Changes in the Radioactivity Regime during the Passage of a Cold Front over The Netherlands

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Abstract

Artificial radioactivity in rainwater was measured during the passage of a cold front over The Netherlands. The specific radioactivity of the frontal rain increased considerably after the frontal veering of the wind. Concurrently the "age" of the fission products decreased from 45 to 17 days, *indicating a supply of atomic debris from recent nuclear explosions in arctic regions.* The specific radioactivity of the air increased considerably on the day of the frontal passage. Trajectories of pre-frontal and post-frontal air indicate that the pre-frontal air comes from a source region in the Mediterranean and the post-frontal air from the arctic regions. The diagram of the frontal passage is analysed.

Introduction

In a recent paper (BLEICHRODT, BLOK, DEKKER and LOCK, 1959) data showing a relation between the amount of artificial radioactivity in rain and rain intensity have been presented. As a rule the specific radioactivity of rain was high during periods of low rain intensity and conversely. It is of importance to mention that only showers or showery precipitation occurring in unstable air masses were described.

The research reported in the present paper refers to the changes in specific artificial radioactivity of rain and air during the passage of a front over The Netherlands.

The meteorological situation

On the morning of 31 October 1958, a cold front approached the Netherlands coast from the west. This front separated two distinct air masses, namely modified tropical air in the east and relatively fresh arctic or polar air in the west. The winds in the tropical air were weak; the air mass was rather stable with a high

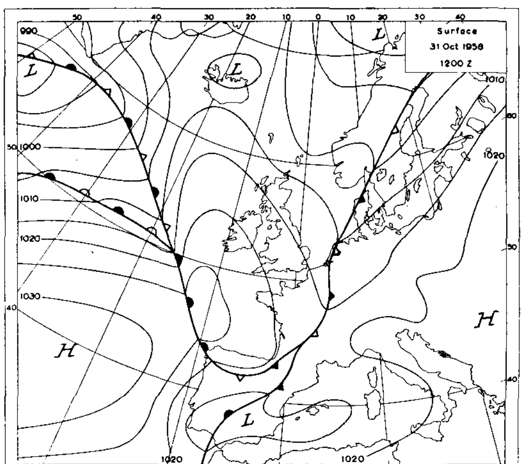

Fig. 1. Synoptic surface map of 31 October 1958, 12.00 GMT.

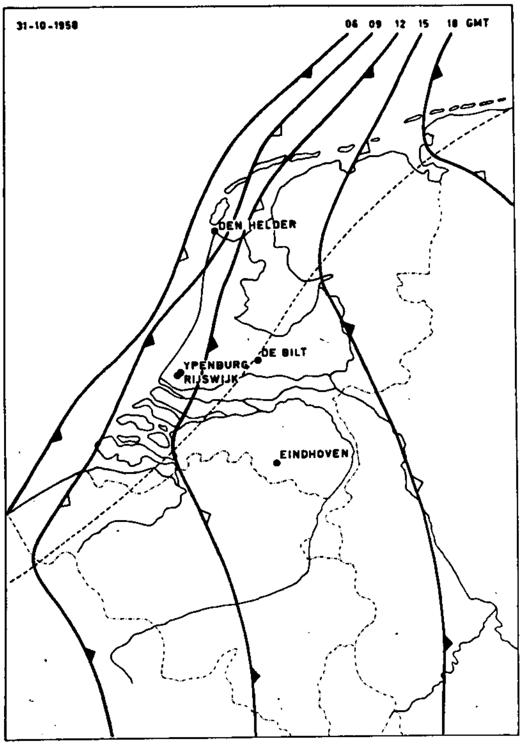

Fig. 2. Isochrones of cold front between 00.00 GMT and 18.00 GMT (31 October 1958); positions of stations.

humidity in the lower layers (fog over large areas). The wind speed was much higher in the polar air, which also was rather unstable and in which especially over the sea shower activity was observed.

Figure 1 shows the synoptic surface map of 12.00 GMT.

Figure 2 gives the successive positions of the cold front over The Netherlands and adjacent areas between 06.00 GMT and 18.00 GMT. It appears that a wave developed in the cold front. The top of the wave moved from SW to NE over the middle of the country; the

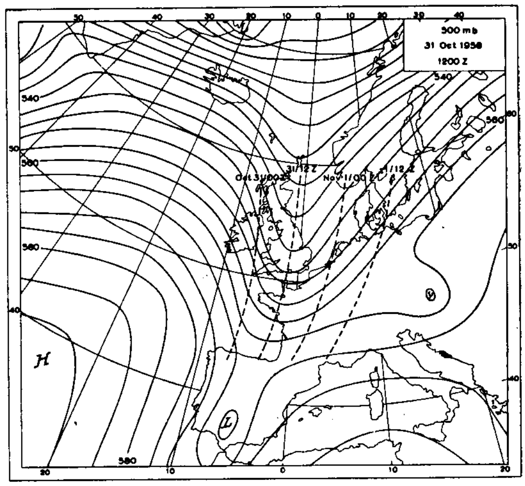

Fig. 3. Absolute topography of 500 mb level of 31 October 1958, 12.00 GMT.

heaviest rains (20 mm or more) were observed just NW of the wave top.

Figure 3 demonstrates the synoptic situation at the 500 mb level, again for 31 October 12.00 GMT. It appears that the surface front is followed by a deep post-frontal trough in the cold air, the successive positions of which are also indicated.

The differences in structure between the tropical and polar air can best be appreciated from table I which gives the temperatures at various levels over De Bilt for 31 October 00.00 and 12.00 GMT and for 1 November 00.00 GMT.

In figure 4 the recordings and observations of various meteorological elements during the frontal passage, at the stations Ypenburg[1] and Rijswijk[1] are reproduced. Visibility (a), relative humidity (b) and temperature (c) were observed at regular intervals at Ypenburg; pressure

[1] The position of the various stations mentioned in this paper is given in figure 2.

Table 1. Upper air temperatures (°C).

Time \ Pressure	1000	850	700	500	400	300	200	100	Tropopause
31 Oct. 00.00 GMT	6	2	— 6	— 21	— 32	— 47	— 58	— 60	— 60 (230 mb)
31 Oct. 12.00 GMT	8	2	— 6	— 22	— 34	— 46	— 52	— 54	— 55 (215 mb)
1 Nov. 00.00 GMT	6	— 2	— 12	— 30	— 40	— 47	— 51	— 57	— 48 (330 mb)

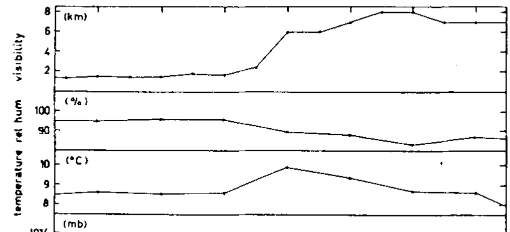

Fig. 4. Diagram of meteorological elements and radioactivity observations at Ypenburg and Rijswijk (31 October 1958). a. visibility; b. relative humidity; c. temperature; d. pressure; e. wind speed; f. wind direction; g. rain intensity; h. radioactivity; i. "age" of fission products.

(d), wind speed (e) and wind direction (f) were recorded at the same station. The rain intensity (g) was observed at Rijswijk. Ypenburg and Rijswijk lie at a distance of 1,600 m. The front arrived at Rijswijk approximately 3 minutes earlier than at Ypenburg. This time difference may be considered negligible in the interpretation of the data.

The frontal passage is indicated by a veering of the wind from southwest to northnorthwest, which took place between 11.00 and 11.30 GMT. During this period high rain intensities were observed. Immediately after the veering the wind speed increased considerably.

Radioactivity in rainwater

Rainwater samples were taken at Rijswijk between 8.20 and 15.20 GMT. The rainwater was caught in a galvanized tray of 5 m^2 which was placed with a slight slope on the flat roof of the Medical Biological Laboratory. A measuring glass was placed under an outlet-pipe at the lowest point of the tray. Every time when about half a liter of rainwater had been collected in the glass, the water film in the tray was quickly swept down with a screen wiper. The total volume of the samples, usually about 1 liter, and the time during which they were obtained were listed. From these data average rain intensities for relatively short periods could be computed. The results are given in figure 4 (g), in which the dots indicate the time of the middle of the sampling period.

Before sampling of rainwater was started the tray had been thoroughly cleaned with tap water in order to remove previously deposited fall-out material.

The artificial radioactivity of the rainwater samples was determined as follows. The bulk of the beta activity was concentrated by stirring the water with 4 g of active coal (Norit C, Norit Verkoopmaatschappij, Amsterdam), which was then filtered off. The filtrate was evaporated to dryness and the residue together with the coal containing filter ashed at 500° C. The ash was weighed and a fixed amount was put in a brass sample cup.

Seven days after collecting the rainwater, the beta radioactivity of the samples was measured (standard error $< 2.5\%$) using an end-window Geiger-Müller tube (Philips 18506) with a diameter of 3 cm and a window thickness of 3 mg/cm^2. The beta radioactivity per liter of rainwater was then computed from the measured activity, the sample weight, the total ash weight and the volume of water collected. The result is shown in figure 4 (h) (dotted radioactivity curve). One radioactivity unit corresponds with about 5 pc/liter.

The beta radioactivity of nine ash portions was measured in duplicate to investigate the possible influence on the changes in specific radioactivity of highly radioactive particles which can be found shortly after nuclear explosions. As the differences between the duplicate measurements proved to be smaller than 10%, it seems justified to exclude this influence.

The beta radioactivity of ten samples was measured periodically during three months. From the decay rate the "age" of the fission

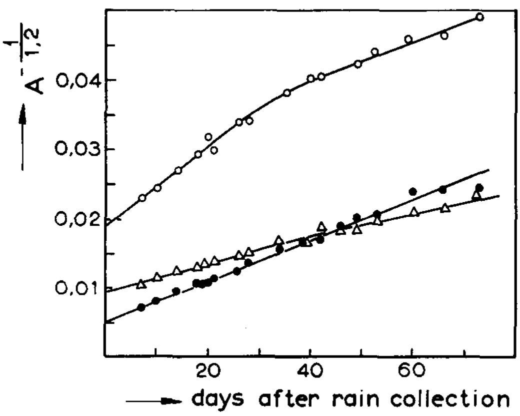

Fig. 5. Decay curves of debris collected at 9.10 (△), 11.10 (o) and 14.40 GMT (●).

products was determined, using the approximate relation:

$$A = A_1 \, t^{-1.2} \qquad (1)$$

where A is the activity at time t and A_1 the activity at $t = 1$. To this end the values of $A^{-1/1.2}$ were plotted versus t and the best fitting curve was extrapolated to $A^{-1/1.2} = 0$. In this way an indication was obtained of the moment when the artificial radioactivity was introduced into the atmosphere.

The decay curves often consist of a combination of two straight line segments with a slightly bent part in between. In the beginning the decay is determined mainly by the relatively fresh fission products. After a few weeks the influence of the older debris becomes noticeable. As example three decay curves are shown in figure 5.

The result of the "age" determinations is plotted in figure 4 (i).

With the aid of the obtained "age"-curve it is possible to calculate the values of the specific radioactivity of the rainwater at the date of sampling. The drawn curve of figure 4 (h) gives the so obtained values of the specific radioactivity at sampling time.

The radioactivity data show the following features. It is obvious that the specific radioactivity of the rain was generally low before the frontal passage with the exception of an isolated peak at about 9.10 GMT. There is a tendency of the radioactivity to decrease with the approaching of the front. During the veering of the wind the activity increases about 50 % and after some variations, in which it occasionally drops to the pre-frontal level, a marked increase to a value approximately 5 times the pre-frontal value starts at 12.45 GMT. It is furthermore noted that neither in the pre-frontal rain nor in the post-frontal rain there is a clear indication of an inverse relationship between rain intensity and radioactivity.

The "age"-curve of figure 4 (i) shows that the "age" of the fission products in the pre-frontal rain was about 45 days and about 17 days in the post-frontal rain. The pre-frontal as well as the post-frontal "age" values are sufficiently uniform and the difference between the two is sufficiently large to warrant the conclusion that the nature of the artificial radioactive material changed completely during the frontal passage.

It should be noted here that the above-mentioned high pre-frontal specific radioactivity which was found at 9.10 GMT will not be related to the high post-frontal values, as exactly for this sample an "age" determination has been made (43 days); this pre-frontal peak must therefore be due to unknown processes. The presence of a very active particle must be considered improbable, as a duplicate measurement showed the same high value of the activity.

Airborne radioactivity

Airborne radioactivity was sampled at ground level at the stations Den Helder, De Bilt, Rijswijk and Eindhoven (for positions cf. figure 2).

By means of a vacuum pump 30 to 50 m^3 (measured with a gasmeter) of air is drawn through a membrane filter (Membranfilter Gesellschaft, Göttingen, Germany) in 24 hours. The effective surface of the filter is 5.2 cm^2. The filters are changed daily at 8.00 GMT. In order to avoid loss of radioactive particles the dust is fixed to the filter paper by treating the filter with a solution of formvar in 1,2-dichloro-ethane (20 g per liter).

The beta activity of the filters was measured at the Medical Biological Laboratory by means of an end-window Geiger-Müller tube (Philips 18506) three days after sampling to allow the natural radioactivity to decay.

In table 2 the airborne artificial radioactivity as measured during the period October 26th—November 5th 1958 is listed.

Table 2. Airborne artificial radioactivity in pc/m³.

Sampling period	Stations				Average
	Den Helder	De Bilt	Rijswijk (Z.H.)	Eindhoven	
October 26/27 ...	1.02	0.47	0.94	0.66	0.77
» 27/28...	1.15	1.06	1.13	0.58	0.98
» 28/29...	1.15	0.95	0.92	0.91	0.98
» 29/30...	0.74	0.81	0.54	0.52	0.65
» 30/31...	1.29	1.36	1.48	1.42	1.39
» 31/Nov.1	2.98	2.59	3.65	2.79	3.00
November 1/2...	2.57	2.11	—	2.16	2.28
» 2/3...	2.18	2.49	[1] —	1.69	2.12
» 3/4...	5.44	5.19	[2] 23.7	5.43	[3] 5.35
» 4/5...	4.51	2.51	2.97	1.90	2.97

[1] No data available.
[2] High value probably due to one or more very active particles.
[3] Value of Rijswijk (Z. H.) excluded.

It appears that the radioactivity was relatively low until the morning of 30 October. The samples collected on the morning of the 31st show an increase of 0.7 pc/m³ in the average. The "age" of the samples collected on 31 October was not determined. It is however highly probable that the radioactivity in the pre-frontal tropical air was the same as that in the pre-frontal rain. In that case the rise on 31 October must be due to processes in the tropical air, of which pre-frontal subsidence or changes in the turbulence conditions could be mentioned.

A strong increase of 1.6 pc/m³ in the average is observed on 1 November, i.e. after the frontal passage. The high activity of the post-frontal rain and the high activity of the post-frontal polar air go together. The highest increase is observed at Rijswijk. The "age" of the airborne radioactivity collected during 31 October—1 November at Rijswijk was 20.5 days. This agrees fairly well with that of the post-frontal rainwater.

It is well-known that the specific artificial radioactivity of the air usually increases with height. The question may be asked whether the radioactivity at the surface increased so strongly solely as a result of a more intense turbulent diffusion due to a higher lapse rate, higher wind speeds and greater vertical wind-shear, or whether the radioactivity on the whole was much higher in the polar air than in the tropical air. In this connection it is important to note that the specific radioactivity of the surface air increased by a factor 2 approximately and that of the rain by a factor 5.

Now the process of rain formation takes place in a thick layer and the specific radioactivity of the rain can, therefore, be considered more representative for the air mass as a whole than the specific radioactivity of surface air. Consequently it may be concluded that the radioactivity in the tropospheric layers above the planetary friction layer in the polar air mass was approximately 5 times higher than in the tropical air, but that in spite of the greater turbulent diffusion the downward transport of radioactive matter was not strong enough to bring about also a fivefold rise in the airborne radioactivity at ground level. This seems to indicate that high values of radioactivity in the polar air were present in the middle of the troposphere and above.

A new rise in the airborne radioactivity was observed during the period 3—4 November. A front with a new outbreak of arctic air in which the wind veered to a northerly direction passed The Netherlands in the afternoon of 3 November 1958. The high value measured at Rijswijk is probably due to one or more very active particles.

Air trajectories

The explanation of the change in the radioactivity regime on 31 October is easily found when the trajectories of the pre-frontal and post-frontal air are investigated. Under the assumption of horizontal motion trajectories have been computed for an air mass present over The Netherlands on 31 October 00.00 GMT (representing the tropical air) and an air mass which covered The Netherlands on 1 November 00.00 GMT (representing the polar air). Fig. 6, 7, 8 and 9 show these trajectories for the 900 mb level (based on gradient winds taken from the surface map) and for the 700, 500, and 300 mb levels respectively.

It appears that up to 500 mb the pre-frontal air originated in the Mediterranean area. The wind speed decreased with increasing altitude, as usually is the case in a predominantly easterly current. The air parcels at 300 mb originated from the North Atlantic.

The post-frontal air came from the arctic

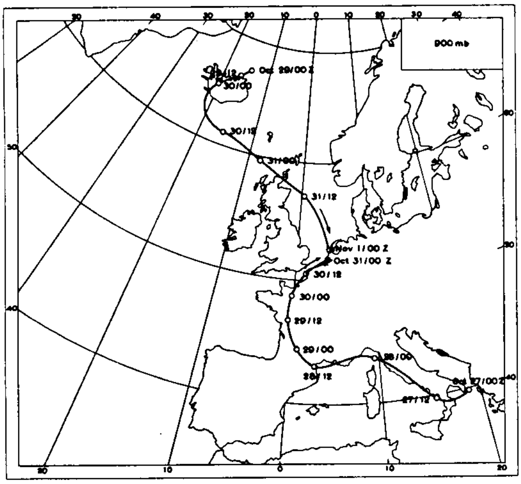

Fig. 6. Air trajectories at 900 mb level.

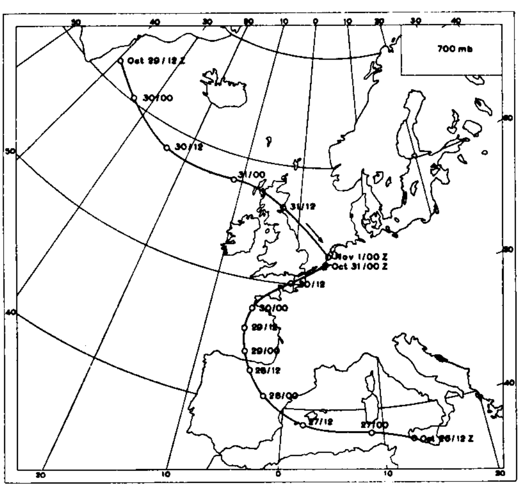

Fig. 7. Air trajectories at 700 mb level.

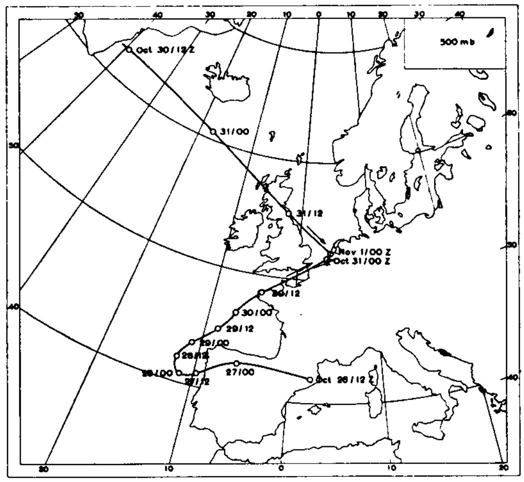

Fig. 8. Air trajectories at 500 mb level.

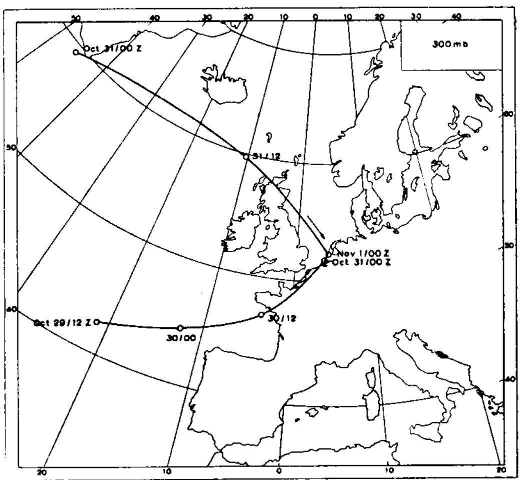

Fig. 9. Air trajectories at 300 mb level.

regions in all levels. The wind speeds increased rapidly with altitude, so that a relatively strong vertical windshear existed.

According to information published by the United States Atomic Energy Commission (HARDY, E. P. and KLEIN, S., 1959) nuclear test explosions were performed north of the arctic circle on 10, 12, 15, 18, 19, 20, 22, 24 and 25 October. The radioactivity found in the polar air is undoubtedly due to one or more of these test explosions. Many more data of radioactivity measurements would be necessary in order to determine at which levels the transport of the radioactive material took place.

It is impossible to state where the atomic debris present in the tropical air was injected into the atmosphere.

Further analysis of the data collected during the frontal passage

Figure 4 raises a number of problems related to cloud physics and to the structure of fronts. For the time being the authors limit themselves to point out that apparently 5 phases can clearly be distinguished in the diagram.

Phase a, from the beginning to 11.00 GMT.

Pre-frontal tropical air with uniform temperature, uniform humidity and low uniform visibility is present over the station. The rain intensity varies, there is a light pressure fall and slowly veering wind of low speed. The radioactivity decreases slowly; the "age" of the radioactive material is constant (approximately 47 days).

Phase b, from 11.00 to 12.00 GMT. This phase is marked by a strong veering of the wind at the beginning of the period and of a considerable stepwise increase of the wind speed at the end of the period. The rain intensity increases to a high value in the beginning of the period and decreases to its original value at the end. The visibility increases considerably. At the end of the period the temperature is slightly higher than at the beginning and the humidity has decreased. There is still a slight pressure fall. The radioactivity increases about 50%; the "age" indicates the presence of a mixture of old and fresh debris.

Phase c, from 12.00 to 12.45 GMT. This is a phase, which is characterized by an approximately constant radioactivity level and a low "age" (18 days). Wind direction and speed are constant and the rain intensity shows a peak. At the beginning of the period the pressure starts to rise. The visibility is high.

Phase d, from 12.45 to 14.40 GMT. This phase is marked by a strong increase of radioactivity of low "age". The wind direction is constant. The wind speed is variable but drops to a lower value at the end of the period. Visibility is high, temperature and humidity have slightly decreased and the pressure rises.

Phase e, from 14.40—end. The radioactivity is at a high level. The wind direction and speed are constant. The pressure rises further while temperature falls and humidity rises apparently under influence of the cooling with the approaching setting of the sun.

Acknowledgement

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