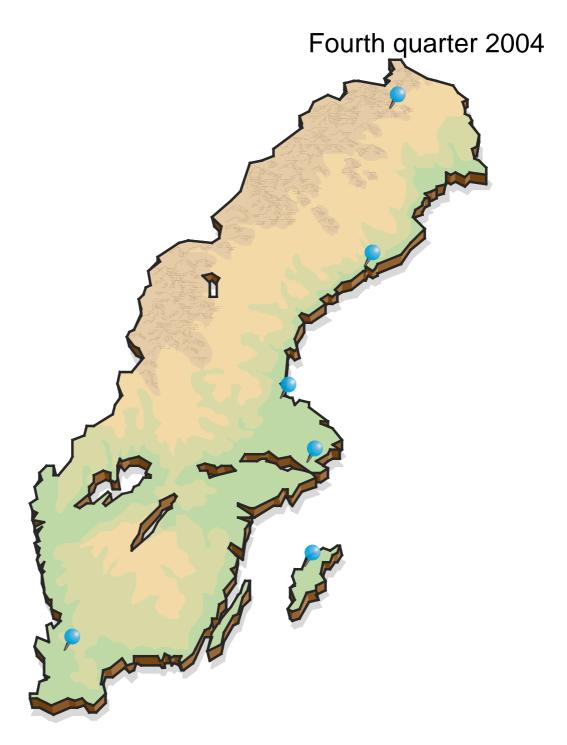


Catharina Söderström Rune Arntsing Peter Jansson Karin Lindh Katarina Wilhelmsen Detection of ¹⁹²Ir in ground level air in Ursvik 20 September 2004 with Quarterly report on measurements of radionuclides in ground level air in Sweden



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Detection of ¹⁹²Ir in ground level air in Ursvik 20 September 2004 with

Quarterly report on measurements of radionuclides in ground level air in Sweden

Fourth quarter 2004

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Detection of ¹⁹² Ir in ground level air in Ursvik 20 S of radionuclides in ground level air in Sweden. Fo		/ report on measurement		
Abstract (not more than 200 words)				
The radioisotope ¹⁹² Ir was found in the filter from ndications of ¹⁹² Ir in filters from any of the other ractivity concentration of ¹⁹² Ir was about 50 μBq/m	national stations during that pe	eriod. The measured		

indications of ¹⁹²Ir in filters from any of the other national stations during that period. The measured activity concentration of ¹⁹²Ir was about 50 μ Bq/m³, corresponding to a total collected activity of 2.36 Bq of ¹⁹²Ir in the filter. The source of the release was the Studsvik Nuclear AB production site about 80 km south of Ursvik and the preliminary estimation of released activity was 58 kBq. Locally performed measurements and dispersion calculations show that the released amount of ¹⁹²Ir was in the order of 1 GBq. This is in agreement both with the measurements at the filter station in Ursvik, and with the dispersion calculations performed by FOI.

Filtering of ground level air is performed weekly at six different locations in Sweden: Kiruna, Umeå, Gävle, Ursvik, Visby and Ljungbyhed. The filters are pressed and the contents of different radionuclides are measured by gamma spectroscopy. Precipitation is also collected at four of the stations: Kiruna, Gävle, Ursvik and Ljungbyhed, the samples are ashed and the contents of radionuclides are measured. The levels of Be-7 and Cs-137 in air and precipitation are presented for the different stations. Other antropogenic radionuclides detected, if any are also presented.

Keywords Airborne radionuclides, deposition, ⁷Be, ¹³⁷Cs, ¹³¹I, ¹⁹²Ir Further bibliographic information Language English ISSN 1650-1942 Pages 12 p. Price acc. to pricelist

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Sammanfattning (högst 200 ord) Isotopen ¹⁹² Ir detekterades i filter från Ursvik, månda nationella stationerna visade på förekomst av ¹⁹² Ir ur av ¹⁹² Ir var ungefär 50 µBq/m³, motsvarande en total Utsläppet visade sig komma ifrån Studsvik Nuclear A och en preliminär uppskattning av utsläppt mängd va Lokala mätningar och spridningsberäkningar visade Detta stämmer väl med uppmätta mängder i Ursvik o	insamlad mängd av 2.36 E AB produktionsanläggning o ar 58 kBq. att utsläppet av ¹⁹² lr var i st	ea 80 km söder om Ursvik orleksordningen 1 GBq.				
Stationer för filtrering av markluft finns på sex olika ställen i Sverige: Kiruna, Umeå, Gävle, Ursvik, Visby och Ljungbyhed. Filtren pressas och analyseras veckovis med hjälp av gammaspektroskopi med germaniumdetektor. Nederbörd samlas in på fyra av dessa stationer: Kiruna, Gävle, Ursvik och Ljungbyhed. Nedrebördsproven askas in och mäts med hjälp av gammaspektroskopi. Halterna av Be-7 och Cs-137 presenteras för luft och nederbörd för de olika stationerna. I de fall andra antopogena radionuklider detekteras presenteras även dessa.						

Nyckelord

Luftburen radioaktivitet, deposition, ⁷Be, ¹³⁷Cs, ¹³¹I, ¹⁹²Ir

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Detection of ¹⁹²Ir in ground level air in Ursvik 20 September 2004

Background

Sampling of particles in surface air is regularly performed at six national stations in Sweden. The sampling is done by filtering air through a glass fibre filter. The exposed filters are compressed and the radioactivity in the samples thus obtained is measured with gamma spectroscopy to determine the concentration of various radioactive elements. This is done for radiation protection purposes and the measured concentration levels of different radioisotopes are regularly reported to the Swedish Radiation Protection Authority (SSI). If concentrations higher than $100~\mu Bq/m^3$ of any antropogenic radionuclide are found SSI is contacted immediately. This is also the case if lower levels of unusual radioisotopes are found.

At the station in Ursvik in the Stockholm area the air flow through the filter is 1700 m³/hour. The filters are changed automatically every 28:th hour. The six filters exposed every week are compressed individually, but measured together as a weekly sample, and can, if required also be measured individually. The weekly sample is measured 3-4 days after the last filter of the week has been taken down and the duration of the measurement is 3-4 days.

Detection of ¹⁹²Ir

When analysing the filters from week 39, 2004 the radioisotope ¹⁹²Ir was found in the sample from Ursvik. There were no indications of ¹⁹²Ir in filters from any of the other national stations. ¹⁹²Ir is not a naturally occurring radioisotope, and has never before been detected in the samples from any of the national stations.

The filters from Ursvik had been exposed during 20-27 September and the measured amount of ^{192}Ir was about $8.3\pm0.1~\mu\text{Bq/m}^3$ for the weekly sample. After having detected ^{192}Ir in the weekly sample, the filters for that week were remeasured individually. Only one of these filters contained ^{192}Ir . This filter had been exposed from Monday 20 September 08:47 UTC until Tuesday 21 September 12:47, and the filtered air volume through the measured part of the filter was 35 000 m³. The measured activity concentration of ^{192}Ir for the 28 hour period was about 50 $\pm0.7~\mu\text{Bq/m}^3$, corresponding to a total collected activity of $2.36\pm0.03~\text{Bq}$ of ^{192}Ir in the filter

Contact was taken with SSI to reveal if the source of iridium was known. However, SSI had no reports on a release of ¹⁹²Ir.

The short time interval of the presence of iridium activity in the filters, together with the fact that iridium was detected at only one of the national stations, indicated that the expected source was local and also that the release had been restricted in time.

To locate the source of the released activity, wind trajectories for the relevant time interval were calculated using the HYSPLIT program from NOAA ARL available at their website (http://www.arl.noaa.gov/ready/). The calculations showed that the winds came from south – southwest winds during the time-period, c.f. figure 1. The results of the filter measurements and the calculated wind trajectories were summarized and sent by e-mail to SSI.

¹ The measured part of the filter corresponds to 75% of the total filter area, the remaining 25 % of each filter is stored in an archive.

NOAA HYSPLIT MODEL Backward trajectories ending at 20 UTC 20 Sep 04 FNL Meteorological Data

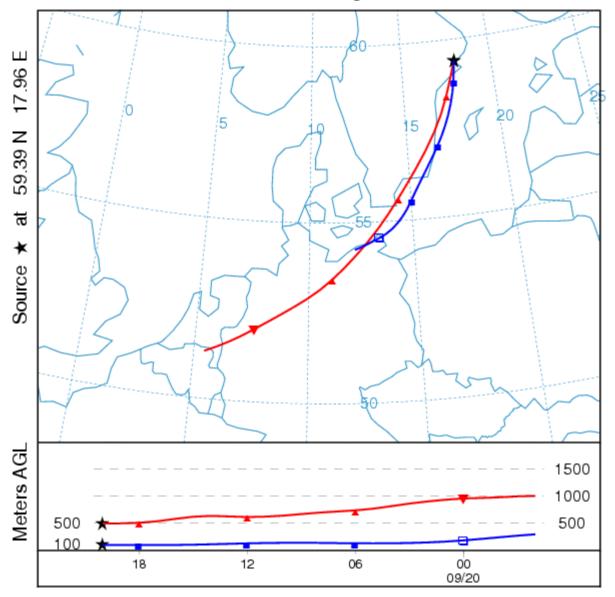


Figure 1. Calculations of 24 hours backward trajectories for the 20:th September, ending in Ursvik at 20:00 UTC. Trajectories are calculated for two different heights, 100(blue) and 500(red) meters. The calculations indicate that the activity was released south or south-west of Ursvik.

Source of the release

Given the information from FOI, SSI contacted Studsvik Nuclear AB located about 80 km south - southwest of Ursvik. Studsvik Nuclear AB then reported that there in fact had been an incident involving accidental release of ¹⁹²Ir at their production site during the 20:th of September, the same day as the activity was found in the filter. The release had led to contamination of laboratories, indoor air and personnel. A preliminary report of the incident was sent to SSI. According to the report, the incident had occurred during the afternoon, and the preliminary estimation of the released activity to the environment was 58 kBq.

Preliminary calculations

With knowledge of the source location and the estimated released amount, calculations of the expected amount collected at the Ursvik sampling site could be performed. These calculations were made by the Department of Environment and Protection, FOI NBC-defence, using more sophisticated models.

The calculations were performed assuming a release during 5 hours between 14:00 UTC and 19:00 UTC and the total amount released during this time was set to 58 kBq, based on data from the report from Studsvik. The particle size was assumed to be 5 μ m. Results from this calculation are shown in figure 2.

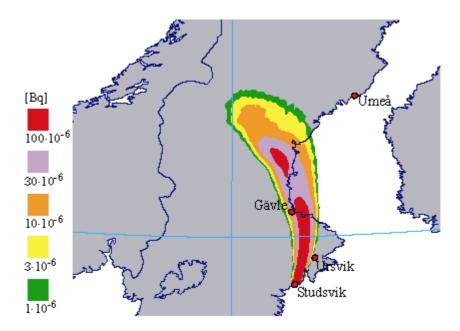


Figure 2. The figure shows the amount of activity (Bq) that would be collected on a filter assuming an air flow of 0.347 m³/s in areas where surface activity concentrations occur. The activity passes as a cloud during a few hours. In the red coloured area the total collected activity from the passing cloud would be 100 μ Bq. The locations of filter stations in the area as well as Studsvik's production site are indicated with red dots.

According to these calculations the release would not have passed Ursvik, but instead the Gävle air sampling station would have been affected by the release. In the central parts of the plume the collected activity would have been in the order of 10^4 Bq if the amount of released activity was, as stated, in the order of 60 kBq. The collected activity in Ursvik was 2.36 Bq, so the released activity has to be increased with at least a factor of 10^4 , assuming that the centre of the plume would have passed Ursvik. Within the uncertainties of the calculations it is possible that the path of the plume could have been more eastbound than shown, this would favour both the detection of iridium in Ursvik and the absence of activity in the filters from Gävle.

Local measurements

Environmental measurements were performed locally at Studsvik by Studsvik Nuclear AB, including *in-situ* measurements, grass-samples, soil-samples and also measurements on ventilation intake filters in surrounding buildings. Calculations of local dispersion were also made and the results were compared with the sample measurements. From these calculations

and measurements it was concluded that the release had indeed been in the order of 1 GBq, which is fully consistent with the calculations based on the Ursvik measurements.

Conclusion

Locally performed measurements and dispersion calculations show that the released amount of ¹⁹²Ir was in the order of 1 GBq. This is in agreement both with the measurements at the filter station in Ursvik, and with the dispersion calculations performed by FOI.

The winds varied during the release period, with the dominant wind direction being south or southeast. This is in accordance with the fact that the activity was found in Ursvik. Furthermore, the fact that no ¹⁹²Ir was measured in Gävle indicates that the wind-direction, and the extension of the plume, were more to the east than first assumed, and hence also resulting in that Ursvik would be more in the centre of the plume. The reason for this could be that local weather conditions such as, e.g. a land breeze, affect the release.

The fact that the activity was present only during one 28 hour period indicates that the release was restricted in time, which was the case since production of ¹⁹²Ir was stopped when it was discovered that personnel had been contaminated.

The incident has shown that the sensitivity of the air sampling system in Sweden system is very high and that the possibility to have a better time resolution than the regularly used weekly resolution is useful for detecting and localizing a release of radioactivity with good precision.

Sampling and analysis procedures

Sampling of ground level air is performed at six different locations in Sweden, as follows:

Kiruna:	67.84° N	20.42° E
Umeå:	63.85° N	20.34° E
Gävle:	60.40° N	17.14° E
Ursvik:	59.39° N	17.96° E
Visby:	57.63° N	18.32° E
Ljungbyhed:	56.08° N	13.23° E



At all stations except at Ursvik, 1000 m³/h of air is filtered through a glass fibre filter (Camfil type CS 5.0). At each station the filters are changed twice weekly (Monday and Thursday or Friday) and sent by mail to our laboratory at Ursvik for measurement and analysis. At Ursvik 1800 m³/h of air is filtered through 2 filters, the filters are changed with a time period of 28 hours.

Weekly samples are made from each station by taking 3/4 of each filter (1/4 of the filter is left for the archive) and compress them together into a small disc (diameter 60 mm, thickness 13 mm). These samples are measured, 3-4 days after the collection, on well shielded High Purity Germanium (HPGe) detectors. From the station at Ursvik, the 12 filters produced per week are assembled in a Marinelli like geometry by pressing them into one circular disc (diameter 94 mm, thickness 16 mm), placed on top of the detector, and into six rectangular bricks (77 mm by 48 mm by 13 mm) placed around the detector.

At four of the stations (Kiruna, Umeå, Ursvik and Ljungbyhed) a small part of the air flow (12m³/h) that has passed the filter is taken through a charcoal cartridge in order to collect gaseous iodine. The cartridges are changed weekly but only analysed if particulate iodine in greater amount has been detected in the filter.

The stations at Kiruna, Gävle, Ursvik and Ljungbyhed are each equipped with a big stainless steel funnel (1m radius) to collect precipitation. Which is passed through a cartridge consisting of a filter part, an anion part and a cation part. The cartridges are changed weekly and sent by mail to our laboratory. Four samples are combined to a monthly sample by ashing. The samples are measured on HPGe detectors. From these measurements the total deposition is calculated.

Radionuclides seen in the filters are normally only the naturally occurring radon daughters and ⁷Be. Most of our stations also detect ¹³⁷Cs, which is due to resuspension of the Chernobyl fallout. In tables I and II the concentrations of ⁷Be and ¹³⁷Cs are presented. The depositions at the stations where we collect precipitation are presented in table III. Sometimes we also detect other anthropogenic radionuclides and in that case these are presented in Table IV.

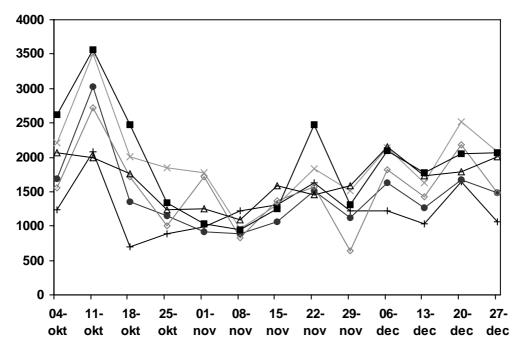
Table I

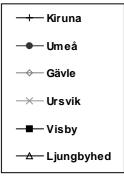
⁷Be concentrations in Sweden, fourth quarter 2004

Week starting	Kirı	una	Umo	eå	Gäı	vle	Urs	svik	Vis	s b y	Ljung	byhed
4-oct	1230 ⁽¹⁾	(0.2)	1690	(0.2)	1560	(0.3)	2210	(0.1)	2610	(0.2)	2060	(0.2)
11-oct	2080 ⁽²⁾		3030	` ,	2720	` ,		(0.1)	3560	` ,		(0.2)
18-oct	700 ⁽³⁾	(0.2)	1360	(0.2)	1720	(0.3)	2000	(0.1)	2480	(0.2)	1760	(0.2)
25-oct	890 ⁽⁴⁾	(0.2)	1160	(0.3)	1010	(0.5)	1850	(0.1)	1340	(0.3)	1230	(0.2)
1-nov	990	(0.3)	910	(0.2)	1720	(0.2)	1780	(0.2)	1030	(0.4)	1260	(0.2)
8-nov	1220 ⁽⁵⁾	(0.2)	890	(0.2)	830	(0.3)	960	(0.2)	950	(0.4)	1100	(0.2)
15-nov	1310 ⁽⁶⁾	(0.2)	1070 ⁽¹⁰⁾	(0.2)	1360	(0.2)	1310	(0.2)	1250	(0.3)	1580	(0.2)
22-nov	1630 ⁽⁷⁾	(0.2)	1520 ⁽⁷⁾	(0.3)	1570 ⁽¹¹⁾	(0.5)	1830	(0.1)	2480	(0.2)	1460	(0.2)
29-nov	1220	(0.2)	1120	(0.2)	650	(0.5)	1510	(0.2)	1300(12)	(0.4)	1590	(0.2)
6-dec	1220	(0.2)	1630	(0.2)	1820	(0.2)	2140	(0.1)	2090	(0.2)	2160	(0.1)
13-dec	1030 ⁽⁸⁾	(0.2)	1270	(0.2)	1430	(0.2)	1630	(0.1)	1770	(0.3)	1740	(0.2)
20-dec	1640 ⁽⁹⁾	(0.2)	1670	(0.2)	2180	(0.2)	2510	(0.1)	2050	(0.2)	1780	(0.2)
27-dec	1060	(0.2)	1490	(0.2)	1480	(0.3)	2080	(0.2)	2070	(0.3)	2000	(0.2)

Values are given in µBq/m³.

Error estimates (1σ %) are given in brackets.





 $^{^{1)}}_{...}$ Six days filter, 4 - 10/10

²⁾ Eight days filter, 10 - 18/10

³⁾ Eight days filter, 18 - 26/10

⁴⁾ Six days filter, 26/10 - 1/11

⁵⁾ Eight days filter, 8 - 16/11

⁶⁾ Seven days filter, 16 -23/11

^{&#}x27;) Six days filter, 23 - 29/11

s) Six days filter, 23 - 29/11 s) Six days filter, 13 - 19/12 ⁹⁾ Eight days filter, 19 - 27/12 ¹⁰⁾ Eight days filter, 15 - 23/11

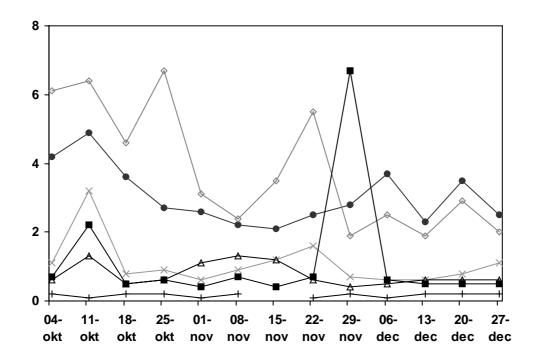
¹¹⁾ Three days filter, 26 - 29/11

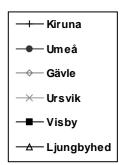
¹²⁾ Five days filter, 1 - 6/12

Table II ¹³⁷Cs concentrations in Sweden, fourth quarter 2004

Week starting	Kir	una	Un	ıeå	Gä	vle	Urs	vik	Vis	sby	Ljung	byhed
4-oct	0.2 ⁽¹⁾	(23)	4.2	(2)	6.1	(3)	1.1	(6)	0.7	(17)	0.6	(12)
11-oct	0.1(2)	(51)	4.9	(2)	6.4	(4)	3.2	(2)	2.2	(7)	1.3	(5)
18-oct	0.2(3)	(26)	3.8	(3)	4.6	(2)	0.8	(7)	0.5	(25)	0.5	(9)
25-oct	0.2(4)	(29)	2.7	(3)	6.7	(3)	0.9	(6)	0.6	(18)	0.6	(9)
1-nov	0.1	(47)	2.6	(3)	3.1	(3)	0.6	(12)	0.4	(27)	1.1	(6)
8-nov	0.2(5)	(26)	2.1	(3)	2.4	(3)	0.8	(7)	0.7	(24)	1.1	(6)
15-nov	<0.1 ⁽⁶⁾		2.5 ⁽¹⁰⁾	(3)	3.5	(2)	1.2	(4)	0.4	(23)	1.2	(4)
22-nov	<0.1 ⁽⁷⁾		2.6 ⁽⁷⁾	(3)	5.5 ⁽¹¹⁾	(7)	1.6	(3)	0.7	(16)	0.6	(8)
29-nov	0.2	(21)	2.8	(2)	1.9	(8)	0.7	(6)	6.7 ⁽¹²⁾	(4)	0.4	(14)
6-dec	0.1	(50)	3.7	(2)	2.5	(4)	0.6	(8)	0.6	(18)	0.5	(11)
13-dec	0.2 ⁽⁸⁾	(31)	2.3	(3)	1.9	(4)	0.6	(8)	0.5	(22)	0.6	(9)
20-dec	0.2(9)	(17)	3.5	(2)	2.9	(3)	0.8	(6)	0.5	(21)	0.6	(10)
27-dec	0.2	(25)	2.5	(3)	2.0	(5)	1.1	(6)	0.5	(27)	0.6	(9)

Values are given in $\mu Bq/m^3.$ Error estimates (10 %) are given in brackets.





 $^{^{1)}}$ Six days filter, 4-10/10

²⁾ Eight days filter, 10 - 18/10

³⁾ Eight days filter, 18 - 26/10

⁴⁾ Six days filter, 26/10 - 1/11

⁵⁾ Eight days filter, 8 - 16/11

⁶⁾ Seven days filter, 16 -23/11

⁷⁾ Six days filter, 23 - 29/11

⁸⁾ Six days filter, 13 – 19/12 ⁹⁾ Eight days filter, 19 - 27/12 ¹⁰⁾ Eight days filter, 15 - 23/11

¹¹⁾ Three days filter, 26 - 29/11

¹²⁾ Five days filter, 1 - 6/12

Table III

Deposition measurements, fourth quarter 2004

Kiruna

Weeks	Period	⁷ Be	¹³⁷ Cs	Precipitation (mm)
40 – 43	27/9 – 26/10	12200 (1.0)	19 (35)	24.5
44 – 47	26/10-23/11	11000 (0.7)	<5	7.0
48 – 51	23/11 – 19/12	4600 (1.3)	<7	4.8

Gävle

Weeks	Period	⁷ Be	¹³⁷ Cs	Precipitation (mm)
39 – 42*	20/9 – 18/10	36600 (0.4)	97 (8)	38.9
43 – 46**	18/10 – 15/11	40900 (0.4)	96 (7)	43.0
47 – 50	15/11 – 13/12	21500 (0.6)	67 (10)	25.7

^{*} Also detected ¹³¹I concentration: 180 (14) mBq/m²

Ursvik

Weeks	Period	^{7}Be	¹³⁷ Cs	Precipitation (mm)
38 - 41	13/9 – 11/10	50600 (0.4)	20 (30)	50.9
42 - 45	11/10 -8/11	52100 (0.4)	45 (18)	38.5
46 - 49	8/11 – 6/12	43300 (0.4)	16 (35)	63.3
50 - 53	6/12 - 27/12	32000 (0.5)	19 (34)	36.3

Ljungbyhed

_	Weeks	Period	⁷ Be	¹³⁷ Cs	Precipitation (mm)
	41 – 44	4/10 – 1/11	74400 (0.3)	30 (20)	50.6
	45 – 48	1/11 – 29/11	91300 (0.3)	22 (25)	54.1
	49 – 52	29/11-27/12	83400 (0.3)	26 (23)	49.1

Values are given in mBq/m².

Error estimates (1 σ %) are given in brackets.

^{**} Also detected ¹³¹I concentration: 70 (29) mBq/m²

Table IV

Other anthropogenic radionuclides detected, fourth quarter 2004

Week starting	Station	Isotope	Concentration	Note
4-oct	Gävle	¹³¹	3.3 (11)	(1)
10-oct	Gävle	¹³¹	1.3 (31)	(1)
8-nov	Gävle	¹³¹	4.6 (3)	(1)
15-nov	Gävle	¹³¹	3.0 (6)	(1)
13-dec	Gävle	¹³¹	1.0 (20)	(1)

Values are given in µBq/m³.

Error estimates (1σ %) are given in brackets.

⁽¹⁾ The activities of ¹³¹I found in Gävle have been shown to correspond to administration of cancer treatment doses for thyroidea cancer at the Gävle-Sandviken County Hospital (ref. Erlandsson et al., "I-131 in air filters at Gävle", presented at NSRP 13th meeting in Åbo, 25-29 August 2002).