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**Monitoring of radiation in airdust, deposition  
and an overall country milk sample.  
Results in the Netherlands in 1996**

Laboratory of Radiation Research  
R.M.W. Overwater (final editor)

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## **PREFACE**

This 1996 annual report presents the results of radiation measurements in airdust, deposition and milk samples in the Netherlands by the Laboratory of Radiation Research (LSO) of the National Institute of Public Health and the Environment (RIVM).

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**ABSTRACT**

This 1996 annual report presents the results of radiation measurements of the atmosphere in the Netherlands by the Laboratory of Radiation Research (LSO) of the National Institute of Public Health and the Environment (RIVM). The programme of RIVM/LSO includes samples of airdust and deposition taken at the RIVM premises in Bilthoven. An overall country milk sample, a mixture of milk samples from four milk factories in the Netherlands, was also analysed.

In 1996, the yearly average gross  $\alpha$ - and gross  $\beta$ -activity concentrations in airdust sampled with a high volume sampler were  $0.0671 \pm 0.0009$  (SD 0.03) and  $0.540 \pm 0.003$  (SD 0.33)  $\text{mBq}\cdot\text{m}^{-3}$ , where SD illustrates the variation in weekly averages during the year. These values are of the same order as those of the period 1992-1995. The yearly average activity concentrations in airdust, measured using  $\gamma$ -spectroscopy, for the nuclides  $^7\text{Be}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  were  $3890 \pm 50$  (SD 1320),  $1.60 \pm 0.03$  (SD 1.2) and  $500 \pm 10$  (SD 390)  $\mu\text{Bq}\cdot\text{m}^{-3}$ .

The yearly total gross  $\alpha$ - and gross  $\beta$ -activities of the deposition in Bilthoven were  $16.4 \pm 1.5$  and  $67 \pm 5$   $\text{Bq}\cdot\text{m}^{-2}$ , and the yearly total deposition of  $^3\text{H}$  was  $970 \pm 50$   $\text{Bq}\cdot\text{m}^{-2}$ . The total activities of  $^{137}\text{Cs}$ ,  $^7\text{Be}$  and  $^{210}\text{Pb}$  in deposition were  $0.55 \pm 0.03$ ,  $920 \pm 20$  and  $64.9 \pm 1.6$   $\text{Bq}\cdot\text{m}^{-2}$ , respectively. The  $^{210}\text{Pb}$ - and  $^{210}\text{Po}$ -analysis results obtained using  $\alpha$ -spectroscopy were  $57 \pm 3$  and  $9 \pm 2$   $\text{Bq}\cdot\text{m}^{-2}$ . These activities are all of the same order as those in previous years.

The yearly average concentration of  $^{137}\text{Cs}$  in consumer milk from a nationwide sample mix was  $0.073 \pm 0.010$   $\text{Bq}\cdot\text{L}^{-1}$ , which is almost equal to that of the previous year. The results for  $^{90}\text{Sr}$  are lower than the detection limit of  $0.2$   $\text{Bq}\cdot\text{L}^{-1}$ .

## SAMENVATTING

In dit jaarverslag over 1996 zijn de resultaten weergegeven van de metingen van radioactiviteit in milieumonsters uit de atmosfeer uitgevoerd door het Laboratorium voor Stralingsonderzoek (LSO) van het Rijksinstituut voor Volksgezondheid en Milieu (RIVM). De analyses zijn uitgevoerd aan monsters luchtstof en depositie genomen op het RIVM-terrein in Bilthoven. Ook zijn mengmonsters melk, gemengd uit melkmonsters afkomstig van vier melkfabrieken verspreid over het land, geanalyseerd.

In 1996 zijn de jaargemiddelde totale  $\alpha$ - en  $\beta$ -activiteitsconcentraties in luchtstof, bemonsterd met een high volume sampler,  $0,0671 \pm 0,0009$  (SD 0,03) en  $0,540 \pm 0,003$  (SD 0,33) mBq·m<sup>-3</sup>, waarbij SD de variatie in de wekelijkse resultaten gedurende het jaar representeert. Deze waarden zijn overeenkomstig die van de periode 1992-1995. De  $\gamma$ -spectrometrische analyses uitgevoerd aan luchtstofmonsters gaven voor de nucliden <sup>7</sup>Be, <sup>137</sup>Cs en <sup>210</sup>Pb de activiteitsconcentraties  $3890 \pm 50$  (SD 1320),  $1,60 \pm 0,03$  (SD 1,2) en  $500 \pm 10$  (SD 390)  $\mu$ Bq·m<sup>-3</sup>.

De jaarlijkse totale  $\alpha$ - en  $\beta$ -activiteiten in depositie in Bilthoven over 1996 bedroegen  $16,4 \pm 1,5$  en  $67 \pm 5$  Bq·m<sup>-2</sup>. De jaarlijkse totale activiteit van <sup>3</sup>H in depositie was  $970 \pm 50$  Bq·m<sup>-2</sup>. De totale jaarlijkse deposities van <sup>137</sup>Cs, <sup>7</sup>Be en <sup>210</sup>Pb, verkregen via  $\gamma$ -spectrometrie, bedroegen  $0,55 \pm 0,03$ ,  $920 \pm 20$  en  $64,9 \pm 1,6$  Bq·m<sup>-2</sup>. De <sup>210</sup>Pb- en <sup>210</sup>Po-resultaten bepaald met  $\alpha$ -spectroscopie waren  $57 \pm 3$  en  $9 \pm 2$  Bq·m<sup>-2</sup>. Al de gemeten waarden zijn van dezelfde orde van grootte als de waarden die in de voorafgaande jaren zijn gemeten.

De jaargemiddelde activiteitsconcentratie van <sup>137</sup>Cs in melk van een landelijk mengmonster is  $0,073 \pm 0,010$  Bq·L<sup>-1</sup>, hetgeen nagenoeg gelijk is aan het resultaat van vorig jaar. Voor <sup>90</sup>Sr is een waarde gemeten beneden de detectielimiet van  $0,2$  Bq·L<sup>-1</sup>.

## 1. INTRODUCTION

Radioactive nuclides of natural origin, such as  $^{40}\text{K}$  and daughters from the uranium and thorium series, are found in the environment. Due to human activities, such as factories processing ores, natural radionuclides are emitted into the environment and consequently may lead to enhanced levels. The presence of man-made radionuclides occur in the environment through, for instance, fallout from experiments with nuclear weapons or discharges from nuclear installations. Under the authority of the State Health Inspectorate, the National Institute of Public Health and the Environment (RIVM) regularly monitors the concentration of a number of radionuclides in airdust and deposition. Milk from four major dairies is sampled by the Veterinary Public Health Inspectorate (VHI) and analysed by RIVM. The results of these measurements are presented in this report.

The monitoring programme is given in Chapter 2. In Chapter 3 results are given for airdust, deposition and for overall country milk. Conclusions are given in Chapter 4. In correspondence with previous reports (e.g. [1]), this report gives a data-presentation only. The results will, in general, be presented visually with a minimum amount of text. More detailed tables will be shown in the Appendix. A detailed description of sampling, sample treatment and analytical method is given in previous reports, e.g. the Monitoring Report of 1995 [1].

## 2. MONITORING PROGRAMME

The monitoring programme in 1996 for the determination of radioactive nuclides in the atmosphere is given in *Table 2.1*. Two types of samples are analysed: airdust and deposition. The sampling was done on the RIVM premises in Bilthoven. Airdust samples were collected weekly ( $\gamma$ -emitters, gross- $\alpha$ , gross- $\beta$ ) with a High Volume Sampler (HVS). Samples of both wet and dry deposition are collected weekly ( $\gamma$ -emitters) and monthly ( $^3\text{H}$ , gross- $\alpha$ , gross- $\beta$ ,  $^{210}\text{Pb}/^{210}\text{Po}$ ). For the  $^{210}\text{Pb}/^{210}\text{Po}$  analysis, three monthly samples are combined into one sample which is analysed quarterly.

The monitoring programme in 1996 for the determination of radioactive nuclides in milk is given in *Table 2.1*. Five-litre samples were taken monthly from four milk factories which are situated as follows: Frico at Groningen (north), Coberco at Deventer (east), Campina at Bergeijk (south), and De Graafstroom at Bleskesgraaf (west). A mixture of these samples is taken to be representative for milk in the Netherlands. Three monthly samples are mixed to a quarterly sample which is analysed on  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$  and  $\gamma$ -emitters. The analysis of  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  in milk is performed using Sr•SPEC™ separation and liquid scintillation counting, a method differing from the method used until 1995 (for further details see [2,3]).

*Table 2.1: Monitoring programme in 1996 for the determination of radioactive nuclides in airdust, deposition and milk.*

Measurement	Location	Sample period	Sample volume	Analysis frequency	Analysis
Airdust	Bilthoven	week	$\pm 50000 \text{ m}^3$	1 x/week	$\gamma$ -em
	Bilthoven	week	$\pm 1000 \text{ m}^3$ *	1 x/week	gross $\alpha$ , gross $\beta$
Deposition	Bilthoven	week	variable	1 x/week	$\gamma$ -em
	Bilthoven	month	variable	1 x/month	gross $\alpha$ , gross $\beta$ , $^3\text{H}$
	Bilthoven	month	variable	1 x/quarter	$^{210}\text{Pb}/^{210}\text{Po}$
Milk	National sample	month	mix	1 x/quarter	Sr, $\gamma$ -em

$\gamma$ -em:  $\gamma$ -spectroscopic analysis in which the contents of specific  $\gamma$ -emitting nuclides are determined

Sr: determination of both  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$

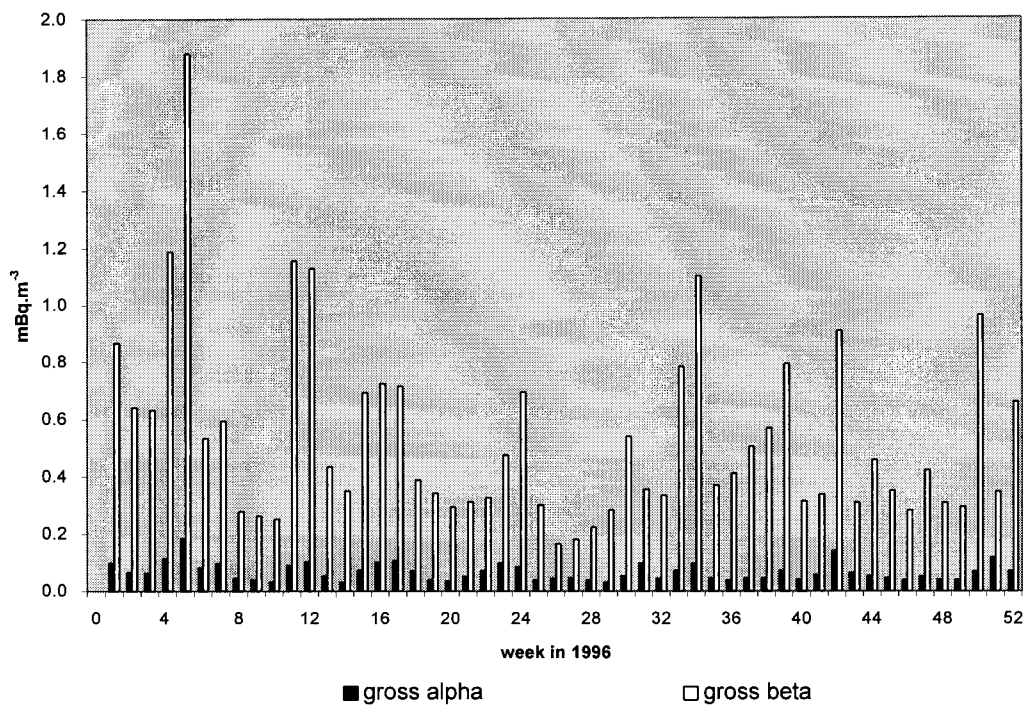
\*: a subsample of 2% is taken from the filter through which  $50000 \text{ m}^3$  is sampled



### 3. RESULTS

#### 3.1 AIRDUST

The weekly results of gross  $\alpha$ - and  $\beta$ -activity concentrations in airdust are given in *Figure 3.1* and *Table A1* (see Appendix). The period between sampling and analysis is five to ten days, which is long compared to the decay time of the short-lived decay products of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$ . Therefore, these naturally occurring decay-products do not contribute to the  $\alpha$ - and  $\beta$ -activity concentrations. Sampling by HVS has started in 1992. The yearly averages of the long-lived gross  $\alpha$ - and  $\beta$ -activity concentrations in 1996 are of the same order as the results in the period 1992-1995 [1,4-6] (see *Figure 3.2*).



*Figure 3.1: Weekly results in 1996 of long-lived gross  $\alpha$ - and gross  $\beta$ -activity concentrations in airdust sampled at the RIVM premises in Bilthoven (The Netherlands).*

The frequency distributions of gross  $\alpha$ -activity and gross  $\beta$ -activity concentrations in airdust are given in *Figures 3.3* and *3.4*, respectively.

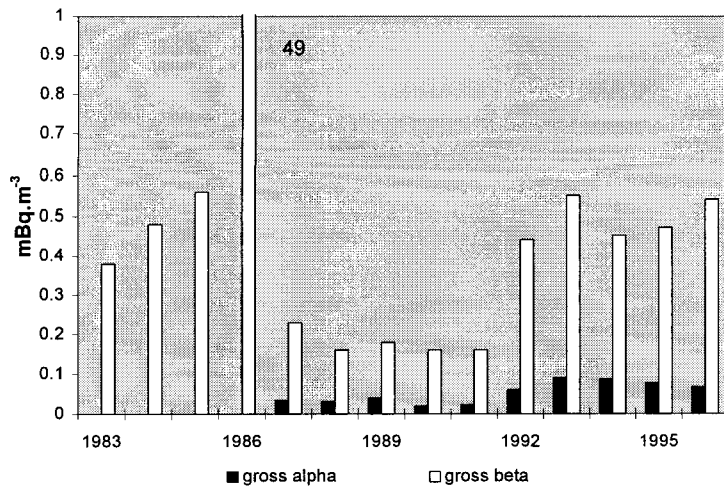


Figure 3.2: Yearly averages of long-lived gross  $\alpha$ - and gross  $\beta$ -activity concentration in air dust for the period 1983-1996 [1,4-12]. For the long-lived gross  $\alpha$ -activity data are only available since 1987. The 49 mBq·m<sup>-3</sup> level in 1986 was caused by the accident at the Chernobyl nuclear power plant.

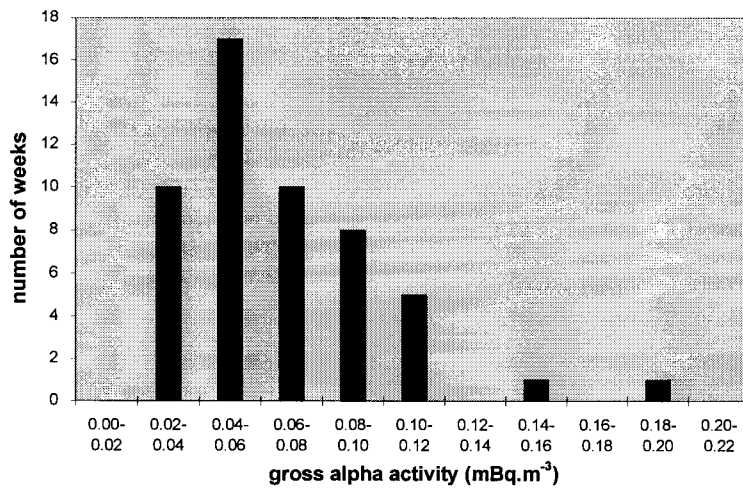


Figure 3.3: Frequency distribution of long-lived gross  $\alpha$ -activity concentration in air dust collected weekly at RIVM, Bilthoven (The Netherlands) in 1996. Mean value is  $0.0671 \pm 0.0009$  (SD 0.03) mBq·m<sup>-3</sup>.

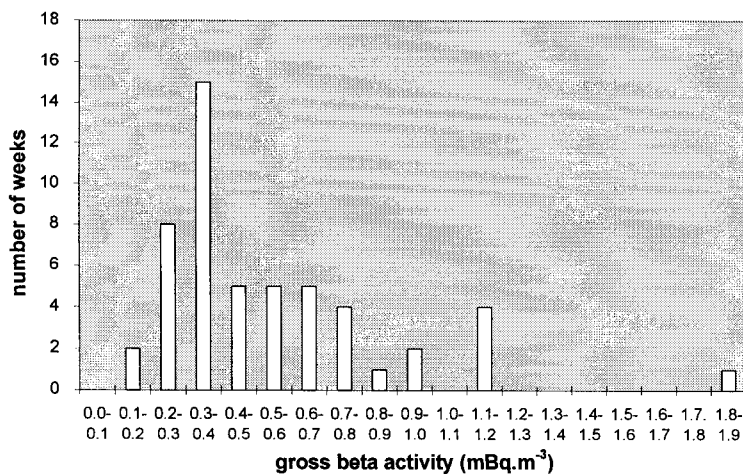
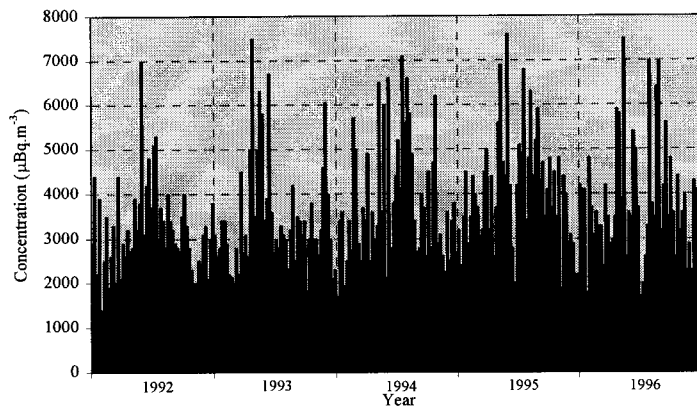
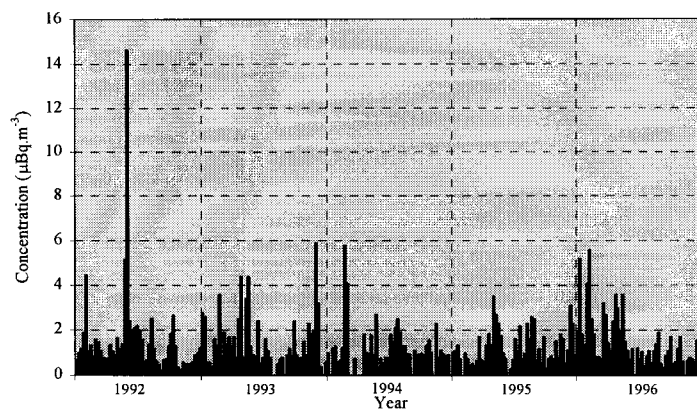


Figure 3.4: Frequency distribution of long-lived gross  $\beta$ -activity concentration in air dust collected weekly at RIVM, Bilthoven (The Netherlands) in 1996. Mean value is  $0.540 \pm 0.003$  (SD 0.33) mBq·m<sup>-3</sup>.

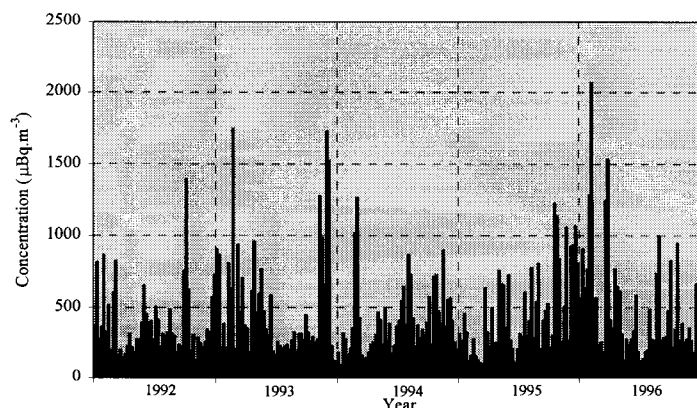
The nuclides  $^7\text{Be}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  are the only nuclides found with values above the detection limit (*Table A2; Figure 3.5, 3.6 and 3.7*). The detection limits for the nuclides considered in the gamma-spectroscopic analysis of the HVS-samples are given in *Table A3*. The values found in 1996 were not significantly different from the values found in the period 1992-1995 [1,4-6].



*Figure 3.5: Activity concentration of  $^7\text{Be}$  in airdust in Bilthoven in the period 1992-1996. Yearly average in 1996 is  $3890 \pm 50$  (SD 1320)  $\mu\text{Bq}\cdot\text{m}^{-3}$ .*



*Figure 3.6: Activity concentration of  $^{137}\text{Cs}$  in airdust in Bilthoven in the period 1992-1996. Yearly average in 1996 is  $1.60 \pm 0.03$  (SD 1.2)  $\mu\text{Bq}\cdot\text{m}^{-3}$ .*



*Figure 3.7: Activity concentration of  $^{210}\text{Pb}$  in airdust in Bilthoven in the period 1992-1996. Yearly average in 1996 is  $500 \pm 10$  (SD 390)  $\mu\text{Bq}\cdot\text{m}^{-3}$ .*

### 3.2 DEPOSITION

The monthly values of deposited long-lived gross  $\alpha$ - and gross  $\beta$ -activity are given in *Figure 3.8* and in *Table A4*. The yearly total depositions of gross- $\alpha$  and gross- $\beta$  were  $16.4 \pm 1.5$  and  $67 \pm 5$   $\text{Bq}\cdot\text{m}^{-2}$ , respectively. These values are not significantly different from those since 1987, as is illustrated in *Figure 3.9* and *Table 3.1*.

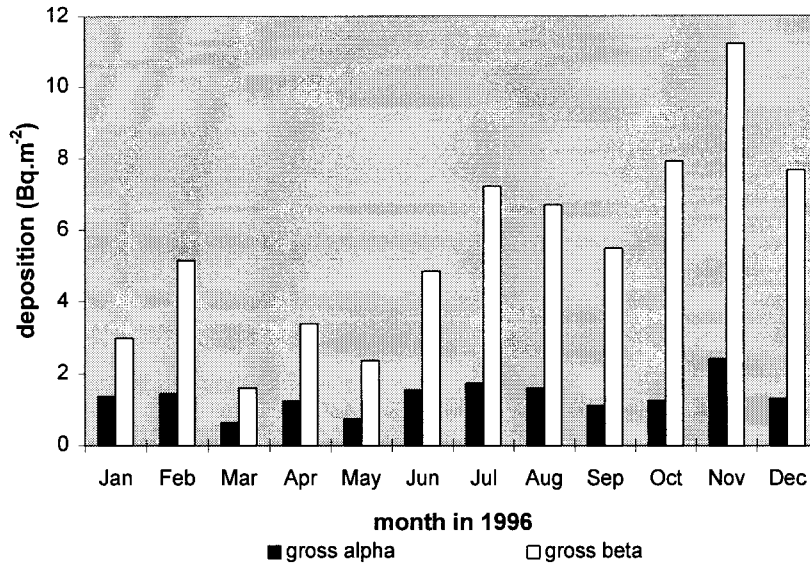


Figure 3.8: Monthly data of long-lived gross  $\alpha$ - and gross  $\beta$ -activity in deposition in 1996.

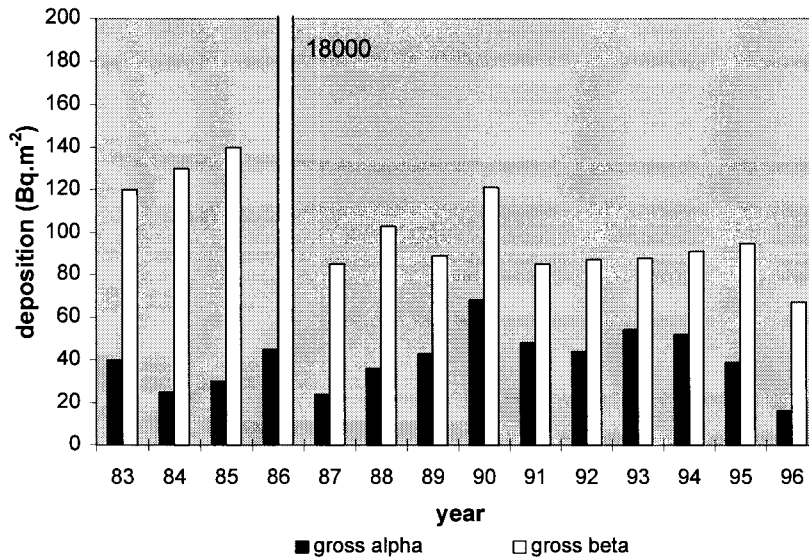


Figure 3.9: Total yearly deposition of long-lived gross  $\alpha$ - and gross  $\beta$ -activity from 1983 to 1996. See also *Table 3.1*. The  $18000 \text{ Bq}\cdot\text{m}^{-2}$  level in 1986 was caused by the accident at the Chernobyl nuclear power plant.

Table 3.1: Yearly totals for long-lived gross  $\alpha$ - and gross  $\beta$ -activity,  $^3\text{H}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in deposition from 1983 to 1996 (see also [13-16]).

Year	Precipitation mm	Gross $\alpha$ $\text{Bq}\cdot\text{m}^{-2}$	Gross $\beta$ $\text{Bq}\cdot\text{m}^{-2}$	$^3\text{H}$ $\text{Bq}\cdot\text{m}^{-2}$	$^{210}\text{Pb}^{\#}$ $\text{Bq}\cdot\text{m}^{-2}$	$^{210}\text{Po}^{\#}$ $\text{Bq}\cdot\text{m}^{-2}$
1983	869	40 (*)	120 (*)	2100	-	-
1984	868	25 (*)	130 (*)	2610	-	-
1985	767	30 (*)	140 (*)	3800	-	-
1986	825	45 (*)	18000 (*)	2400	15	3
1987	975	24 $\pm$ 1	85 $\pm$ 3	2630	52	6
1988	887	36 $\pm$ 2	103 $\pm$ 3	1700 $\pm$ 40	110 $\pm$ 3	25 $\pm$ 1
1989	706	43 $\pm$ 1	89 $\pm$ 3	1560 $\pm$ 130	94 $\pm$ 7	24 $\pm$ 4
1990	756	68 $\pm$ 1	121 $\pm$ 4	1360 $\pm$ 120	85 $\pm$ 4	16 $\pm$ 2
1991	699	48 $\pm$ 1	85 $\pm$ 1	1060 $\pm$ 50	56 $\pm$ 1	10 $\pm$ 1
1992	946	44 $\pm$ 1	87 $\pm$ 1	1440 $\pm$ 50	83 $\pm$ 5	11 $\pm$ 1
1993	886	54.3 $\pm$ 0.7	87.9 $\pm$ 0.8	1310 $\pm$ 30	78 $\pm$ 3	6.0 $\pm$ 0.6
1994	1039	52.0 $\pm$ 0.7	91.2 $\pm$ 1.0	1210 $\pm$ 30	82 $\pm$ 3	12.7 $\pm$ 0.7
1995	724	39 $\pm$ 4	95 $\pm$ 8	970 $\pm$ 40	(-) <sup>1</sup>	(-) <sup>1</sup>
1996	626	16.4 $\pm$ 1.5	67 $\pm$ 5	970 $\pm$ 50	57 $\pm$ 3	9 $\pm$ 2

(\*) Different method starting from 1987.

- No analysis.

# Data from  $\alpha$ -spectroscopy.

(-)<sup>1</sup> Result rejected [18]

The monthly values of  $^3\text{H}$  and the weekly values of  $^7\text{Be}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  are given in Tables A4 and A6. The yearly total deposition of  $^3\text{H}$  is  $970 \pm 50 \text{ Bq}\cdot\text{m}^{-2}$ , which is equal to the value of 1995 – the lowest value since 1970 [17] (see Figure 3.10).

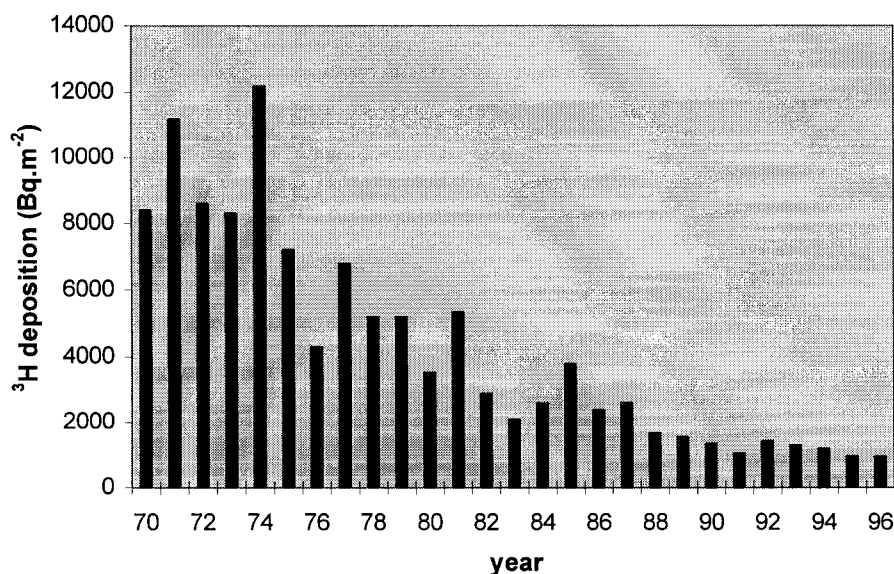


Figure 3.10: Total deposition of  $^3\text{H}$  in the period 1970-1996.

Significant activities of the naturally occurring nuclides  $^7\text{Be}$  and  $^{210}\text{Pb}$  were found, with yearly total depositions of  $920 \pm 20$  and  $64.9 \pm 1.6 \text{ Bq}\cdot\text{m}^{-2}$ . For  $^{137}\text{Cs}$  weekly depositions with a maximum of  $0.057 \pm 0.008 \text{ Bq}\cdot\text{m}^{-2}$  (detection limit is about  $0.005 \text{ Bq}\cdot\text{m}^{-2}$ ) and a yearly total deposition of  $0.55 \pm 0.03 \text{ Bq}\cdot\text{m}^{-2}$  were found. For  $^{134}\text{Cs}$  no activity was found higher than the detection limit of about  $0.2 \text{ Bq}\cdot\text{m}^{-2}$ . In week 40 the nuclide  $^{54}\text{Mn}$  was found with an activity of  $0.011 \pm 0.003 \text{ Bq}\cdot\text{m}^{-2}$ .

Figure 3.11 presents the relationship between the monthly deposition of  $^3\text{H}$  and the amount of precipitation. In Figure 3.12 the same relationship is presented for the weekly deposition of  $^7\text{Be}$  and in Figure 3.13 of  $^{210}\text{Pb}$ . Figure 3.11 indicates a correlation between the amount of rainfall and the deposition of  $^3\text{H}$ . For  $^7\text{Be}$  and  $^{210}\text{Pb}$ , as indicated in Figures 3.12 and 3.13, such a correlation is less clear.

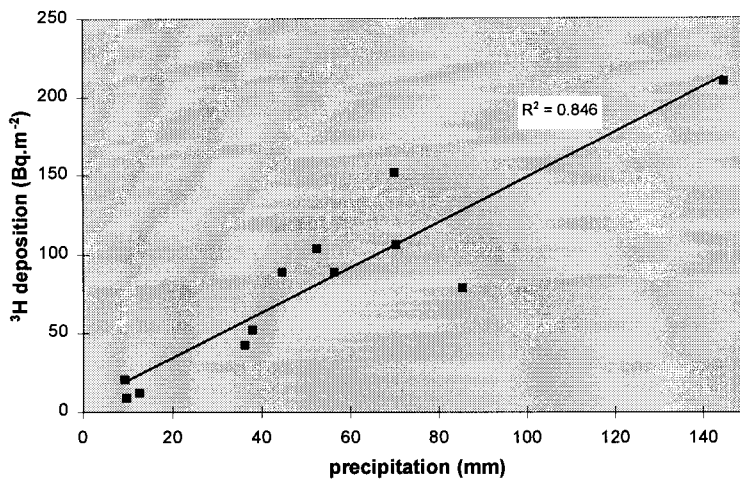


Figure 3.11: The monthly deposition of  $^3\text{H}$  in 1996 versus precipitation.

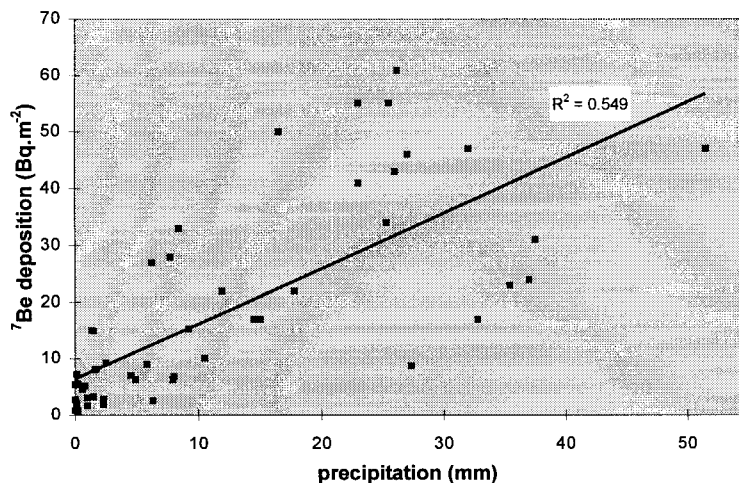


Figure 3.12: The weekly deposition of  $^7\text{Be}$  in 1996 versus precipitation.

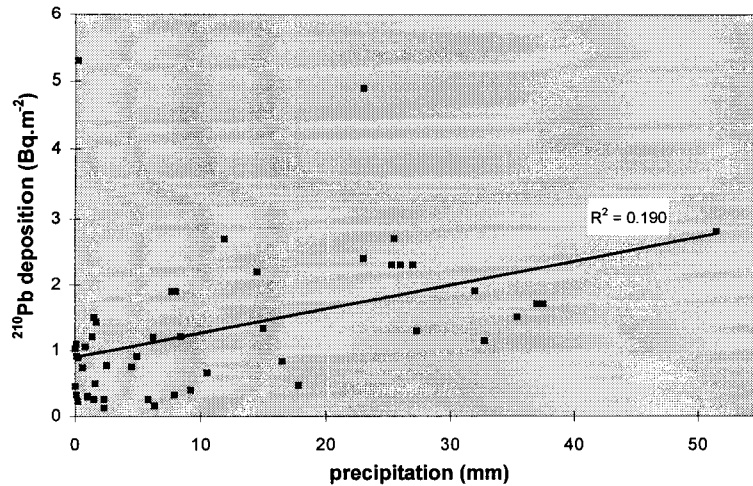
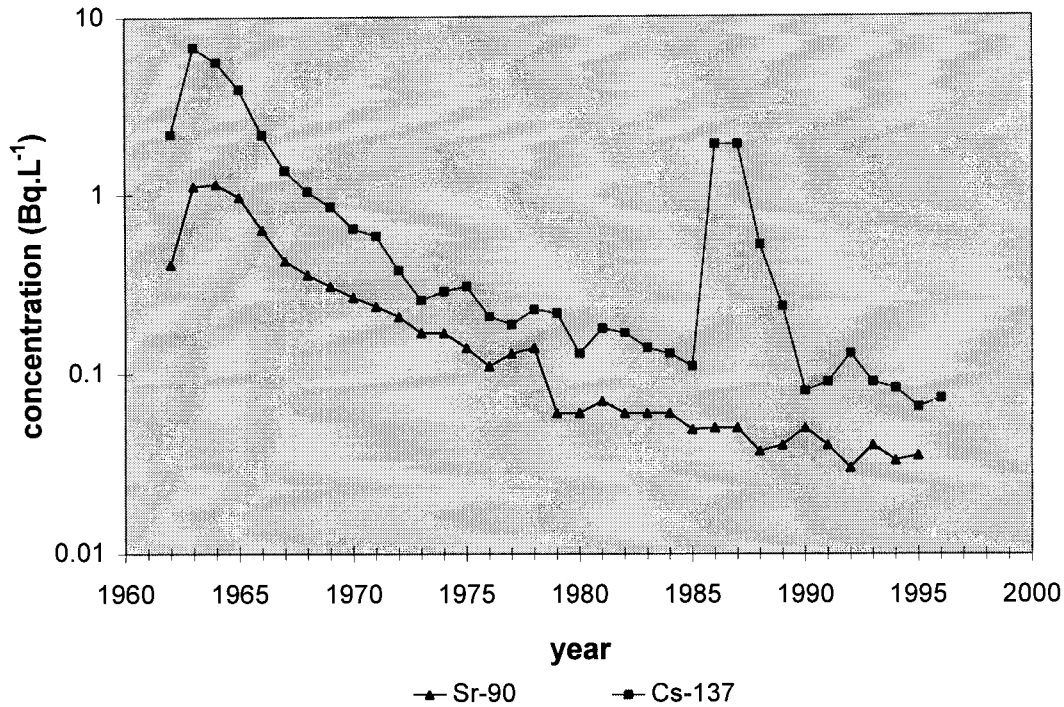


Figure 3.13: The weekly deposition of  $^{210}\text{Pb}$  in 1996 versus precipitation.

The quarterly results for the  $^{210}\text{Pb}/^{210}\text{Po}$ -analysis by  $\alpha$ -spectroscopy are given in *Table A5*. In 1995 a contamination of the samples occurred and no results for  $^{210}\text{Pb}$  en  $^{210}\text{Po}$  were reported [18]. The results obtained in 1996 for  $^{210}\text{Pb}$  for  $\alpha$ -spectroscopy are in good agreement with the result for  $\gamma$ -spectroscopy (see *Tables A5* and *A6*). The gross- $\alpha$  result is mainly due to  $^{210}\text{Po}$ , while the gross- $\beta$  result is mainly due to  $^{210}\text{Pb}$ . This is depicted in the similarity of the results of gross- $\alpha$  and  $^{210}\text{Po}$  as well as the similarity of the results of gross- $\beta$  and  $^{210}\text{Pb}$ .

### 3.3 OVERALL COUNTRY MILK SAMPLE

In 1996, the average result for  $^{137}\text{Cs}$  in the mixed milk sample is  $0.073 \pm 0.010 \text{ Bq}\cdot\text{L}^{-1}$ , which is down to the level just before the Chernobyl accident. This is illustrated in *Figure 3.14* (see also *Table A7* and refs. [19-21]). For  $^{40}\text{K}$  the average yearly result ( $49.0 \pm 1.5 \text{ Bq}\cdot\text{L}^{-1}$ ) is not significantly different from the results of previous years. The activity concentrations of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ ,  $^{134}\text{Cs}$  and  $^7\text{Be}$  are below the detection limit.



*Figure 3.14: The yearly average concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in milk in the Netherlands since 1962. For  $^{90}\text{Sr}$  a detection limit of  $0.2 \text{ Bq}\cdot\text{L}^{-1}$  was measured in 1996. Note the logarithmic scale.*



#### 4. CONCLUSIONS

The yearly averages in airdust and the yearly totals in deposition are summarized in *Table 4.1*. More detailed data are given in *Tables A1* and *A2* for airdust, and *Table A4, A5* and *A6* for deposition.

No unexpected activity concentrations have been observed for long-lived nuclides in airdust and deposition. All the results are of the same order as the values for the period 1992-1995. The  $^3\text{H}$  values are the lowest since 1970 (*Figure 3.10*). The  $^{137}\text{Cs}$  values are back to the pre-Chernobyl values.

*Table 4.1: Results summarized for 1996: yearly average activities in airdust and in deposition.*

Type of activity	Average airdust activity concentration $\text{mBq}\cdot\text{m}^{-3}$ (SD)	Total Deposition $\text{Bq}\cdot\text{m}^{-2}$
Gross $\alpha$	$0.0671 \pm 0.0009$ (0.03)	$16.4 \pm 1.5$
Gross $\beta$	$0.540 \pm 0.003$ (0.33)	$67 \pm 5$
$^3\text{H}$	-	$970 \pm 50$
$^{137}\text{Cs}$	$0.00160 \pm 0.00003$ (0.0012)	$0.55 \pm 0.03$
$^7\text{Be}$	$3.89 \pm 0.05$ (1.32)	$920 \pm 20$
$^{210}\text{Pb}$ @	$0.500 \pm 0.010$ (0.39)	$64.9 \pm 1.6$
$^{210}\text{Pb}$ #	-	$57 \pm 3$
$^{210}\text{Po}$	-	$9 \pm 2$

- no analysis

@ data from  $\gamma$ -spectroscopy.

# data from  $\alpha$ -spectroscopy.

The concentration of  $^{137}\text{Cs}$  (yearly average of  $0.073 \pm 0.010 \text{ Bq}\cdot\text{L}^{-1}$ ) in an overall country milk sample is of the same order of magnitude as the values recorded just before Chernobyl. For  $^{40}\text{K}$  (yearly average of  $49.0 \pm 1.5 \text{ Bq}\cdot\text{L}^{-1}$ ) the results are not significantly different from the results of previous years. The activity concentrations of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ ,  $^{134}\text{Cs}$  and  $^7\text{Be}$  are below the detection limit.

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## APPENDIX

Table A1: Weekly results of gross  $\alpha$ - and gross  $\beta$ -activities in airdust sampled by HVS in 1996 on the RIVM premises in Bilthoven (The Netherlands). The error in the yearly average is equal to the square root of the sum of the squared weekly errors divided by the number of weeks. SD is the standard deviation of the weekly results.

week number	gross- $\alpha$ mBq·m <sup>-3</sup>	gross- $\beta$ mBq·m <sup>-3</sup>	week number	gross- $\alpha$ mBq·m <sup>-3</sup>	gross- $\beta$ mBq·m <sup>-3</sup>
1	0.099 ± 0.008	0.87 ± 0.02	27	0.046 ± 0.005	0.182 ± 0.009
2	0.066 ± 0.006	0.64 ± 0.02	28	0.039 ± 0.005	0.224 ± 0.010
3	0.063 ± 0.006	0.63 ± 0.02	29	0.032 ± 0.005	0.283 ± 0.012
4	0.115 ± 0.010	1.19 ± 0.04	30	0.053 ± 0.006	0.54 ± 0.02
5	0.185 ± 0.011	1.88 ± 0.06	31	0.098 ± 0.008	0.357 ± 0.014
6	0.082 ± 0.007	0.53 ± 0.02	32	0.046 ± 0.005	0.335 ± 0.014
7	0.098 ± 0.008	0.60 ± 0.02	33	0.072 ± 0.007	0.78 ± 0.03
8	0.046 ± 0.005	0.278 ± 0.012	34	0.097 ± 0.008	1.10 ± 0.04
9	0.040 ± 0.005	0.262 ± 0.012	35	0.046 ± 0.005	0.372 ± 0.015
10	0.032 ± 0.005	0.252 ± 0.011	36	0.039 ± 0.005	0.413 ± 0.016
11	0.091 ± 0.008	1.16 ± 0.04	37	0.046 ± 0.005	0.506 ± 0.019
12	0.102 ± 0.008	1.13 ± 0.04	38	0.046 ± 0.005	0.57 ± 0.02
13	0.053 ± 0.006	0.437 ± 0.017	39	0.071 ± 0.007	0.79 ± 0.03
14	0.031 ± 0.005	0.351 ± 0.015	40	0.040 ± 0.005	0.314 ± 0.013
15	0.075 ± 0.006	0.69 ± 0.03	41	0.058 ± 0.006	0.339 ± 0.014
16	0.101 ± 0.008	0.72 ± 0.03	42	0.143 ± 0.011	0.91 ± 0.03
17	0.107 ± 0.009	0.72 ± 0.03	43	0.064 ± 0.006	0.310 ± 0.014
18	0.071 ± 0.007	0.390 ± 0.016	44	0.054 ± 0.006	0.460 ± 0.018
19	0.039 ± 0.005	0.343 ± 0.014	45	0.046 ± 0.005	0.352 ± 0.014
20	0.035 ± 0.006	0.295 ± 0.014	46	0.040 ± 0.005	0.282 ± 0.012
21	0.052 ± 0.005	0.313 ± 0.012	47	0.052 ± 0.006	0.424 ± 0.017
22	0.072 ± 0.007	0.328 ± 0.014	48	0.040 ± 0.005	0.309 ± 0.013
23	0.097 ± 0.008	0.476 ± 0.018	49	0.040 ± 0.005	0.294 ± 0.012
24	0.086 ± 0.007	0.69 ± 0.03	50	0.070 ± 0.007	0.96 ± 0.03
25	0.039 ± 0.005	0.301 ± 0.013	51	0.118 ± 0.010	0.348 ± 0.015
26	0.045 ± 0.005	0.166 ± 0.009	52	0.070 ± 0.006	0.659 ± 0.019
			Avg.	0.0671 ± 0.0009	0.540 ± 0.003
			SD	0.03	0.33

Table A2: Weekly results of  $^7\text{Be}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  concentrations in air dust sampled by HVS in 1996 on the RIVM premises in Bilthoven (The Netherlands). Well-type detector. Empty fields indicate that the value was below the detection limit (Table A3). The error in the yearly average is equal to the square root of the sum of the squared weekly errors divided by the number of weeks. SD is the standard deviation of the weekly results.

week number	$^7\text{Be}$ $\mu\text{Bq}\cdot\text{m}^{-3}$	$^{137}\text{Cs}$ $\mu\text{Bq}\cdot\text{m}^{-3}$	$^{210}\text{Pb}$ $\mu\text{Bq}\cdot\text{m}^{-3}$
1	4100 ± 400	5.2 ± 0.5	910 ± 80
2	4100 ± 400	1.8 ± 0.2	640 ± 60
3	1800 ± 150	1.35 ± 0.13	770 ± 70
4	4800 ± 400	4.1 ± 0.3	1290 ± 110
5	3700 ± 200	5.6 ± 0.3	2070 ± 130
6	3100 ± 300	2.5 ± 0.3	570 ± 50
7	3600 ± 300	1.8 ± 0.2	570 ± 50
8	3300 ± 300	0.84 ± 0.13	230 ± 20
9	3300 ± 300	0.83 ± 0.19	260 ± 20
10	2400 ± 200	0.72 ± 0.14	180 ± 20
11	4200 ± 400	3.2 ± 0.3	1250 ± 110
12	3500 ± 300	2.7 ± 0.3	1540 ± 130
13	2900 ± 300	1.43 ± 0.17	420 ± 40
14	3000 ± 300	1.07 ± 0.17	360 ± 30
15	3500 ± 300	2.4 ± 0.2	770 ± 70
16	5900 ± 500	3.6 ± 0.3	650 ± 60
17	5800 ± 500	3.0 ± 0.3	620 ± 50
18	3900 ± 300	2.0 ± 0.3	320 ± 30
19	7500 ± 1300	3.6 ± 0.4	130 ± 10
20	2600 ± 200	1.5 ± 0.2	280 ± 20
21	3600 ± 300	1.10 ± 0.14	240 ± 20
22	3500 ± 300	0.71 ± 0.13	280 ± 20
23	5400 ± 500	1.18 ± 0.16	340 ± 30
24	5000 ± 400		590 ± 50
25	3700 ± 300	1.20 ± 0.20	190 ± 20
26	1700 ± 150	0.74 ± 0.16	120 ± 10

Table A2: Continued

week number	<sup>7</sup> Be μBq·m <sup>-3</sup>	<sup>137</sup> Cs μBq·m <sup>-3</sup>	<sup>210</sup> Pb μBq·m <sup>-3</sup>
27	2000 ± 180	1.1 ± 0.2	140 ± 10
28	2600 ± 200	0.44 ± 0.17	190 ± 20
29	3300 ± 300	0.85 ± 0.15	200 ± 20
30	7000 ± 600	1.09 ± 0.16	490 ± 40
31	3800 ± 300	0.57 ± 0.1	280 ± 30
32	3500 ± 300	1.09 ± 0.15	270 ± 20
33	6400 ± 600	1.4 ± 0.2	740 ± 70
34	7000 ± 600	1.9 ± 0.2	1000 ± 90
35	3200 ± 300	0.53 ± 0.14	270 ± 20
36	4200 ± 400	0.36 ± 0.17	290 ± 30
37	5600 ± 500	0.88 ± 0.14	300 ± 30
38	3500 ± 300	1.1 ± 0.11	480 ± 40
39	4800 ± 400	1.7 ± 0.2	830 ± 70
40	3600 ± 300	0.53 ± 0.13	220 ± 20
41	2600 ± 200	0.61 ± 0.09	350 ± 30
42	4400 ± 400	1.2 ± 0.16	950 ± 80
43	3200 ± 300	1.7 ± 0.2	270 ± 20
44	3600 ± 300	0.66 ± 0.15	390 ± 30
45	4000 ± 400		260 ± 20
46	2300 ± 200	0.67 ± 0.13	280 ± 20
47	2900 ± 300	0.81 ± 0.12	360 ± 30
48	2300 ± 200	0.75 ± 0.1	270 ± 20
49	4300 ± 400	0.62 ± 0.09	180 ± 20
50	4100 ± 400	1.58 ± 0.18	670 ± 60
51	3100 ± 300	0.97 ± 0.16	170 ± 20
52	5200 ± 500	2.6 ± 0.4	570 ± 50
Avg.	3890 ± 50	1.60 ± 0.03	500 ± 10
SD	1320	1.2	390

Table A3: Detection limits in  $\mu\text{Bq}\cdot\text{m}^{-3}$  of the well-type detector for a seven days sampling period, ten days between sampling and start of measurement, 200000 seconds counting time and a sample volume of about  $50000\text{ m}^3$  [22].

Nuclide	Detection limit	Nuclide	Detection limit
$^7\text{Be}$	1.4	$^{113}\text{Sn}$	0.1
$^{22}\text{Na}$	0.2	$^{115\text{m}}\text{Cd}$	8
$^{24}\text{Na}$	600 *	$^{123\text{m}}\text{Te}$	0.1
$^{40}\text{K}$	7	$^{124}\text{Sb}$	0.4
$^{51}\text{Cr}$	1.6	$^{125}\text{Sb}$	0.4
$^{54}\text{Mn}$	0.1	$^{129\text{m}}\text{Te}$	5
$^{57}\text{Co}$	0.1	$^{131}\text{I}$	1.3 **
$^{58}\text{Co}$	0.1	$^{132}\text{Te}$	3
$^{59}\text{Fe}$	0.3	$^{134}\text{Cs}$	0.2
$^{60}\text{Co}$	0.2	$^{136}\text{Cs}$	0.2
$^{65}\text{Zn}$	0.3	$^{137}\text{Cs}$	0.1
$^{75}\text{Se}$	0.2	$^{140}\text{Ba}$	0.6
$^{95}\text{Nb}$	0.2	$^{140}\text{La}$	22
$^{95}\text{Zr}$	0.4	$^{141}\text{Ce}$	0.2
$^{99}\text{Mo}$	20	$^{144}\text{Ce}$	0.7
$^{103}\text{Ru}$	0.1	$^{202}\text{Tl}$	0.3
$^{106}\text{Ru}$	0.9	$^{210}\text{Pb}$	5
$^{109}\text{Cd}$	3	$^{237}\text{U}$	2
$^{110\text{m}}\text{Ag}$	0.2		

\* Due to the relatively short half-life of  $^{24}\text{Na}$  and the long waiting time for the measurement (10 days waiting time), this nuclide cannot be determined on the well-type detector. Therefore, the detection limit for the coaxial detector is given (3 days waiting time, 100000 seconds counting time).

\*\* Due to the sample preparation procedure the volatile nuclide  $^{131}\text{I}$  cannot be determined on the well-type detector. Therefore, the detection limit for the coaxial detector is given (3 days waiting time, 100000 seconds counting time).

Table A4: Monthly values of precipitation and of  $^3\text{H}$ , long-lived gross  $\alpha$ - and gross  $\beta$ -activity in deposition sampled in 1996 on the RIVM premises in Bilthoven (The Netherlands). The error in the sum is equal to the square root of the sum of the squared monthly errors.

Month 1996	Precipitation mm	$^3\text{H}$ $\text{Bq}\cdot\text{m}^{-2}$	gross- $\alpha$ $\text{Bq}\cdot\text{m}^{-2}$	gross- $\beta$ $\text{Bq}\cdot\text{m}^{-2}$
January	9.5	9.5 $\pm$ 2.0	1.4 $\pm$ 0.5	3.0 $\pm$ 0.9
February	70.1	107 $\pm$ 13	1.4 $\pm$ 0.4	5.2 $\pm$ 1.3
March	12.4	13 $\pm$ 3	0.6 $\pm$ 0.2	1.6 $\pm$ 0.5
April	9.2	21.4 $\pm$ 1.8	1.2 $\pm$ 0.4	3.4 $\pm$ 0.9
May	37.6	53 $\pm$ 8	0.7 $\pm$ 0.2	2.4 $\pm$ 0.6
June	35.9	43 $\pm$ 8	1.6 $\pm$ 0.5	4.9 $\pm$ 1.3
July	52.1	104 $\pm$ 11	1.7 $\pm$ 0.5	7.2 $\pm$ 1.7
August	69.6	152 $\pm$ 15	1.6 $\pm$ 0.4	6.7 $\pm$ 1.6
September	44.4	89 $\pm$ 10	1.1 $\pm$ 0.4	5.5 $\pm$ 1.4
October	85	79 $\pm$ 18	1.3 $\pm$ 0.4	7.9 $\pm$ 1.9
November	144.3	210 $\pm$ 30	2.4 $\pm$ 0.6	11 $\pm$ 3
December	56.2	89 $\pm$ 12	1.3 $\pm$ 0.4	7.7 $\pm$ 1.9
Sum	626	970 $\pm$ 50	16.4 $\pm$ 1.5	67 $\pm$ 5

Table A5: Quarterly values of  $^{210}\text{Pb}$  en  $^{210}\text{Po}$  in deposition sampled in 1996 on the RIVM premises in Bilthoven (The Netherlands). Measurements are carried out using  $\alpha$ -spectroscopy. The error in the sum is equal to the square root of the sum of the squared quarterly errors.

Period 1996	$^{210}\text{Pb}$ $\text{Bq}\cdot\text{m}^{-2}$	$^{210}\text{Po}$ $\text{Bq}\cdot\text{m}^{-2}$
January - March	8.1 $\pm$ 0.9	1.9 $\pm$ 1.7
April - June	8.6 $\pm$ 0.9	2.9 $\pm$ 0.9
July - September	16.4 $\pm$ 1.6	2.8 $\pm$ 0.8
October - December	24.0 $\pm$ 2.3	1.7 $\pm$ 1.1
Sum	57 $\pm$ 3	9 $\pm$ 2



Table A6: Weekly values of  $^7\text{Be}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  deposition sampled in 1996 on the RIVM premises in Bilthoven (The Netherlands). Measurements are carried out using  $\gamma$ -spectroscopy. Empty fields indicate that the value was below the detection limit. The errors in the sum is equal to the square root of the sum of the squared weekly errors. SD is the standard deviation of the weekly results.

week number	precipitation mm	$^7\text{Be}$ $\text{Bq}\cdot\text{m}^{-2}$	$^{137}\text{Cs}$ $\text{Bq}\cdot\text{m}^{-2}$	$^{210}\text{Pb}$ $\text{Bq}\cdot\text{m}^{-2}$
1	1.7	8.1 ± 1.0		1.43 ± 0.18
2	8.0	6.8 ± 0.8		1.9 ± 0.2
3	0.2	0.70 ± 0.10		0.23 ± 0.05
4	0.1	2.2 ± 0.3		1.1 ± 0.15
5	0.2	5.6 ± 0.7	0.030 ± 0.007	5.3 ± 0.6
6	0.6	4.6 ± 0.6		0.74 ± 0.11
7	35.4	23 ± 3	0.021 ± 0.004	1.5 ± 0.2
8	26.2	61 ± 7	0.048 ± 0.009	(-) <sup>1</sup>
9	7.9	6.3 ± 0.7		0.33 ± 0.07
10	6.3	2.6 ± 0.3	0.020 ± 0.006	0.16 ± 0.05
11	0.0	0.84 ± 0.11		
12	1.5	3.3 ± 0.4		0.26 ± 0.06
13	2.3	2.0 ± 0.2	0.010 ± 0.003	0.13 ± 0.06
14	2.3	2.9 ± 0.4	0.017 ± 0.005	0.26 ± 0.06
15	8.4	33 ± 4		1.21 ± 0.17
16	0.0	2.8 ± 0.3	0.018 ± 0.005	0.46 ± 0.08
17	0.8	5.2 ± 0.6	0.025 ± 0.005	1.06 ± 0.16
18	0.25	5.4 ± 0.6	0.030 ± 0.006	0.89 ± 0.13
19	4.5	7.0 ± 0.8	0.057 ± 0.008	0.75 ± 0.12
20	0.1	1.2 ± 0.2	0.023 ± 0.004	0.33 ± 0.07
21	15.0	17 ± 2	0.035 ± 0.005	1.33 ± 0.18
22	17.8	22 ± 3		0.47 ± 0.1
23	1.5	14.9 ± 1.8		1.5 ± 0.2
24	7.7	28 ± 3	0.022 ± 0.006	1.9 ± 0.2
25	0.13	7.2 ± 0.9		0.91 ± 0.16
26	4.9	6.3 ± 0.7	0.022 ± 0.006	0.91 ± 0.14

(-)<sup>1</sup> Result is rejected

Table A6: Continued.

week number	precipitation mm	$^7\text{Be}$ $\text{Bq}\cdot\text{m}^{-2}$	$^{137}\text{Cs}$ $\text{Bq}\cdot\text{m}^{-2}$	$^{210}\text{Pb}$ $\text{Bq}\cdot\text{m}^{-2}$
27	32.8	17 ± 2	0.016 ± 0.005	1.14 ± 0.17
28	16.5	50 ± 6		0.83 ± 0.14
29	0.0	5.4 ± 0.6	0.020 ± 0.006	1.03 ± 0.14
30	23.0	55 ± 6	0.030 ± 0.006	4.9 ± 0.6
31	1.6	8.0 ± 0.9		0.5 ± 0.09
32	14.5	17 ± 2	0.017 ± 0.004	2.2 ± 0.3
33	10.5	10.1 ± 1.2	0.017 ± 0.005	0.66 ± 0.11
34	1.4	15.0 ± 1.8	0.021 ± 0.007	1.21 ± 0.17
35	37.5	31 ± 4		1.7 ± 0.2
36	5.8	9 ± 1.1		0.26 ± 0.06
37	9.2	15.3 ± 1.8	0.014 ± 0.004	0.4 ± 0.07
38	1.0	3.1 ± 0.4		0.3 ± 0.07
39	11.9	22 ± 3	0.019 ± 0.004	2.7 ± 0.3
40	32.0	47 ± 6	0.014 ± 0.004	1.9 ± 0.3
41	6.2	27 ± 3		1.2 ± 0.16
42	0.25	0.89 ± 0.12		
43	26.0	43 ± 5		2.3 ± 0.5
44	37.0	24 ± 3		1.7 ± 0.3
45	51.5	47 ± 6		2.8 ± 0.4
46	25.5	55 ± 6		2.7 ± 0.4
47	27.0	46 ± 5		2.3 ± 0.3
48	23.0	41 ± 5		2.4 ± 0.3
49	25.3	34 ± 4		2.3 ± 0.3
50	1.0	1.7 ± 0.2		0.31 ± 0.06
51	27.3	8.7 ± 1.0		1.29 ± 0.17
52	2.5	9.3 ± 1.1		0.77 ± 0.12
Sum		920 ± 20	0.550 ± 0.030	64.9 ± 1.6
SD		17	0.010	1.1

Table A7: Quarterly activity concentrations of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$  and  $\gamma$ -emitters in 1996 in a mixed sample of four regional milk samples from the north, south, east and west of the Netherlands. The error in the yearly average is equal to the square root of the sum of the squared quarterly errors divided by four.

date 1996	$^{89}\text{Sr}$ $\text{Bq}\cdot\text{L}^{-1}$	$^{90}\text{Sr}$ $\text{Bq}\cdot\text{L}^{-1}$	$^7\text{Be}$ $\text{Bq}\cdot\text{L}^{-1}$	$^{134}\text{Cs}$ $\text{Bq}\cdot\text{L}^{-1}$	$^{137}\text{Cs}$ $\text{Bq}\cdot\text{L}^{-1}$	$^{40}\text{K}$ $\text{Bq}\cdot\text{L}^{-1}$
1/1 - 31/3	< 0.5	< 0.2	< 0.5	< 0.02	$0.07 \pm 0.02$	$50 \pm 3$
1/4 - 30/6	< 0.5	< 0.2	< 0.4	< 0.03	$0.07 \pm 0.02$	$48 \pm 3$
1/7 - 30/9	< 0.5	< 0.2	< 0.5	< 0.02	$0.06 \pm 0.02$	$49 \pm 3$
1/10 - 31/12	< 0.5	< 0.2	< 0.3	< 0.03	$0.09 \pm 0.02$	$49 \pm 3$
Average	< 0.5	< 0.2	< 0.4	< 0.03	$0.073 \pm 0.010$	$49.0 \pm 1.5$