare, or four tonnes every fourth year.[9] The change was made to reduce the levels of heavy metals in fields.

The contribution of fallout radiocesium in sludge to the total radiation dose was considered insignificant.

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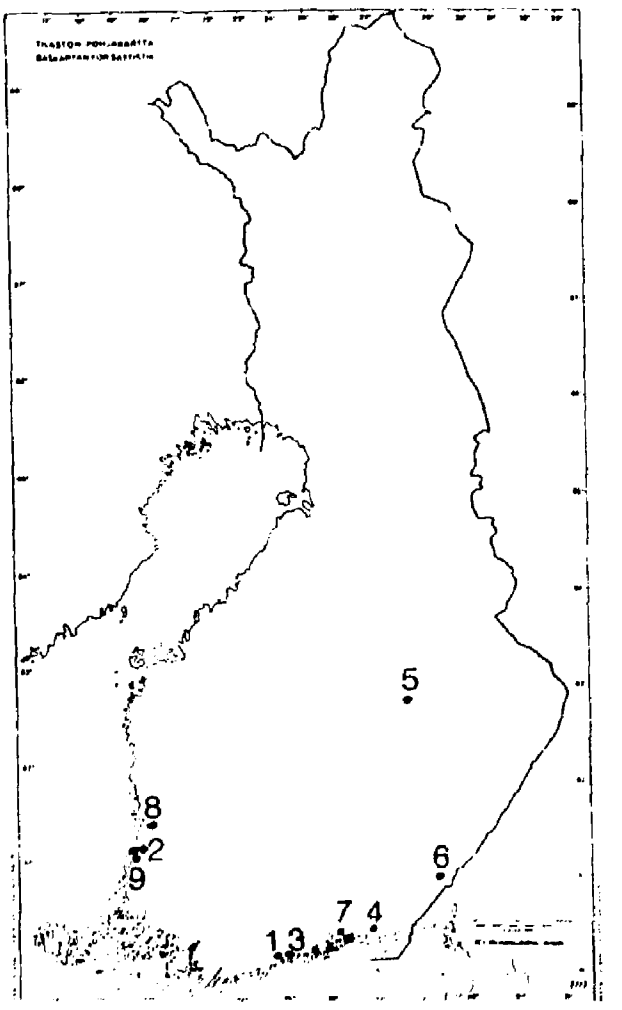

Fig. 1. The sampling sites for sewage sludge

1. Espoo, Suomenoja
2. Eurajoki, Kirkonkylä
3. Helsinki, Kyläsaari
4. Kotka, Mussalo
4. Kotka, Sunila
5. Kuopio, Lehtoniemi
6. Lappeenranta, Toikansuo
7. Loviisa, Vårdö
8. Pori, Luotisinmäki
9. Rauma, Maanpäänniemi

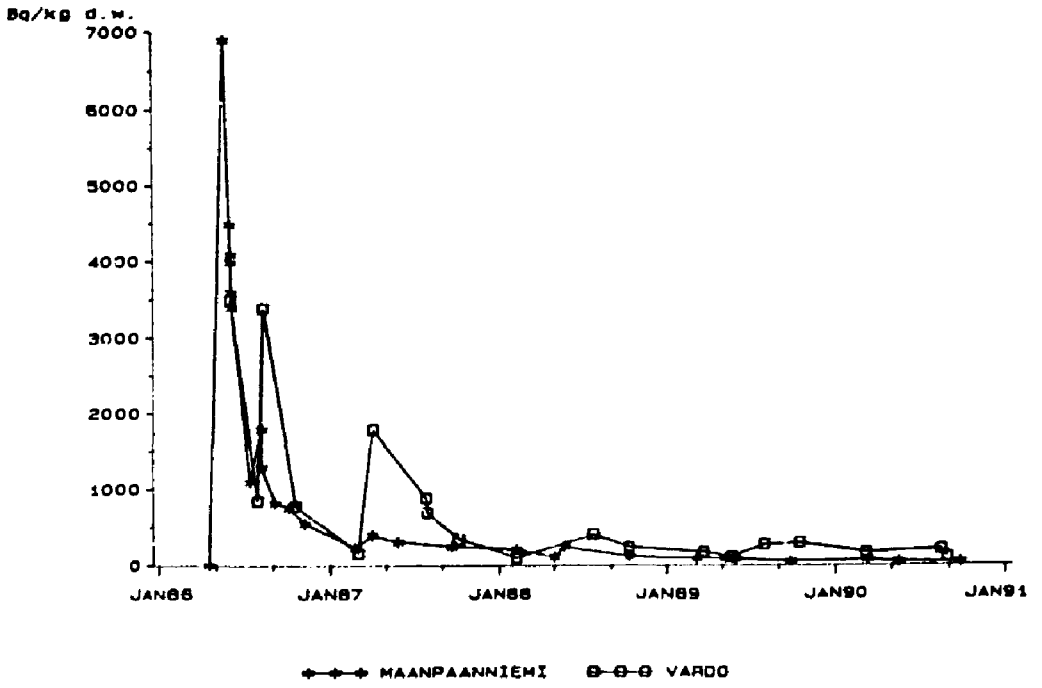

Fig.2. Concentrations of ^{137}Cs in sewage sludge at Rauma (Maanpäänniemi) and at Loviisa (Vårdö) in 1986–1990.

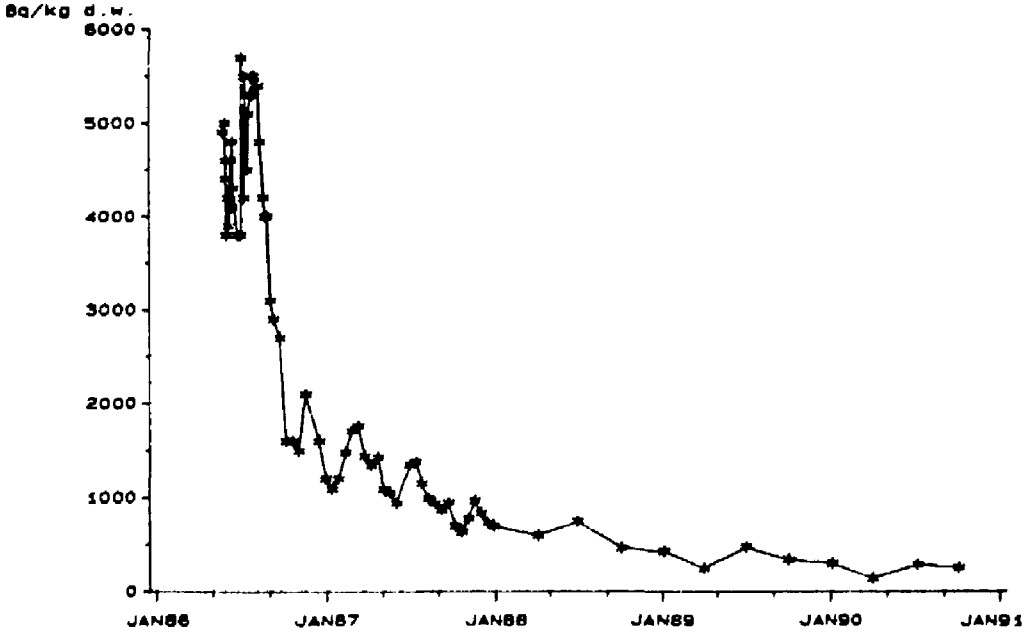

Fig.3. Concentrations of ^{137}Cs in sewage sludge at Helsinki (Kyläsaari) in 1986–1990.

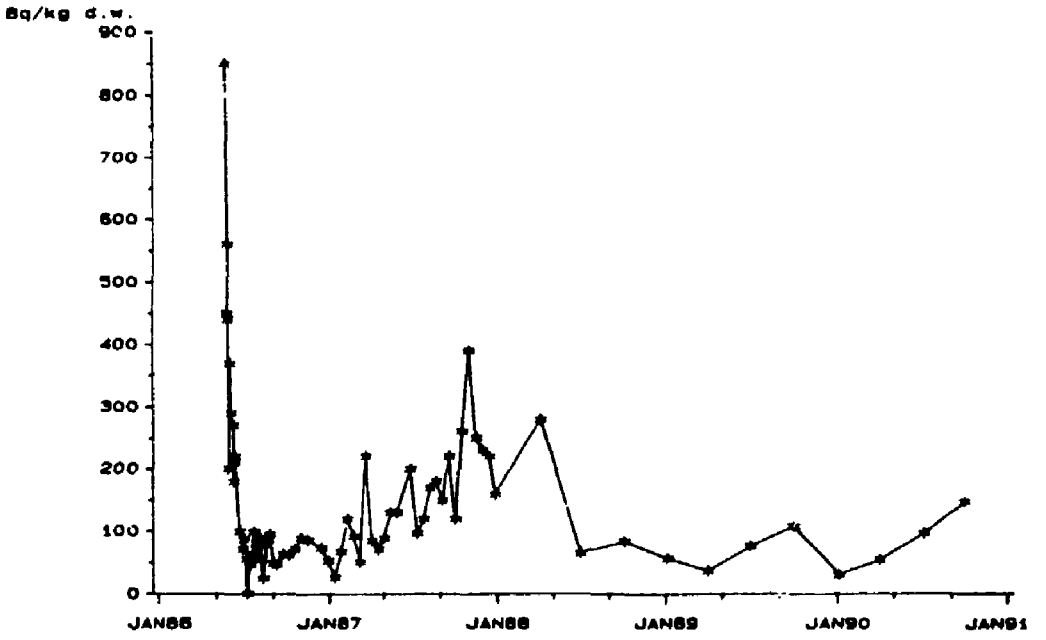

Fig.4. Concentrations of ^{131}I in sewage sludge at Helsinki (Kyläsaari) in 1986–1990.

Table I. Treatment methods, mean values for dry matter in sludge, daily discharges and estimated amounts of sludge produced at wastewater treatment plants.[10]

	Treatment method	Dry matter %	Discharge $m^3\ d^{-1}$	Amount of sludge $m^3\ d^{-1}$	Fallout region
Espoo, Suomenoja	RS[a]	22	70865	70	1
Eurajoki, Kirkonkylä	RS	1.7	598	9	3
Helsinki, Kyläsaari	RS	18	111462	134	1
Kotka, Mussalo	RS	16	11869	23	4
Kotka, Sunila	RS	15	13556	22	4
Kuopio, Lehtoniemi	RS	28	25830	18	3
Lappeenranta, Toikansuo	ES[b]	8	18090		1
Loviisa, Vårdö	SS[c]	19	2682	3–4	4
Pori, Luotsinmäki	KS[d]	17	30388	82	3
Rauma, Maanpäänniemi	RS	14	8869	14–24	3

[a] simultaneous precipitation
[b] pre-precipitation
[c] direct precipitation
[d] chemical treatment

Table II. Gamma-emitting radionuclides in sewage sludge (Bq kg^{-1} dry wt.) in 1988.

Wastewater treatment plant	Sampling date	Measuring time min.	Dry matter %	^{7}Be	^{40}K	^{51}Cr	^{54}Mn	^{57}Co	^{58}Co	^{60}Co
Eurajoki	23.5.88[a]	1010	1.7	0[c]	240	0	4.4	0	0	7.7
Helsinki,	5.4.88	82	19.9	120	280	220	0	11	0	0
Kyläsaari	1.7.88	60	21.9	300	310	100	0	110	0	0
	3.10.88	99	14.9	380	190	290	0	13	0	0
Kotka, Mussalo	10.10.88	4033	16.8	44	140	0	0	0	0	0
Kotka,	8.–10.2.88	1017	16.6	64	74	0	0	0	0	0
Sunila	25.7.88	1053	14.2	34	55	0	0	0	0	0
	10.10.88	1065	13.7	56	–	0	0	0	0	0
Kuopio,	18.9.88	407	27.3	110	0	0	0	0	0	0
Lehtoniemi	19.9.88	403	30.0	81	0	0	0	0	0	0
	20.9.88	121	26.9	56	74	0	0	0	0	0
	21.9.88	422	27.8	47	75	0	0	0	0	0
Loviisa,	8.2.88	1033	17.5	39	84	0	0	0	0	0
Vårdö	25.–28.7.88[b]	1043	17.0	110	130	0	11	0	6.6	6.3
	10.10.88	1080	18.8	110	160	0	0	0	0	2.4
Pori,	2.2.88	1028	16.9	35	39	0	0	0	0	0
Luotsin-	23.–26.5.88	1019	18.9	51	75	0	0	0	0	0
mäki	10.10.88	1093	16.9	42	0	0	0	0	0	0
Rauma,	8.2.88	3910	12.0	140	140	0	0	0	0	2.0
Maanpään-	2.5.88[a]	1021	11.4	120	150	0	0	0	0	0
niemi	23.–26.5.88[a]	1079	11.3	160	140	0	5	0	0	3.8
	10.10.88	1102	11.7	120	110	0	0	0	0	0

Table II. cont.

^{106}Ru	^{110m}Ag	^{125}Sb	^{131}I	^{134}Cs	^{137}Cs	^{144}Ce
0	0	0	0	32	140	0
0	0	0	280	180	600	0
0	0	0	66	220	750	0
0	0	0	85	120	470	0
0	0	8.9	110	57	210	0
0	0	9.9	7.0	68	210	0
14	0	9.7	6.5	69	240	0
0	0	12	0	80	300	0
0	0	0	390	21	100	0
0	0	0	210	20	100	0
0	0	0	830	21	98	0
0	0	0	660	25	94	0
0	0	0	0	35	100	0
0	5.5	7	29	120	410	0
0	0	4.7	570	65	240	0
0	0	0	0	23	68	0
0	0	0	7.7	41	150	0
0	0	0	0	40	150	0
69	0	17	0	70	210	0
43	0	0	0	35	110	0
45	0	12	71	71	250	24
28	0	14	0	32	120	0

Annual maintenance shutdown period

[a] Olkiluoto nuclear power station 30.4.–9.6.1988

[b] Loviisa nuclear power station 24.6.–23.8.1988

[c] below detection limit

Table III. Gamma-emitting radionuclides in sewage sludge (Bq kg^{-1} dry wt.) in 1989.

Wastewater treatment plant	Sampling date	Measuring time min.	Dry material %	^{7}Be	^{40}K	^{51}Cr	^{57}Co	^{59}Fe	^{60}Co	^{110m}Ag	^{131}I	^{134}Cs	^{137}Cs
Espoo, Suomenoja	14.3.89	168	24.7	65	87	0[c]	0	0	0	0	5	11	42
Eurajoki, Kirkonkylä	15.5.89[a]	1165	1.3	0	200	0	0	0	7	0	0	15	66
	29.5.89[a]	4018		27	190	30	0	0	19	0	0	15	66
Helsinki, Kyläsaari	4.1.89	120	19.1	94	160	200	0	0	0	0	57	110	430
	3.4.89	76	22.5	310	250	95	0	0	0	0	37	54	250
	2.7.89	60	17.3	220	0	200	17	16	0	0	77	100	480
	2.10.89	58	16.4	160	0	97	14	0	0	0	110	60	340
Kotka, Mussalo	6.3.89	4052	13.6	93	140	0	0	0	0	0	48	60	250
Kotka, Sunila	6.3.89	4012	10.7	77	120	0	1	0	0	0	310	44	190
	31.7.–3.8.89	1114	15.3	0	110	0	0	0	0	0	0	27	120
Lappeenranta, Toikansuo	15.3.89		7.9	0	0	0	0	0	0	0	7900[d]	0	16

Table III cont.

Wastewater treatment plant	Sampling date	Measuring time min.	Dry material %	^{7}Be	^{40}K	^{51}Cr	^{57}Co	^{59}Fe	^{60}Co	^{110m}Ag	^{131}I	^{134}Cs	^{137}Cs
Loviisa,	21.3.89	1025	17.1	71	130	0	0	0	0	0	0	45	180
Vårdö	22.5.89	1034	–	67	0	0	0	0	0	0	11	28	110
	31.7.–3.8.89[b]	1128	18.5	160	140	0	0	0	0	10	0	59	280
	16.10.89	1002	19.2	150	220	0	0	0	0	0	0	50	300
Pori,	6.3.89	671	18.2	88	77	0	0	0	0	0	0	15	72
Luotsinmäki	15.–18.5.89	4147	18.4	26	0	0	0	0	0	0	13	11	55
	29.5.–1.6.89	1077	18.8	0	0	0	0	0	0	0	8	13	60
Rauma,	6.3.89	1772	11.7	150	230	0	0	0	0	0	57	21	92
Maanpäänniemi	8.–11.5.89[a]	1062	10.4	94	0	0	4.7	0	0	0	76	18	98
	29.5.–1.6.89[a]	1060	13.1	87	110	0	9.8	0	4	0	110	18	86
	25.–28.9.89	1040	13.3	0	0	0	0	0	0	0	0	8.8	49

[a] Annual maintenance shutdown period at Olkiluoto nuclear power station 30.4.–15.5.1989 and 20.5.–31.5.1989
[b] Annual maintenance shutdown period at Loviisa nuclear power station 9.7.–21.8.1989
[c] below detection limit
[d] at a nearby hospital, patien were given ^{131}I therapy on March 9 and 10, 1989.

Table IV. Gamma-emitting radionuclides in sewage sludge (Bq kg^{-1} dry wt.) in 1990.

Wasterwater treatment plant	Sampling date	Measuring time min.	Dry material %	^{7}Be	^{40}K	^{51}Cr	^{54}Mn	^{57}Co	^{58}Co	^{60}Co	^{110m}Ag	^{125}Sb	^{131}I	^{134}Cs	^{137}Cs
Eurajoki,	15.5.90[a]	1012	2.2	0[c]	150	0	0	0	0	6.3	0	0	0	6.4	33
Kirkonkylä	28.5.90[a]	3843	1.9	0	47	4	1.2	0	0.8	1.7	0	0	0	1.8	11
Helsinki,	4.1.90	1097	19.5	170	200	130	0	4.2	0	0	0	0	31	53	300
Kyläsaari	2.4.90	116	21.9	160	260	110	0	0	0	0	0	0	55	23	140
	6.7.90	192	20.8	150	230	81	0	28	0	0	0	0	98	46	290
	1.10.90	126	17.6	230	120	86	0	0	0	0	0	0	150	32	250
Kotka,	12.3.90	1513	13.5	68	110	0	0	0	0	0	0	0	370	28	150
Mussalo	3.–6.9.90	335	20.7	0	52	0	0	0	0	0	0	7	87	10	65
Kotka,	12.3.90	3902	14.1	71	86	0	0	0	0	0	0	0	0	27	140
Sunila	3.–6.9.90	341	19.7	0	0	0	0	0	0	0	0	0	25	11	74
Loviisa,	8.3.90	1147	17.4	120	210	0	0	0	0	0	0	0	66	32	170
Vårdö	15.8.90[b]	227	20.0	120	–	0	0	0	0	0	4	0	6	31	220
	27.–30.8.90[b]	368	17.6	63	–	0	0	0	0	0	5	0	0	17	110

Table IV. cont

Wasterwater treatment plant	Sampling date	Measuring time min.	Dry material %	^{7}Be	^{40}K	^{51}Cr	^{54}Mn	^{57}Co	^{58}Co	^{60}Co	^{110m}Ag	^{125}Sb	^{131}I	^{134}Cs	^{137}Cs
Pori,															
Luotsinmäki	12.3.90	1213	18.3	31	71	0	0	0	0	0	0	0	0	7.1	42
	14.–17.5.90	3855	19.5	22	41	0	0	0	0	0	0	0	0	5.4	32
	25.9.90	1098	18.3	27	0	0	0	0	0	0	0	0	4	7.3	41
Rauma,	12.3.90	1077	15.5	110	150	0	0	0	0	0	0	0	26	11	74
Maan-	14.–17.5.90[a]	305	15.2	26	100	0	0	0	0	0	0	0	100	5.7	45
päänniemi	25.9.90	359	13.1	30	81	0	0	0	0	0	0	0	0	6.7	43

[a] Annual maintenance shutdown period at Olkiluoto nuclear power station 6.5.–9.6.1990
[b] Annual maintenance shutdown period at Loviisa nuclear power station 28.7.–24.9.1990
[c] below detection limit

Table V. Gamma-emitting radionuclides in sludge (Bq kg^{-1} dry wt.) from the septic tank at the housing area in the vicinity of the Loviisa nuclear power plant.

Sampling date	^{51}Cr	^{54}Mn	^{58}Co	^{59}Fe	^{60}Co	^{95}Zr	^{106}Ru	^{110m}Ag	^{124}Sb	^{131}I	^{134}Cs	^{137}Cs	^{141}Ce
13.7.88	47	71	270	27	190	0	0	170	180	17	14	61	0
19.7.88	40	33	130	13	100	0	0	160	160	5.7	5.1	22	0
27.7.88	46	90	210	33	180	0	0	260	180	7.2	15	61	0
27.7.89	0	14	48	0	57	0	0	350	120	0	10	49	0
3.8.89	0	12	32	0	49	22	24	-	72	22	15	69	0
10.8.89	50	15	43	0	71	32	39	240	70	0	15	84	20
15.8.90	23	11	75	28	46	11	0	320	67	0	4.6	28	0
29.8.90	28	75	80	36	120	14	20	290	94	0	8	47	0
5.9.90	0	23	46	0	68	0	12	240	74	0	0	0	0

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