National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Monitoring of radioactivity in the Netherlands

Air dust and deposition – results 2020 and 2021

RIVM letter report 2023-0087
P.P. Bosch

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Synopsis

Monitoring of radioactivity in the Netherlands

Air dust and deposition – results 2020 and 2021

In 2020 and 2021, the Netherlands fulfilled its annual European obligation to measure how much radioactivity is present in the environment. Radioactivity levels in air dust and deposition were normal and similar to previous years.

All countries of the European Union are required to perform these measurements each year under the terms of the Euratom Treaty of 1957. The Netherlands performs these measurements following the guidance issued in 2000.

The measurements represent the background values for radioactivity that are present under normal circumstances. They can be used as reference values, for instance, during a nuclear emergency.

The results on radioactivity in the environment in the Netherlands are reported to the European Commission by the National Institute for Public Health and the Environment (RIVM) on behalf of the Authority for Nuclear Safety and Radiation Protection (ANVS).

Keywords: radioactivity, air dust, deposition

Publiekssamenvatting

Monitoring van radioactiviteit in Nederland

Luchtstof en depositie – resultaten 2020 en 2021

In 2020 en 2021 voldeed Nederland aan de Europese verplichting om elk jaar de hoeveelheid radioactiviteit in het milieu te meten. De niveaus radioactiviteit in stof in de lucht en op de bodem laten een normaal beeld zien, net als in eerdere jaren.

Alle landen van de Europese Unie zijn volgens het Euratom-verdrag uit 1957 verplicht om deze metingen te doen. Nederland volgt daarbij de aanbevelingen uit 2000 op om de metingen op een bepaalde manier uit te voeren.

De metingen leveren achtergrondwaarden op, ofwel radioactiviteitsniveaus die er onder normale omstandigheden zijn. Deze waarden kunnen bij bijvoorbeeld calamiteiten of rampen als referentie dienen.

Het RIVM brengt namens de Autoriteit Nucleaire Veiligheid en Stralingsbescherming (ANVS) verslag uit aan de Europese Unie over radioactiviteit in het Nederlandse milieu.

Kernwoorden: radioactiviteit, luchtstof, neerslag

Disclaimer

The Authority for Nuclear Safety and Radiation Protection (ANVS, Koningskade 4, The Hague; https://english.autoriteitnvs.nl/) has given RIVM, Centre for Environmental Safety and Security, department SMA (RIVM, Antonie van Leeuwenhoeklaan 9, 3721 MA Bilthoven) the assignment to perform the measurements presented here and to make this report.

Department SMA of the Centre for Environmental Safety and Security of the National Institute for Public Health and the Environment (RIVM) has been accredited by the Dutch Accreditation Council (RvA) according to NEN-EN-ISO/IEC 17025:2017 (RvA: L153 Testing) for a number of procedures. In this report, all measurements are performed within the scope of this accreditation (except the ^{3}H measurements, which are performed by Rijkswaterstaat according to their ISO 17025 accreditation). The sampling and any opinions and/or interpretations that are expressed are outside the scope of accreditation.

Contents

Summary

In 2020 and 2021 radioactivity was monitored in air dust and deposition at one central location in the Netherlands. The yearly average activity concentration in air dust and the yearly total activity in deposition was determined for gross α, gross β, ^{7}Be, ^{137}Cs and ^{210}Pb. In addition, the yearly total activity in deposition of ^{3}H and ^{210}Po was determined. Gross α and gross β are the total activity of radionuclides emitting α and β radiation, respectively. The results, which are presented in Table 1, are within the range of those presented in previous years.

Table 1 Yearly averages and yearly totals for activity concentrations in air dust and deposition, respectively, at Bilthoven in 2020 and 2021.

Matrix/ parameter	Values (1) 2020	Values (1) 2021	Frequency (per year)
Air dust (average)			
Gross α (2)	0.019 mBq·m^{-3}	0.022 mBq·m^{-3}	52
Gross β	0.381 ± 0.007 mBq·m^{-3}	0.395 ± 0.007 mBq·m^{-3}	52
^{7}Be	3,400 ± 30 µBq·m^{-3}	3,490 ± 40 µBq·m^{-3}	52
^{137}Cs	0.270 ± 0.006 µBq·m^{-3}	0.201 ± 0.004 µBq·m^{-3}	52
^{210}Pb	294 ± 4 µBq·m^{-3}	311 ± 4 µBq·m^{-3}	52
Matrix/ parameter	**Values (1) 2020**	**Values (1) 2021**	**Frequency (per year)**
Deposition (total)			
Gross α (2)	68 ± 2 Bq·m^{-2}	62.7 ± 1.7 Bq·m^{-2}	12
Gross β	99.1 ± 1.8 Bq·m^{-2}	104.0 ± 1.8 Bq·m^{-2}	12
^{3}H	800 ± 30 Bq·m^{-2}	830 ± 40 Bq·m^{-2}	12
^{7}Be	1,502 ± 19 Bq·m^{-2}	1,670 ± 20 Bq·m^{-2}	52
^{137}Cs (3)	0.0 – 1.1 Bq·m^{-2}	0.0 – 1.1 Bq·m^{-2}	52
^{210}Pb	100.1 ± 1.4 Bq·m^{-2}	120.7 ± 1.7 Bq·m^{-2}	52
^{210}Po	39.9 ± 1.1 Bq·m^{-2}	39.8 ± 1.0 Bq·m^{-2}	12

(1) Uncertainties are given as 1σ (68% confidence level) as defined in the Appendix.
(2) Values are indicative due to large uncertainties caused by variations in the amount of dust on the filters [1].
(3) Lower and upper limits of the 68% confidence interval are given as defined in the Appendix.

[1] NEN, 2006. NEN 5636, Radioactivity measurements – Determination of artificial gross-alpha activity, artificial gross-beta activity and gamma spectrometry of air filters and calculation of the volumic activity of sampled air. NEN, Delft.

1 Introduction

This report presents the results of the monitoring programme for the determination of radionuclides in air dust and in deposition. The sampling was performed at the RIVM premises in Bilthoven, the Netherlands. Details of the sampling and measuring procedures at RIVM can be found elsewhere [2], with the exception of (a change in) the collection efficiency of filter type used to collect the air dust. The collection efficiency of the new batch of G-3 filters was determined to be 91 ± 3% and is used in the calculation of the 2020 and 2021 data. The measurements performed by RIVM are accredited according to ISO 17025 [3]. Air dust samples for the measurement of gross α, gross β and γ emitters were collected weekly using a high-volume sampler and analysed spectroscopically for specific γ-emitting radionuclides. A sub-sample of the total filter is analysed for gross α and gross β activities. Samples of deposition were collected weekly for γ emitters and monthly for gross α, gross β, ^{3}H and ^{210}Po. The monthly samples for ^{3}H were analysed by Rijkswaterstaat (RWS). These measurements are also accredited according to ISO 17025 [4].

[2] Environmental radioactivity in the Netherlands, Results in 2018, CP Tanzi (ed.), RIVM report 2019-0216, 2020.
[3] Raad van Accreditatie, scope L153. Web page https://www.rva.nl/ (March 2021).
[4] Raad van Accreditatie, scope L521. Web page https://www.rva.nl/ (March 2021).

2 Results

2.1 Results for long-lived α and β activity in air dust

The weekly results of gross α and β activity concentrations in air dust are given for 2020 in Figure 1 and for 2021 in Figure 2. The yearly averages from 1992 onwards are given in Figure 3. The activity concentrations of ^{210}Pb (as measured by γ-spectrometry) is also shown in Figure 1 and Figure 2. Normally, as is also the case in 2020 and 2021, there is a good correlation between gross β and naturally occurring γ-emitting ^{210}Pb (in equilibrium with its β-emitting daughter ^{210}Bi) activity concentrations, which suggests that, under normal conditions, ^{210}Pb/^{210}Bi is the main contributor to the gross β value.

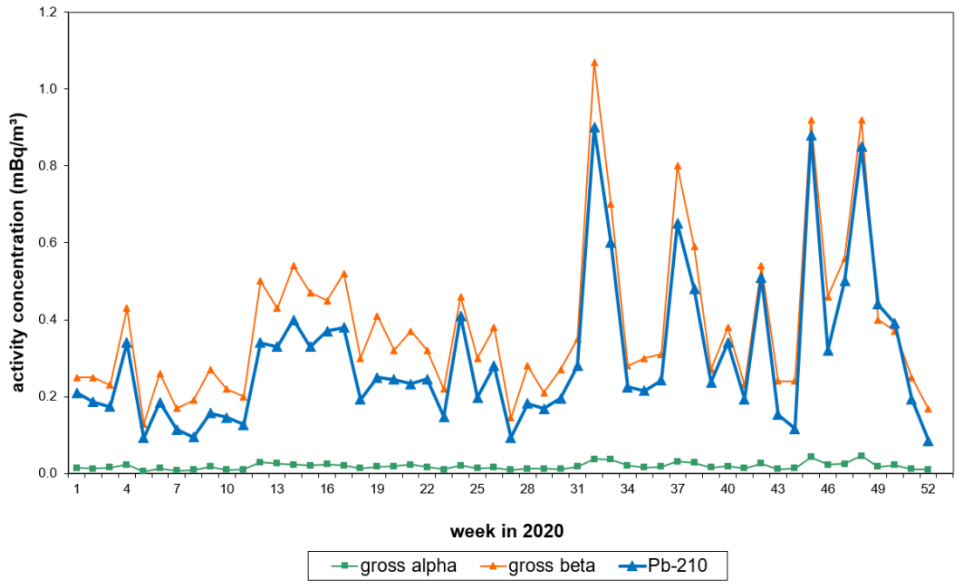

Figure 1 Weekly average ^{210}Pb and gross α and β activity concentrations of long-lived radionuclides in air dust sampled at RIVM in 2020. Weekly average gross β and ^{210}Pb activity concentrations in air dust at RIVM show a good correlation.

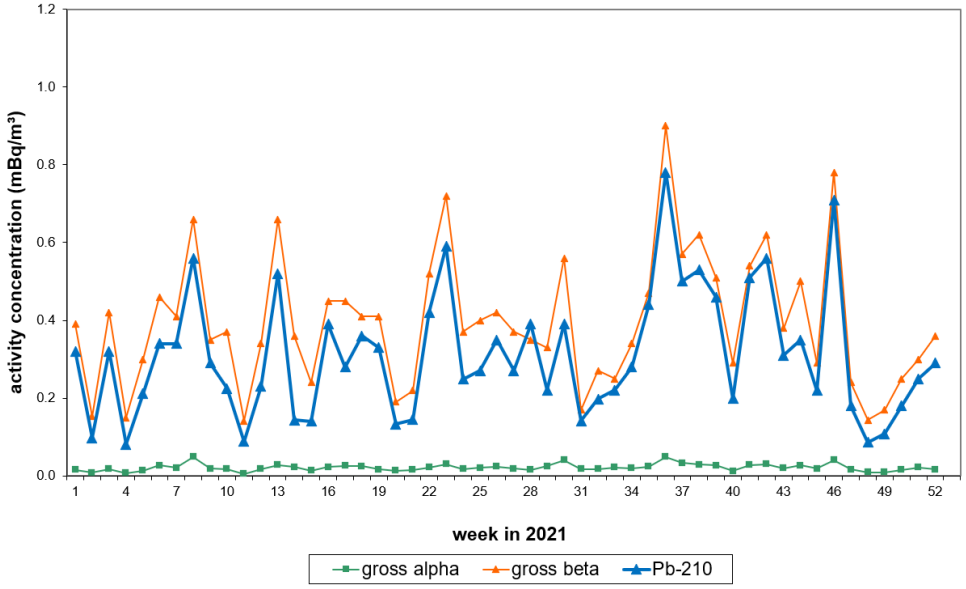

Figure 2 Weekly average ^{210}Pb and gross α and β activity concentrations of long-lived radionuclides in air dust sampled at RIVM in 2021. Weekly average gross β and ^{210}Pb activity concentrations in air dust at RIVM show a good correlation.

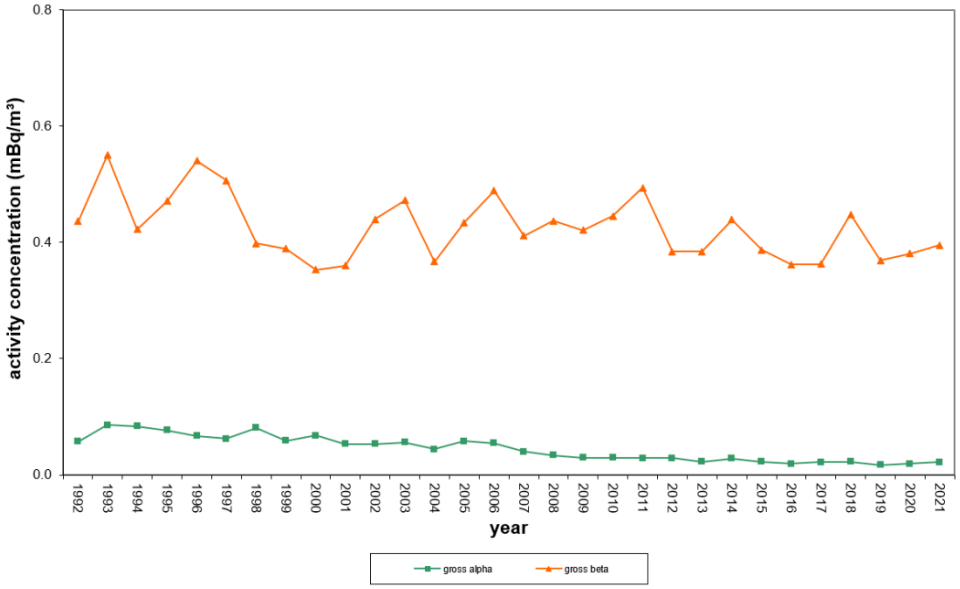

Figure 3 Yearly average gross α and gross β activity concentrations of long lived radionuclides in air dust at RIVM since 1992.

2.2 Results for γ-emitting radionuclides in air dust

Several γ emitting radionuclides were detected in the sampled air dust: ^{7}Be (52 times in both 2020 and 2021), ^{137}Cs (43 times in 2020 and 50 times in 2021) and ^{210}Pb (52 times in both 2020 and 2021). The results for these radionuclides since 1991 are presented in Figure 4, Figure 5 and Figure 6, respectively.

Figure 4 shows ^{7}Be detected in the sampled air dust. Natural ^{7}Be (half-life of 53.3 days) is formed by spallation reactions of cosmogenic radiation with atmospheric nuclei such as nitrogen and oxygen, resulting

in the formation of BeO or $Be(OH)_2$. Approximately 70% of ^7Be is produced in the stratosphere (with an estimated residence time of one to two years) and 30% in the troposphere (with an estimated residence time of approximately six weeks). Most of the ^7Be produced in the stratosphere does not reach the troposphere, except during spring, when seasonal thinning of the tropopause takes place at mid-latitudes, resulting in air exchange between the stratosphere and the troposphere.

In the troposphere, ^7Be rapidly associates mainly with submicron sized aerosol particles, and gravitational settling and precipitation processes accomplish its transfer to the earth's surface. Seasonal variations in the concentration of ^7Be in surface air are influenced by the following main atmospheric processes: wet and dry deposition, mass exchange between stratosphere and troposphere, vertical transport in the troposphere, and horizontal transport of air masses from the subtropics and mid-latitudes to the tropics and polar regions.

In Figure 4 the peaks of the ^7Be activity concentration during the spring and summer periods reflect the seasonal variations in the transport rate of air from the stratosphere to the troposphere. The influence of the solar cycle can also be seen: the elevated values of 1997 and the 2007–2009 period, and the lower values of the 2000–2002 and 2014-2015 periods are consistent with the solar minima (measured by radio flux and sunspot count) of the 1996–1997 and 2008–2009 periods, and the solar maximum of the 2000–2002 and 2014-2015 periods, respectively [5]. In the summer of 1991, two severe geomagnetic storms caused a significant worldwide disturbance of the earth's geomagnetic field. This resulted in a considerable decrease in cosmogenic radiation, which was unprecedented in at least the previous four decades [6]. The absence of a 1991 summer peak in the ^7Be activity concentration can be explained by the decrease in cosmogenic radiation. The concentrations found for ^7Be in 2020 and 2021 fit into the pattern described above.

[5] Solar Cycle Progression. Web page: www.swpc.noaa.gov/products/solar-cycle-progression (March 2021).
[6] R.C.G.M. Smetsers and R.O. Blaauboer, 1997. A dynamic compensation method for natural ambient dose rate based on 6 years' data from the Dutch Radioactivity Monitoring Network. Radiation Protection Dosimetry 69 (1), 19–31

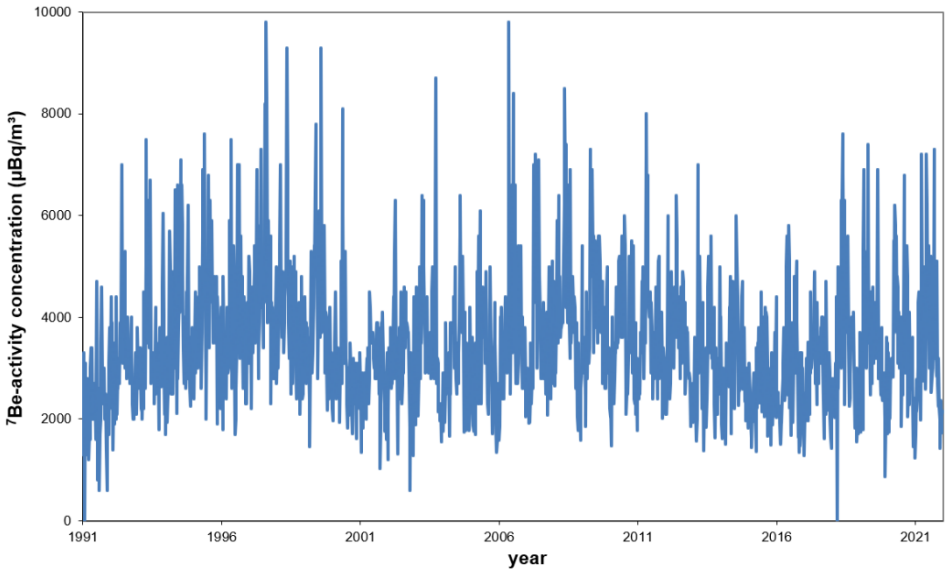

Figure 4 Weekly average ^{7}Be activity concentrations in air dust at RIVM since 1991.

Figure 5 shows ^{137}Cs detected in the sampled air dust. The nuclide ^{137}Cs (half-life of 30.2 years) is of anthropogenic origin. The two main sources of ^{137}Cs in the environment are atmospheric nuclear weapons tests and some nuclear accidents, most notably the Chernobyl accident of 1986 and the Fukushima Daiichi accident of 2011. Resuspension of previously deposited activity has been the main source of airborne ^{137}Cs activity in the Netherlands since 1986. Figure 5 shows a lack of data between 2000 and the middle of 2009. During that period the detection limit was higher than it had been during the 1991–1999 period due to a different detector set-up. Since July 2009, a new detector set-up has been used, which results in lower detection limits (similar to those used before 2000).

In Figure 5 two peaks are clearly visible:

- May 1992: several wildfires occurred near the Chernobyl area [7] and the level of airborne ^{137}Cs activity increased ten times in the 30 km exclusion zone around Chernobyl. It is possible that the airborne ^{137}Cs was transported to Western Europe by a strong easterly wind in the same period.
- 18 March to 10 June 2011: elevated levels of ^{137}Cs activity were measured as a result of the incident at Fukushima (Japan). More detailed results on ^{137}Cs and other radionuclides during that period are presented in [8].

In addition, on 29 May 1998, an incident occurred at Algeciras (Spain), an iron foundry melted a ^{137}Cs source concealed in scrap metal [9]. As a result, elevated levels of airborne ^{137}Cs activity were measured in

[7] S.I. Dusha-Gudym, 2005. Transport of radioactive materials by wildland fires in the Chernobyl accident zone: how to address the problem. International Forest Fire News 32. Web page: gfmc.online/wp-content/uploads/20-Dusha-Gudym-3.pdf (March 2020).
[8] G.J. Knetsch (ed.), 2013. Environmental radioactivity in the Netherlands. Results in 2011. RIVM Bilthoven, Report no. 610891004.
[9] T. Hantke, F.J. Aldenkamp, R.M.W. Overwater, H. Slaper, 1998. De jacht op een 137Cs-wolk in Zuid-Europa – 'aftermath' van een ongeval in Algeciras. NVS Nieuws 23 (4).

France, Germany, Italy and Switzerland during late May and early June. Figure 5 shows a slightly elevated level of ^{137}Cs activity around the same period (29 May to 5 June 1998). Such slightly elevated levels are not uncommon, as can be seen in Figure 5. These elevated values may be related to the resuspension of previously deposited dust, especially during a period of strong winds from the continent.

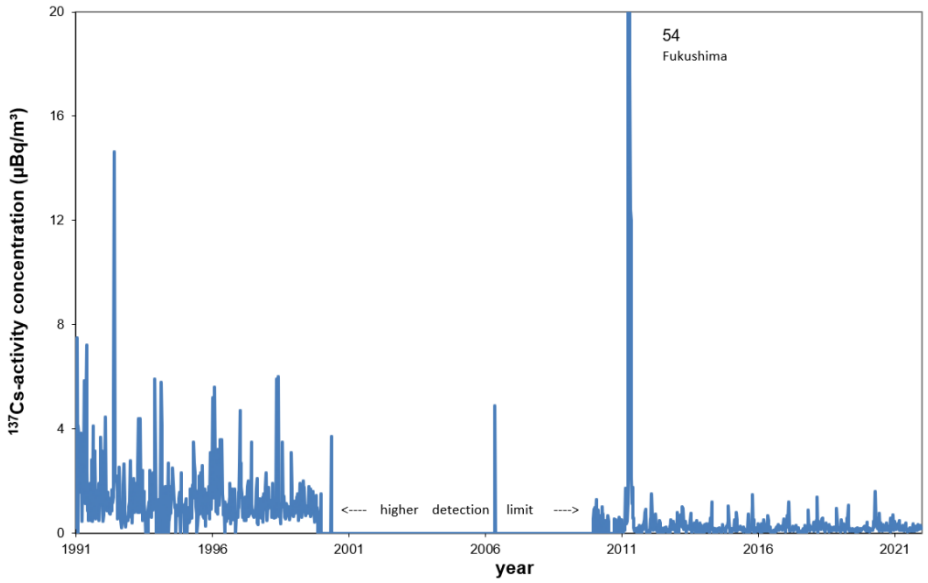

Figure 5 Weekly average ^{137}Cs activity concentrations in air dust at RIVM since 1991.

Figure 6 shows ^{210}Pb detected in the sampled air dust. The primary source of atmospheric ^{210}Pb (half-life of 22.3 years) is the decay of ^{222}Rn exhaled from continental surfaces. The reported UNSCEAR reference level [10] of ^{210}Pb in air dust is 500 µBq·m^{-3}. Unusual (high) ^{210}Pb values might be explained by natural phenomena such as an explosive volcanic eruption, Saharan dust [11, 12, 13] or the resuspension of (local) dust. The weekly average ^{210}Pb activity concentrations in 2020 and 2021 were within the range of those found in previous years.

[10] UNSCEAR, 2000. Sources and effects of ionizing radiation. Volume 1: Sources.
[11] E.Y. Nho, B. Ardouin, M.F. Le Cloarec, M. Ramonet, 1996. Origins of ^{210}Po in the atmosphere at Lamto, Ivory Coast: biomass burning and Saharan dust. Atmospheric Environment 30 (22), 3705–3714.
[12] M.A. Mélières, M. Pourchet, S. Richard, 2003. Surface air concentration and deposition of lead-210 in French Guiana: two years of continuous monitoring. Journal of Environmental Radioactivity 66, 261–269.
[13] P.G. Appleby, A.O. Koulikov, L. Camarero, M. Ventura, 2002. The input and transmission of fall-out radionuclides through Redó, a high mountain lake in the Spanish Pyrenees. Water, Air & Soil Pollution: Focus 2, 19–31.

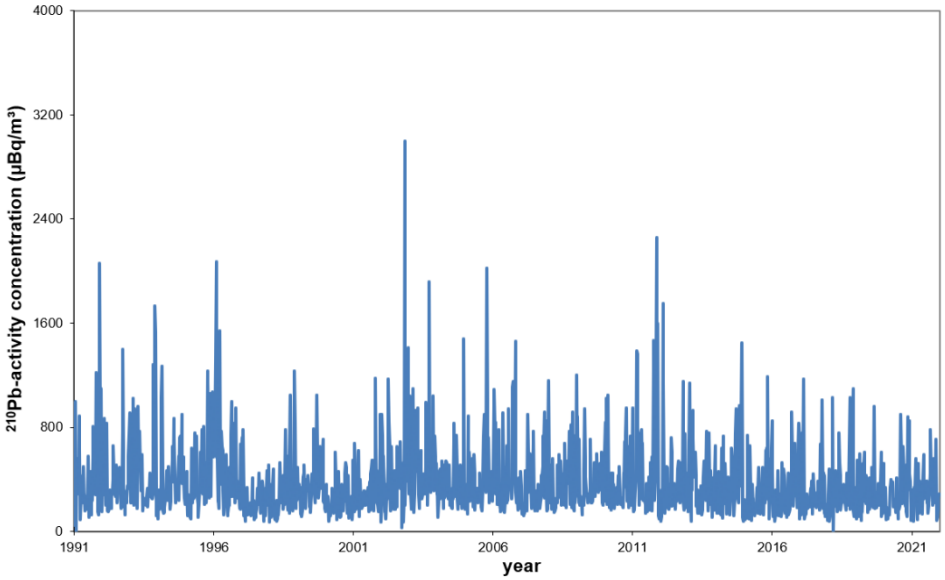

Figure 6 Weekly average ^{210}Pb activity concentrations in air dust at RIVM since 1991.

2.3 Results for long-lived α and β activity in deposition

The monthly deposited gross α and gross β activities of long lived radionuclides are given in Figure 7 and Figure 10 for 2020 and Figure 8 and Figure 11 for 2021. The yearly total depositions of gross α and gross β are shown in Figure 9 and Figure 12 respectively. In April 2020 and June 2021 a higher deposition of gross α and gross ß in respect to all other months is visible. The yearly total deposition of gross α and gross β is within the range of those from previous years.

The monthly α spectroscopy results for ^{210}Po are given in Figure 13. In April 2020 and June 2021 a higher deposition of ^{210}Po in respect to all other months is visible, as is for gross α and gross ß. The monthly total depositions of ^{210}Po is within the range of those from previous years. The results for the years 2009 to 2019 have been recalculated with respect to previous reports. This change is due to a change in the radioactive decay correction. The new concentrations are all lower than previously reported (up to ± 10 percent lower).

The yearly total deposition of ^{3}H is given in Figure 14. In 2020 and 2021, the yearly totals consisted both of 12 samples which were all above the detection limit. The range in 2020 and 2021 did not differ significantly from those measured since 1993. Up to 1998, samples were electrolytically enriched before counting, which resulted in a much lower detection limit for ^{3}H than the one achieved from 1998 onwards (1.6-2.0 Bq·L^{-1}). Since 2017 the samples are analysed for ^{3}H by Rijkswaterstaat (RWS) with a lower detection limit (0.3 Bq·L^{-1}) than in the period between 1998 and 2016.

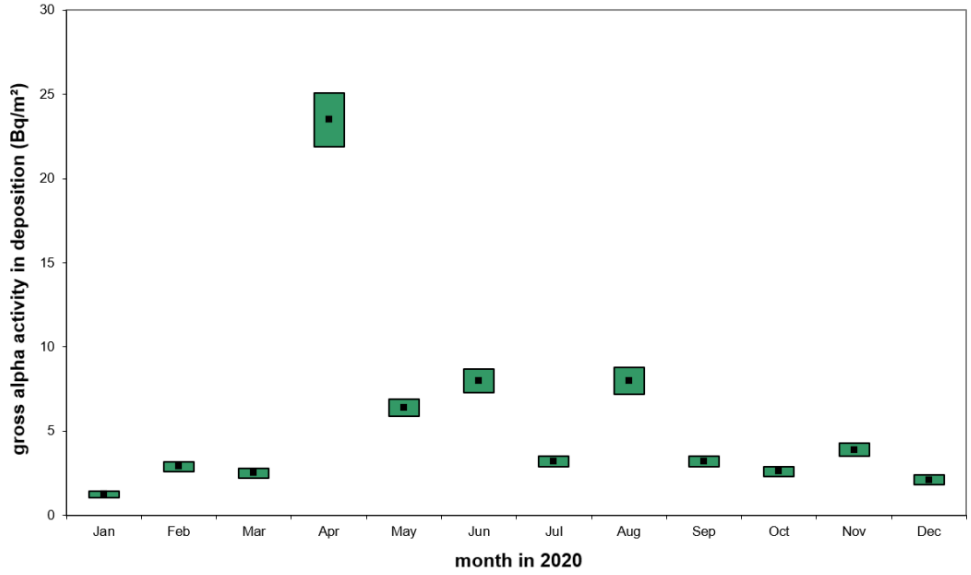

Figure 7 Monthly totals of deposition (black dots) of gross α activity of long-lived radionuclides at RIVM in 2020 are shown with a 68% confidence interval (coloured bars).

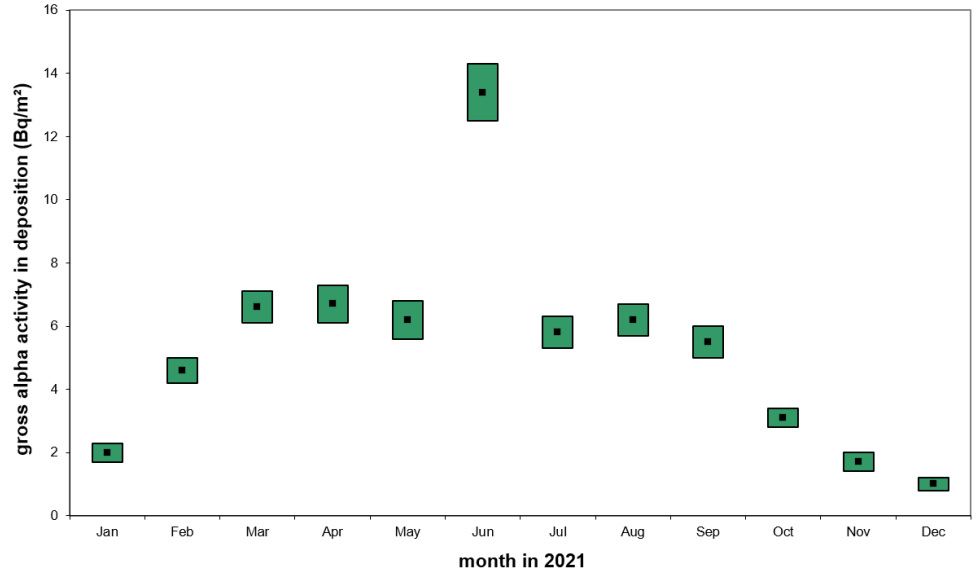

Figure 8 Monthly totals of deposition (black dots) of gross α activity of long-lived radionuclides at RIVM in 2021 are shown with a 68% confidence interval (coloured bars).

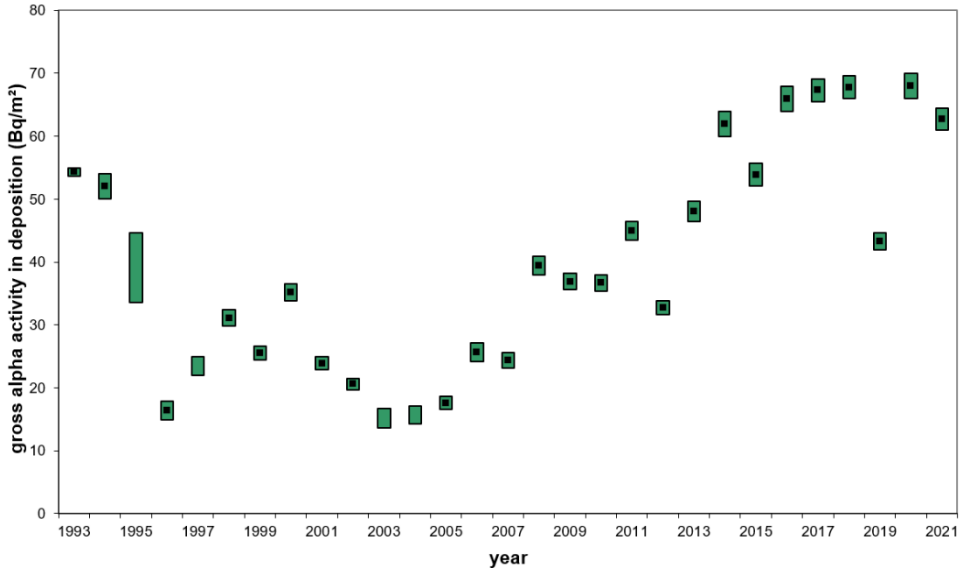

Figure 9 Yearly deposition of gross α activity (black dots) of long lived radionuclides at RIVM since 1993 shown with a 68% confidence interval (coloured bars). If the sum leading to the yearly total contains at least one detection limit, only the 68% confidence interval is shown.

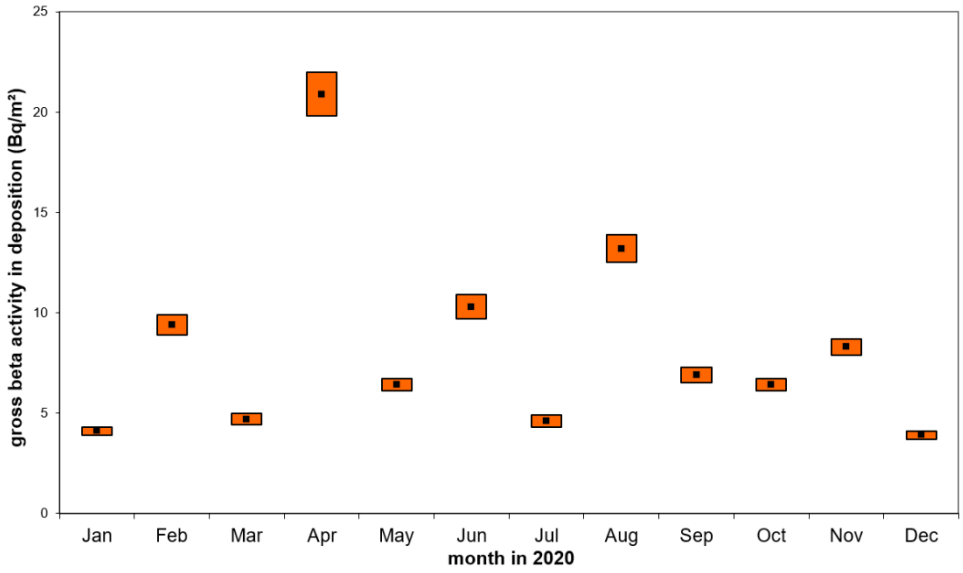

Figure 10 Monthly totals of deposition (black dots) of gross β activity of long-lived radionuclides at RIVM in 2020 are shown with a 68% confidence interval (coloured bars).

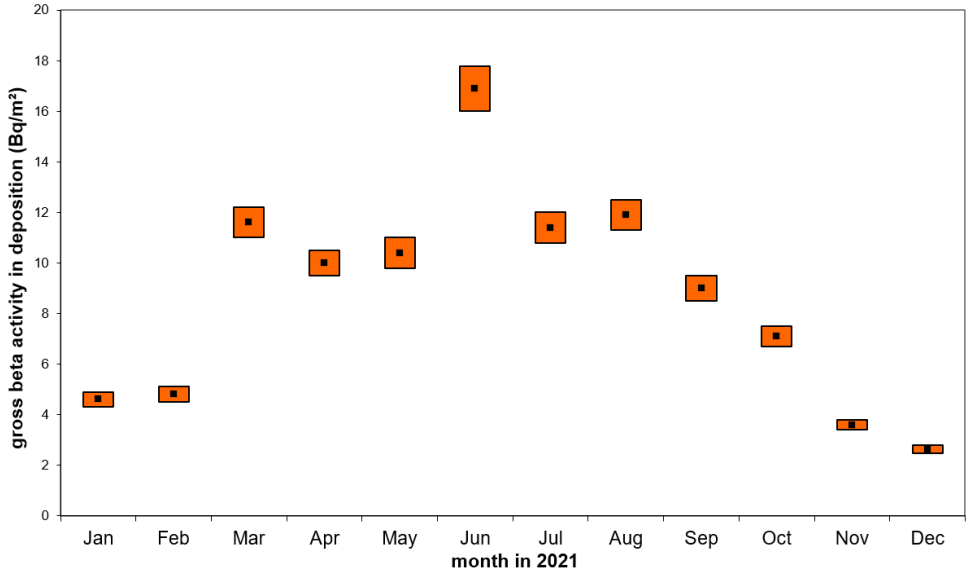

Figure 11 Monthly totals of deposition (black dots) of gross β activity of long-lived radionuclides at RIVM in 2021 are shown with a 68% confidence interval (coloured bars).

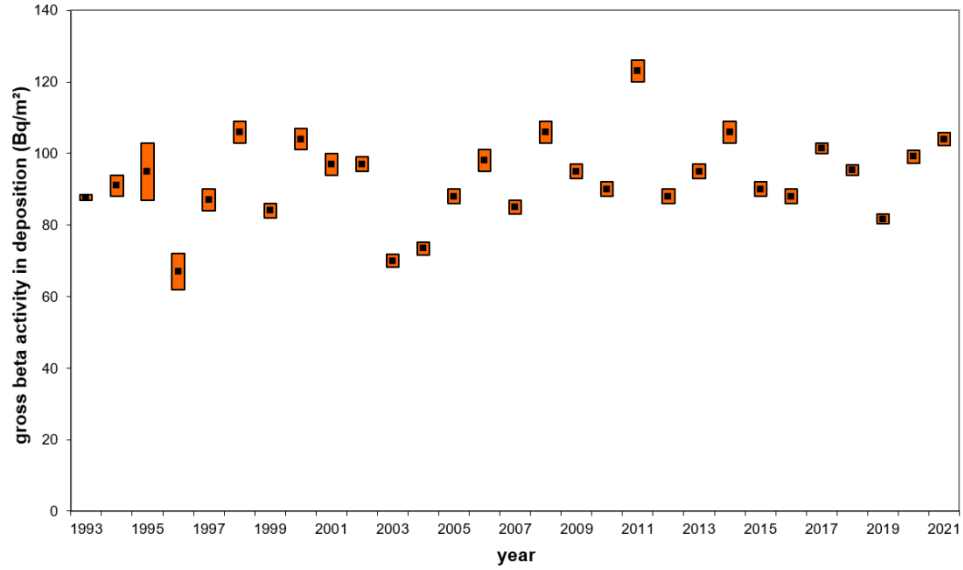

Figure 12 Yearly deposition of gross β activity (black dots) of long lived radionuclides at RIVM since 1993 shown with a 68% confidence interval (coloured bars). If the sum leading to the yearly total contains at least one detection limit, only the 68% confidence interval is shown.

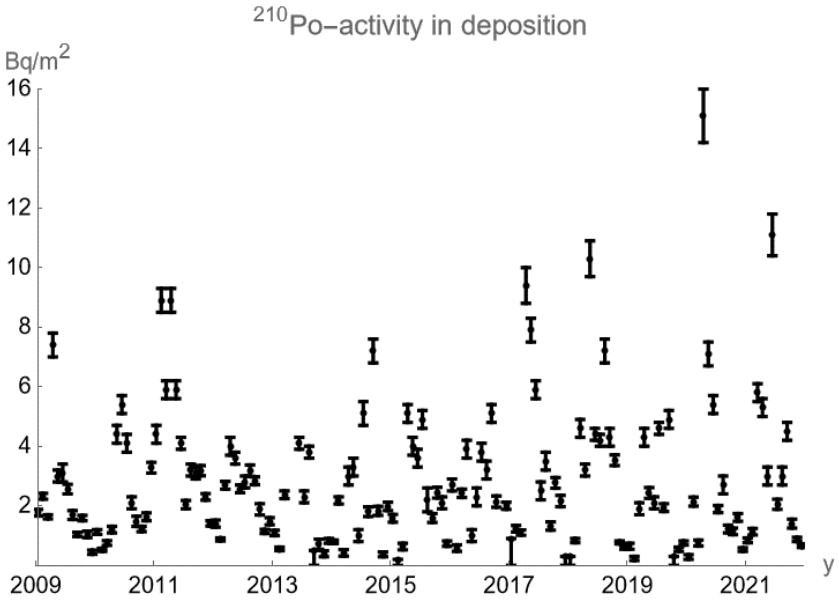

Figure 13 Monthly totals of deposition (black dots) of ^{210}Po activity at RIVM since 2009 are shown with a 68% confidence interval (bars). If the total is less than the detection limit, only the 68% confidence interval is shown.

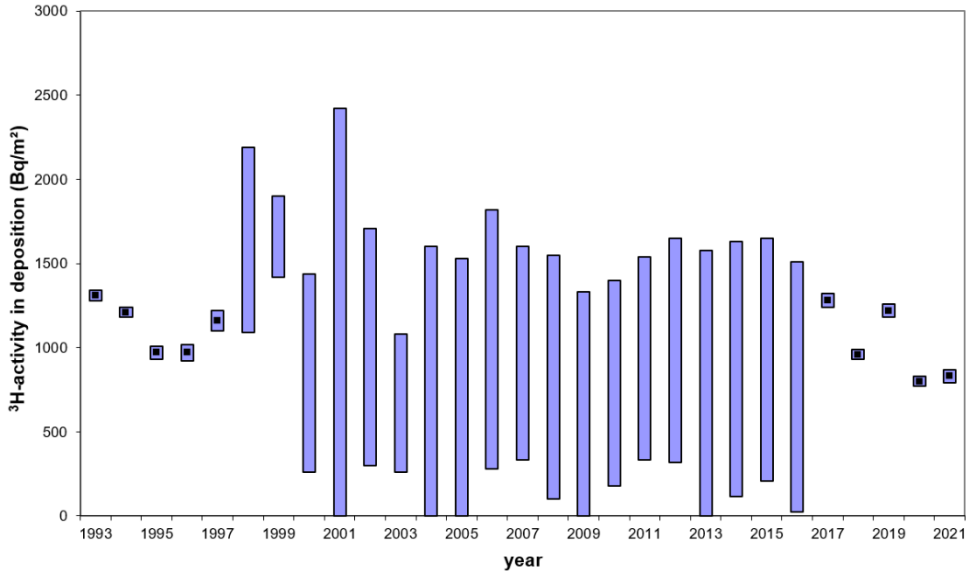

Figure 14 Yearly deposition of ^{3}H activity (black dots) sampled at RIVM since 1993 are shown with a 68% confidence interval (coloured bars). If the sum leading to the yearly total contains at least one detection limit, only the 68% confidence interval is shown. Since 2017, the samples are analysed by Rijkswaterstaat (RWS).

2.4 Results for γ-emitting radionuclides in deposition

The naturally occurring radionuclides ^{7}Be (Figure 15 for 2020 and Figure 16 for 2021) and ^{210}Pb (Figure 18 for 2020 and Figure 19 for 2021) were found in all of the 52 weekly deposition samples for both 2020 and 2021. The nuclide ^{137}Cs was found in two weekly deposition samples in 2020. In all other samples, the ^{137}Cs concentration was below the detection level (the detection limit for ^{137}Cs is 0.02 Bq·m^{-2} on average).

The results for previous years are given in Figure 17, Figure 20 and Figure 21. Figure 21 shows a change in the trend for ^{137}Cs between 2000 and the middle of 2009. During that period, the detection limit was higher than it had been during the 1993–1999 period due to a different detector set-up. Since July 2009, a new detector set-up has been used, which results in lower detection limits. Since the ^{137}Cs deposition values are mostly below the detection limit, no conclusion can be drawn concerning the correlation between the measured ^{137}Cs values in air dust and the measured ^{137}Cs values in deposition.

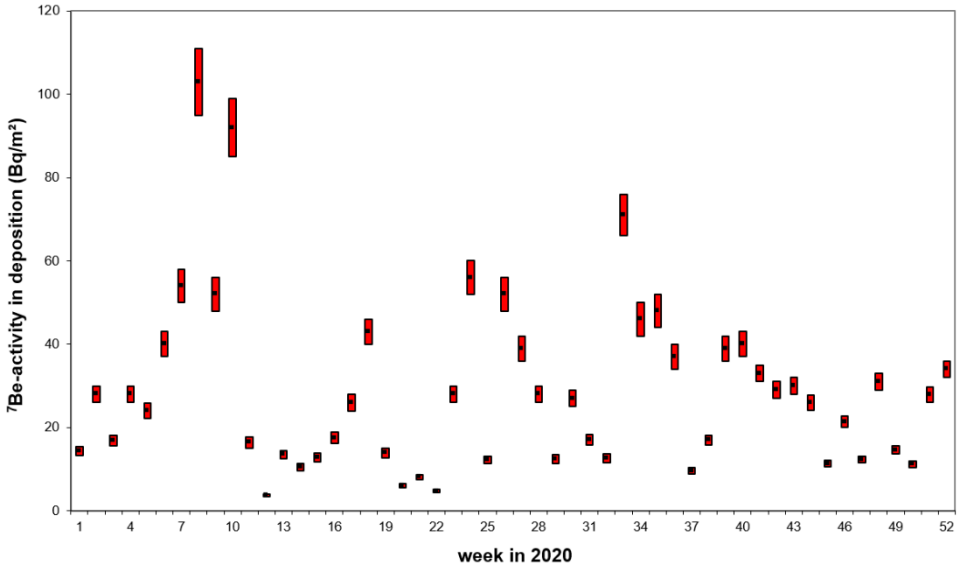

Figure 15 Weekly totals of deposition (black dots) of ^{7}Be activity at RIVM in 2020 are shown with a 68% confidence interval (coloured bars).

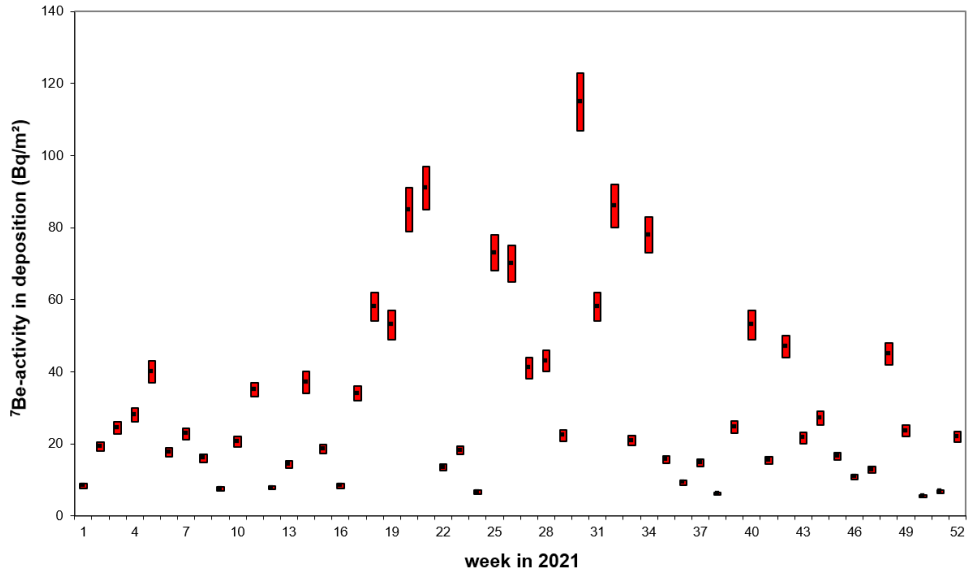

Figure 16 Weekly totals of deposition (black dots) of ^{7}Be activity at RIVM in 2021 are shown with a 68% confidence interval (coloured bars).

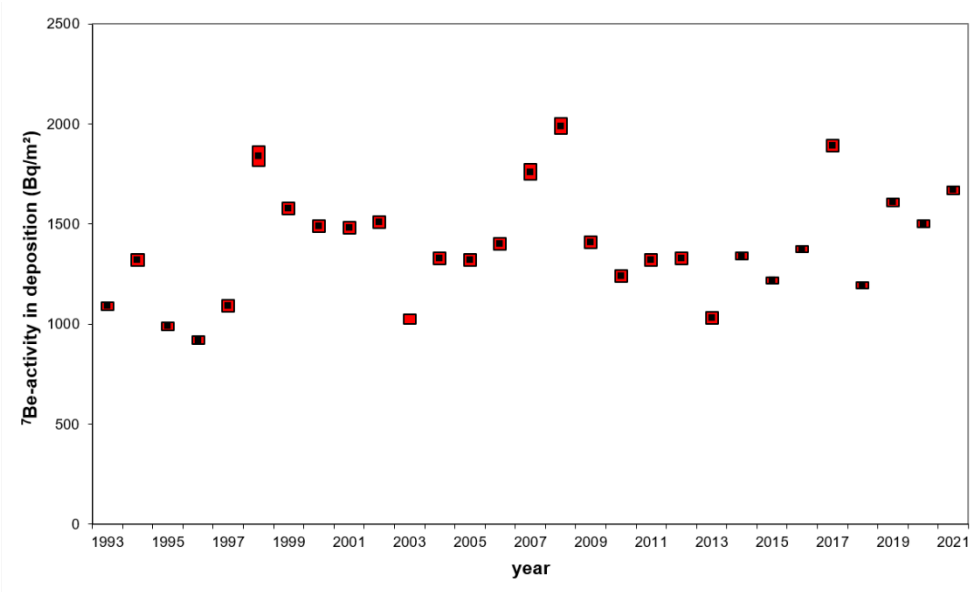

Figure 17 Yearly totals of ^{7}Be activity deposited at RIVM (black dots) since 1993 shown with a 68% confidence interval (coloured bars). In 2003 the sum leading to the yearly total contained at least one detection limit, and only the 68% confidence interval is shown.

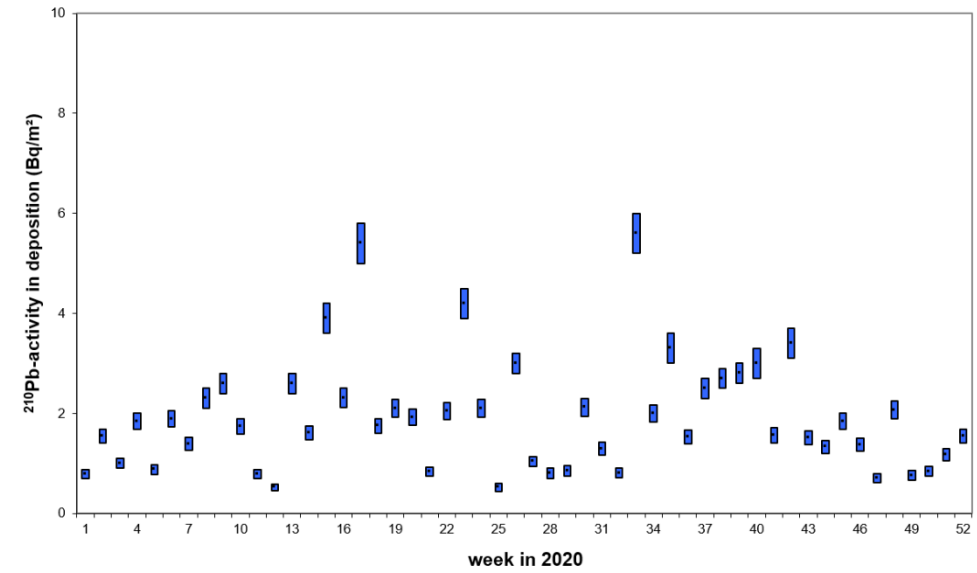

Figure 18 Weekly totals of deposition (black dots) of ^{210}Pb activity at RIVM in 2020 are shown with a 68% confidence interval (coloured bars).

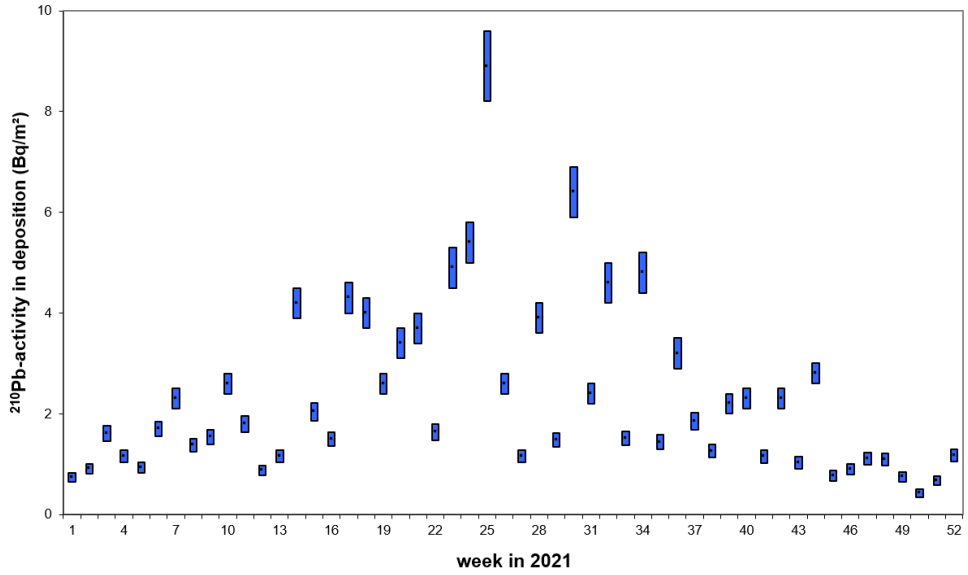

Figure 19 Weekly totals of deposition (black dots) of ^{210}Pb activity at RIVM in 2021 are shown with a 68% confidence interval (coloured bars).

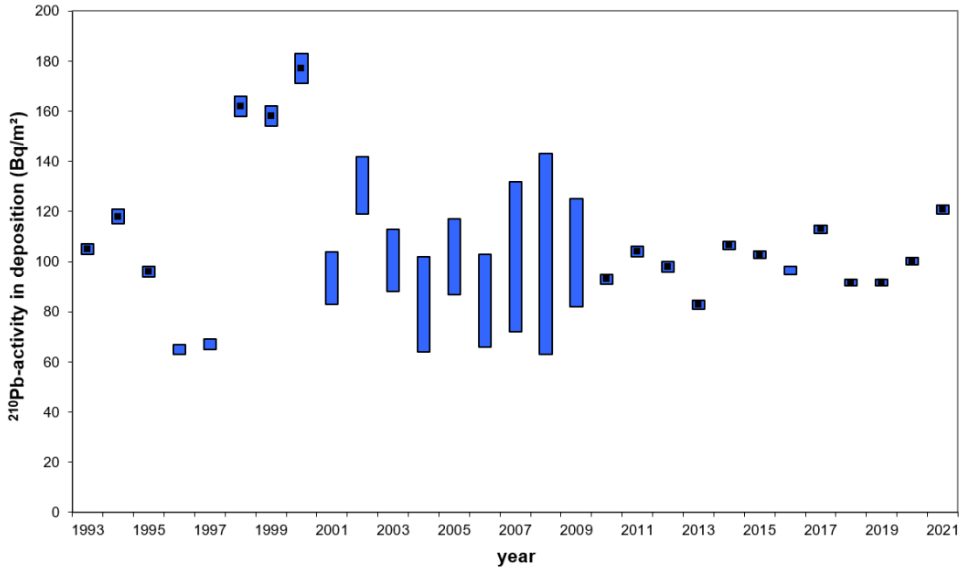

Figure 20 Yearly totals of ^{210}Pb activity deposited at RIVM (black dots) since 1993 shown with a 68% confidence interval (coloured bars). If the sum leading to the yearly total contains at least one detection limit, only the 68% confidence interval is shown.

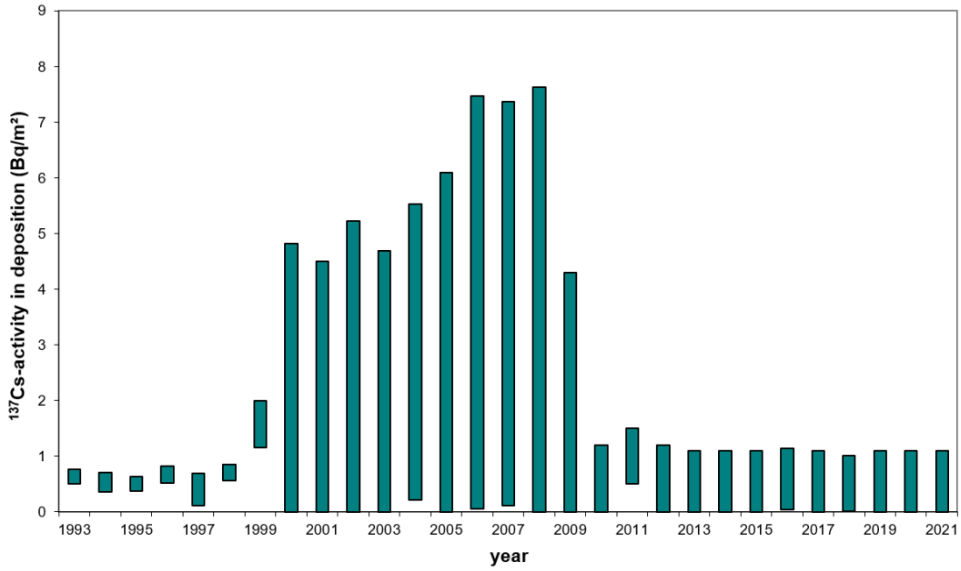

Figure 21 Yearly totals of ^{137}Cs activity deposited at RIVM since 1993. Yearly totals are shown solely as a 68% confidence interval since the sum leading to the yearly total contains at least one detection limit.

Appendix: Presentation of data

The methods described below were applied by RIVM to the data of air dust and deposition.

Correction for radioactive decay

In general, the activities of specific radionuclides are corrected for radioactive decay. The activities measured in the sample are multiplied by a decay factor, combining the time from halfway through the sampling period to the time of analysis, the decay during the measurement and the half-life of the radionuclide. If the radionuclides are unknown, as with gross α and gross β, no correction for radioactive decay is made.

Calculation of sums and averages

In the calculation of weekly, monthly or yearly averages or sums, the original results before rounding-off are used. If a certain radionuclide was not detected, the detection limit is used in the calculation of the sums. In that case, only a range (lower and upper limit) is given instead of a total with an uncertainty. Both range and total with an uncertainty are presented with a 68% confidence interval.

The lower and upper limits are calculated as follows:

$$\text{Lower limit} = \sum x_i - \sqrt{\sum s_i^2}$$

$$\text{Upper limit} = \sum x_i + \sqrt{\sum s_i^2} + \sum MDA_j$$

where

x_i = weekly or monthly result that is not a detection limit;

$\sqrt{\sum s_i^2}$ = the uncertainty in the sum;

s_i = uncertainty in the weekly or monthly result (1σ);

MDA_j = weekly or monthly result that is a detection limit.

The detection limits are omitted in the calculation of the averages. If data are not reported (e.g. a sample was not analysed/available), the sampling period is not taken into account for the calculation of the sum or average.

Calculation of uncertainties

The uncertainties of the individual results are a combination of the statistical uncertainties and the estimations of the experimental uncertainties. In the yearly total, the uncertainty is the square root of the sum of the squared weekly or monthly uncertainties. In the yearly average, the uncertainty is the square root of the sum of the squared weekly uncertainties divided by the number of weeks.

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