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On the Spring Maximum of Radioactive Fallout from Nuclear Test Explosions

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Abstract. Some results of measurements are presented pertaining to the maximum of fallout which was observed during the spring of the last few years. Evidence is presented that the height of this maximum is chiefly determined by the amount of radioactive debris present in the temperate or polar lower stratosphere, whereas seasonal factors determine the time of appearance of the maximum.

Introduction. During the last few years a seasonal variation in the concentration of long-lived fission products in air and in rain water, exhibiting a maximum in the spring and a minimum in the autumn, has been observed in several countries of the northern hemisphere [*Stewart, Osmond, Crooks, and Fisher,* 1957; *Stewart, Osmond, Crooks, Fisher, and Owers,* 1959; *Crooks, Osmond, Owers, and Fisher,* 1959; *Pierson, Crooks, and Fisher,* 1960*a*; *HASL-42, 51, 65, 69, 77, 84,* and *88*; *Lockhart, Baus, Patterson, and Saunders,* 1960*a*; *Aarkrog and Lippert,* 1959, 1960; *Bergh, Finstad, Lund, Michelsen, and Ottar,* 1959; *Edvarson and Löw,* 1960, *Schumann and Eulitz,* 1960]. Observations in the southern hemisphere suggest an analogous fluctuation, the amplitude being much smaller however [*Stewart and others,* 1959; *Crooks and others,* 1959; *Lockhart and others,* 1960*a*; *Achard, Beninson, and Migliori,* 1960].

Stewart and others [1957] and *Machta* [1958] attributed the spring maximum to an increased downward transport of air in the polar winter stratosphere. *Martell* [1959*a*, *b*] pointed out that the fallout during the spring might be due chiefly to Soviet nuclear tests in the autumn of the preceding year.

The hypothesis of Martell does not preclude the possibility of a 'real' seasonal effect causing the stratospheric radioactive dust from nuclear explosions at northern latitudes during the autumn to be fed into the troposphere, particularly during the spring of the next year. Similar tests carried out in July, for example, would then also give rise to a maximum of fallout in the spring of the next year. In other words, the height of the maximum is closely correlated with the amount of debris present in the stratosphere at higher latitudes, and the time of appearance depends on meteorological factors. Evidence for this view is presented in this paper.

Methods. The concentration of Sr^{89}, Sr^{90}, and Cs^{137} in monthly rain-water samples was determined by using the procedures of *Osmond, Owers, Healy, and Mead* [1959]. Monthly rain-water samples were obtained by exposing pots with a diameter of about 45 cm and a height of about 40 cm. Inactive carriers were added before exposure. The amount of rainfall during the collection period was measured with a rain gage which was emptied every 2 or 3 days.

The radioactivity of the isolated nuclides was measured by means of a Geiger-Müller tube (Philips type 18506) which had been calibrated with standardized solutions obtained from the Isotope Division, A.E.R.E., Harwell, England, or The Radiochemical Centre, Amersham, England.

All measurements were carried out in duplicate. In this paper average values are given. Duplicates usually differed less than 10 per cent.

Airborne radioactive particles were collected by drawing air through a membrane filter (Membranfiltergesellschaft, Göttingen, Germany) or a glass fiber filter (MSA 1106 B) with an effective diameter of 26 mm. The volume of processed air (40 to 60 m^3 day^{-1}) was measured with a gas meter connected at the outlet of the pump. Filters were changed daily. The radioactivity on the filters was measured with a Geiger-Müller tube (Philips type 18506) after decay of the natural radioactivity. The counting

setup was calibrated as accurately as possible. The geometrical efficiency was assessed by means of an absorption curve of a U_3O_8 sample of known weight. Self-absorption and absorption in the counter window (3 mg cm^{-2}) were estimated by calculation.

Because of the complex mixture of nuclides and the dependence of counting efficiency on the age of the fission products, an accuracy of better than 20 per cent cannot be claimed for the absolute values of the specific gross β radioactivity of air. Besides, daily figures for atmospheric radioactivity may deviate appreciably from the true mean value because particles with high radioactivity are sometimes present in such small numbers that a sample of 40 to 60 m^3 of air cannot be considered to be representative. Averages over more extended periods are, of course, much less affected.

Results and discussion. A seasonal variation in the concentration of various nuclides in rain water has also been observed at Rijswijk, Z.H., (52°3′N, 4°20′E). In Figure 1 the Cs^{137} concentration in monthly rain-water samples is plotted.

Abnormally low amounts of rainfall can result in high specific radioactivity of monthly samples due to the increased relative contribution of dry fallout. This was presumably the cause of the peak in September 1959. Rainfall during this month amounted to only 3.1 mm. The spring maxima however cannot be explained by rainfall peculiarities, as has already been shown by *Stewart and others* [1957] and follows also from the measurements of atmospheric radioactivity to be presented.

Radioactivity measurements during 1956 and 1959 support the view of Martell that most of the fallout during the spring is caused by Soviet tests at the end of the preceding year, the debris from these tests being stored in the stratosphere for a relatively short time only.

The observations of gross radioactivity in ground-level air during the first half of 1956 have been used in drawing the graph of Figure 2, which shows the specific atmospheric radioactivity at Rijswijk, Z.H., averaged over fortnightly periods and corrected for decay since November 23, 1955, the date of the last explosion of the Soviet tests in 1955 [*HASL-65*]. For this correction the decay formula $A = A_1 t^{-1.2}$ was used (t = days since explosion, A_1 = radioactivity at $t = 1$, A = radioactivity at time t). The choice of this date seems justified because age determinations by means of decay measurements pointed to the end of 1955 as the time of explosion, except during two periods of younger fallout indicated in the graph. The gross atmospheric radioactivity in the spring of 1956 was therefore mainly due to Soviet tests in November 1955. The contribution of these tests to the long-lived fraction of the fallout could not be

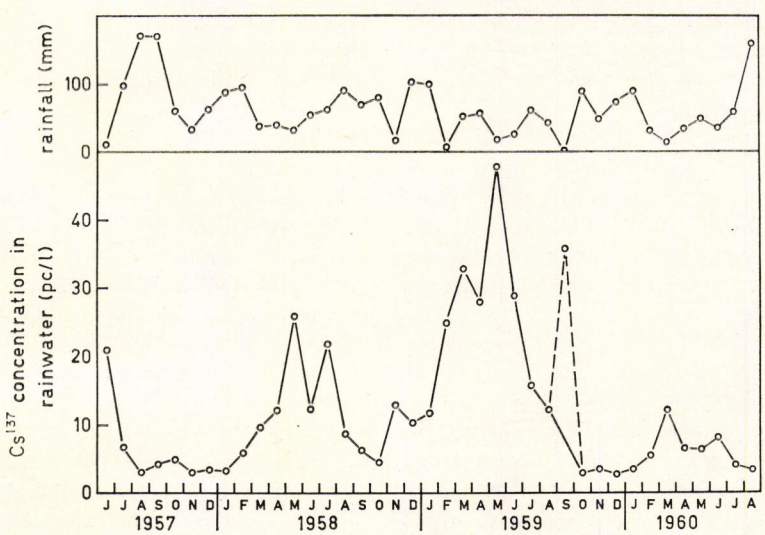

Fig. 1. Specific Cs^{137} activity in rain water and monthly amounts of rainfall at Rijswijk, Z.H., during the period June 1957 to August 1960 (1 pc/l = 10^{-12} curie per liter).

assessed. Data on the radiochemical composition of the fallout in the spring of 1956 are limited [*Stewart and others*, 1957; *Collins and Hallden*, 1958], and the exact times and yields of the Soviet explosions in 1955 are not known.

It is evident, however, that these tests brought about a rather heavy fallout, particularly during the spring of 1956. At that time we explained this wrongly by assuming a very large yield for these tests because *Stewart and others* [1955] had found that only about 12 per cent per year of the radioactivity in the stratosphere entered the troposphere. The yield being about 2 megatons, however [*Libby*, 1959], the observations provide evidence for a rather short storage time in the stratosphere for these tests.

After the Soviet tests in October 1958, the same type of curve was found for the air radioactivity. The specific air radioactivity corrected for decay since October 15, 1958, is shown in Figure 3. The radioactivity during the first half of 1959 was 1 order of magnitude higher than that in 1956, as was the yield of the October tests in 1958 as compared with the November tests in 1955 [*Libby*, 1959].

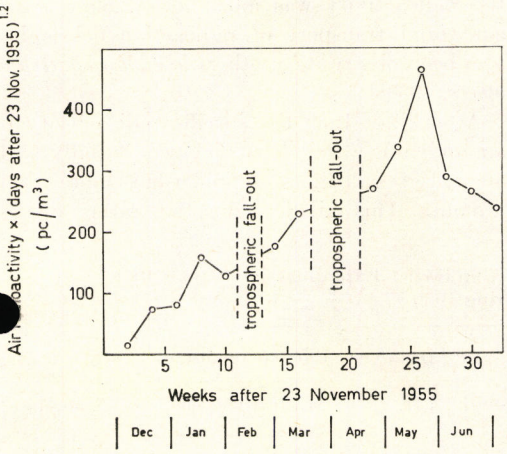

Fig. 2. Specific gross β radioactivity of fission products in air at Rijswijk, Z.H., (52°3′N, 4°20′E) during the period December 1955 to June 1956, corrected for decay since November 23, 1955. Every point represents the average over 14 consecutive days. During the periods indicated (between the dashed lines) the rate of decay of air-filter activity showed the presence of younger (presumably tropospheric) fallout. The measurements during these periods have been omitted from the graph (1 pc/m³ = 10^{-12} curie per m³).

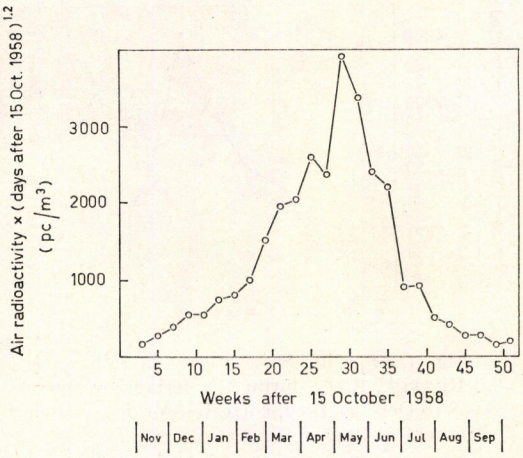

Fig. 3. Specific gross β radioactivity of fission products in air in The Netherlands during the period November 1958 to October 1959, corrected for decay since October 15, 1958. Every point represents the average over 14 consecutive days and over 4 sampling stations: Rijswijk, Z. H., (52°3′N, 4°20′E), De Bilt (52°6′N, 5°11′E), Den Helder (52°58′N, 4°45′E), and Eindhoven (51°26′N, 5°30′E). The three latter stations were operated by the Royal Netherlands Meteorological Institute.

The pattern of the radioactivity of rain water due to the October tests was similar. This is apparent from Figure 4, where the concentration of Sr^{89} in monthly rain-water samples corrected for decay since October 15, 1958, is plotted for the period November 1958 to October 1959.

From decay measurements of gross β radioactivity it follows that older fission products could account for only a small fraction of the total radioactivity during 1959. Even the long-lived fission products originated, for the greater part, from the explosions in October 1958. This is shown in Table 1, where the percentage of Sr^{90} from the latter tests is given, assuming that essentially all the Sr^{89} originated from these tests and that the Sr^{89}/Sr^{90} ratio was 180 at the time of explosion (October 15, 1958). The results are in good agreement with the values that can be computed from the Sr^{89}/Sr^{90} ratios observed at Milford Haven and Abingdon [*Crooks and others*, 1959]. In Table 1 these figures are also given for comparison.

At the time of the maximum, 80 to 90 per cent of the total Sr^{90} appears to have originated from the Soviet tests. The average over the first half of 1959 is 79 per cent at Rijswijk, Z.H.

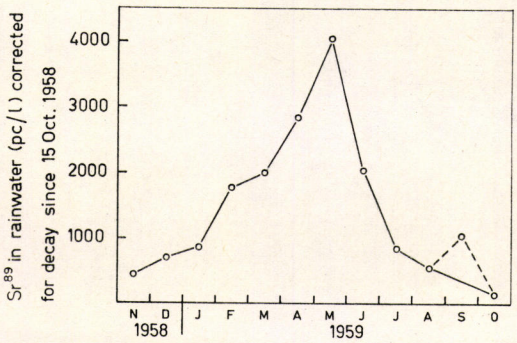

Fig. 4. Specific Sr^{89} activity in rain water at Rijswijk, Z.H., during the period November 1958 to October 1959, corrected for decay since October 15, 1958.

This is higher than the figures of 40 to 50 per cent and 60 per cent which have been computed by *Ambrosen* [1960] and by *Peirson and others* [1960*b*], respectively. The differences from the British results are not due to experimental errors (Table 1) but are probably caused by a different way of computation.

The percentages given in Table 1 might be somewhat high because the amount of Sr^{89} from test explosions before October 1958 has been neglected. *Lockhart and others* [1960*b*] estimate the contribution of Sr^{90} from the Hardtack detonations to the total Sr^{90} fallout in the northern hemisphere during the first half of 1959 to be about 8 per cent. If the average time of explosion is assumed to be July 15, 1958, and the Sr^{89}/Sr^{90} ratio of Hardtack debris at that time to be 180, it can be calculated that the error in attributing the Sr^{89} after January 1, 1959, to the Soviet October series amounts to about 3 per cent. The contribution of Sr^{89} from tests prior to the Hardtack series is probably negligible.

If the amount of Sr^{90} associated with Sr^{89} is subtracted from the total amount of Sr^{90}, a spring maximum in fallout not due to the Soviet tests during the autumn of 1958 is found. The relatively small difference between the two quantities of Sr^{90} is, however, rather sensitive to errors in the determination of Sr^{89} and Sr^{90}. The reality of the maximum is therefore uncertain.

The high concentration of gross atmospheric radioactivity during early 1959 has also been observed at stations at tropical latitudes. The bulk of the radioactivity originated from the tests in October 1958, as could be ascertained from the rate of decay. Figure 5 shows the specific air radioactivity corrected for decay since October 15, 1958, for Curaçao, Paramaribo, and Hollandia (stations set up by the Royal Netherlands Meteorological Institute).

Both stations north of the equator registered a high radioactivity during the spring of 1959. At Hollandia, a few degrees south of the equator, the radioactivity was much lower. Slow trans-equatorial transport of radioactive debris has also been observed by others [e.g., *Lockhart and others*, 1960*b*].

After May 1959 the specific radioacitvity of air and rain water decreased very rapidly with an initial half-time of somewhat more than 1 month. This might be due to a rather sudden

TABLE 1. Percentage of Sr^{90} Originating from Soviet Explosions in October 1958 in Fallout during 1959

Month, 1959	Total Sr^{90}, pc/l	Sr^{89},* pc/l	Sr^{90} from Oct. 1958 (percentage of total)		
			Rijswijk, Z.H.	Milford Haven	Abingdon
January	6.9	247	70	69	49
February	13.6	324	72	69	66
March	16.3	250	68	70	79
April	17.6	234	90	87	80
May	25.5	220	89	82	85
June	13.5	74	86	72	84
July	6.8	24	84		72
August	4.5	8.7	70		
September	7.2	10.6	82		
October	1.28	1.0	66		

* At the middle of the month, 1 pc/l = 10^{-12} curie per liter.

decrease in the rate of input of radioactive dust from the stratosphere, after which the radioactivity in the troposphere diminished with the half-time characteristic for the elimination of small particles from this part of the atmosphere.

The possibility that the fallout during the first half of 1959 consisted mainly of bigger particles with a significant falling velocity determining their residence time in the stratosphere must be excluded. During the period concerned all filters from one of the stations in The Netherlands were autoradiographed by exposing a sensitive X-ray film for 3 weeks.

If the radioactivity carried by debris particles from a particular detonation increases with size of particles, the 'hot spots' found on the autoradiographs will be associated with relatively big particles. Comparison of the radioactivity of filters producing autoradiographs with about the same uniform density (measured with a densitometer) but with greatly different numbers of hot spots showed the contribution of the bigger particles to the total radioactivity to be small.

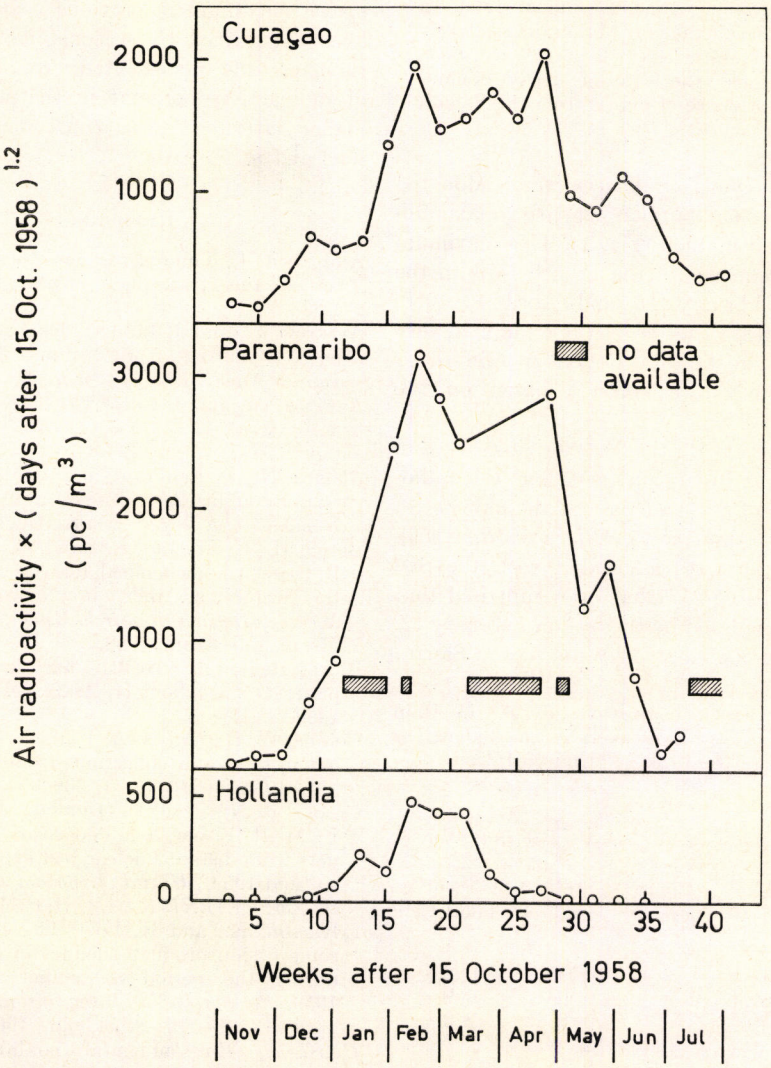

Fig. 5. Specific gross β radioactivity of fission products in air at Curaçao (12°10′N, 69°0′W), Paramaribo (5°50′N, 55°10′W), and Hollandia (2°25′S, 140°30′E) during the period November 1958 to July 1959, corrected for decay since October 15, 1958.

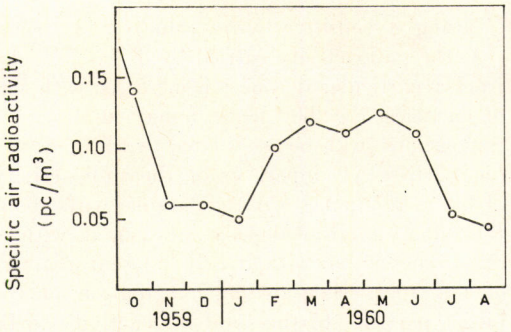

Fig. 6. Specific gross β radioactivity of fission products in air at Rijswijk, Z. H., during the period October 1959 to August 1960. In calculating the average for March the relatively high value of March 1 (1.12 pc/m³) has been omitted because the rate of decay demonstrated the presence of debris from the Sahara test on February 13.

Although no nuclear devices were detonated during 1959, a small but distinct spring maximum has been found in 1960 (Fig. 1). This maximum is not a consequence of the French tests in the Sahara on February 13 and April 1, 1960. Cs^{137} from these tests constituted only a very small fraction of the total present in rain water as follows from a calculation based on the Sr^{89} concentration.

Assuming that all the Sr^{89} originated from fission of Pu^{239} by fission neutrons, the radioactivity ratio of Sr^{89} and Cs^{137} at the moment of explosion was about 45 [*Björnerstedt*, 1959]. The amount of Cs^{137} associated with the Sr^{89} is shown in Table 2. It has been supposed that all Sr^{89} was generated by the test on February 13, nothing being contributed by the second Sahara test. This results in an overestimation of the amount of Cs^{137} associated with Sr^{89}. It is evident that the French tests can account for at most 2.3 per cent of the total Cs^{137} concentration.

TABLE 2. The Concentration of Cs^{137} in Rain Water Due to Test Explosions in the Sahara as Calculated from the Sr^{89} Concentration

Sampling Period 1960	Cs^{137} Conc., pc/l	Sr^{89} Conc.,* pc/l	Cs^{137} Associated with Sr^{89}	
			In pc/l	In Percentage of Total Cs^{137}
February	5.5	2.4	0.05	0.9
March	12.3	3.1	0.11	0.9
April	6.6	6.7	0.15	2.3
May	6.4	0†	...	...

* At the middle of the month.
† No Sr^{89} detected.

The specific radioactivity of the air also exhibited a maximum, as is shown in Figure 6. It is very improbable, therefore, that the Cs^{137} maximum is brought about by rainfall characteristics, although the peak in March may be partly due to light rainfall (Fig. 1).

It seems, therefore, that the maximum in 1960 was a real seasonal effect. This supports the view that the time of appearance of the spring maximum in previous years has likewise been determined by seasonal factors, although its magnitude was essentially due to the amount of radioactivity present in the stratosphere at higher latitudes as a consequence of recent nuclear tests.

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