Advection over Sweden of Radioactive Dust from the First French nuclear Test Explosion

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Abstract

An examination of the occurrence over Sweden of radioactive debris from the first French nuclear test in February 1960 has been undertaken. The distribution of radioactive deposit was uneven. The time of arrival of debris can be determined accurately, and a comparison is made with meteorological parameters. Special autoradiographical and gammaspectrometrical examinations have been carried out, which suggest fractionation effects to be present in the scavenging processes.

Introduction

Since the summer of 1959 the amount of fission products in the atmosphere originating from nuclear test explosions have decreased to a very low level. This facilitates the identification of small amounts of newly introduced radioactivity. The first French nuclear test explosion in Sahara, which took place in the morning of February 13th 1960 and which according to press information was a "tower shot" with a yield of about 70 kilotons, thus gave an opportunity to study the effect of one single insertion of fission product radioactivity into the atmosphere. The data concerning the arrival of the French debris to Sweden have been summarized and may be of interest in the study of the mechanism of fallout. The observed fallout is very much less than during the spring of 1959, when debris from Russian high yield explosions were deposited.

Meteorological situation

During the last days of February 1960 the surface synoptic situation showed a well-developed and almost stationary cold anticyclonic circulation over Scandinavia (Figure 1). The centre of this anticyclone was somewhere to the north of Norway. A deep depression dominated the circulation west of the British Isles bringing warmer and moister air towards the western and northwestern regions of Europe. The upper air map displayed a ridge in an almost north-south direction across Western Europe (Figure 2). In conjunction with the surface depression a closed cyclonic circulation was situated west of the British Isles.

At the end of the month the surface anticyclone and upper ridge began to move slowly eastwards. Evidence of warm air advection over Southwestern Scandinavia was clearly indicated by substantial temperature increases in the troposphere as shown by the radiosonde ascends of Copenhagen and Gothenburg on March 1st 00 GMT.

A precipitation area which was associated with the warm frontal system between the airmasses came in over the southwestern part of Sweden at about March 1st 06 GMT. It passed over a greater part of the country, the

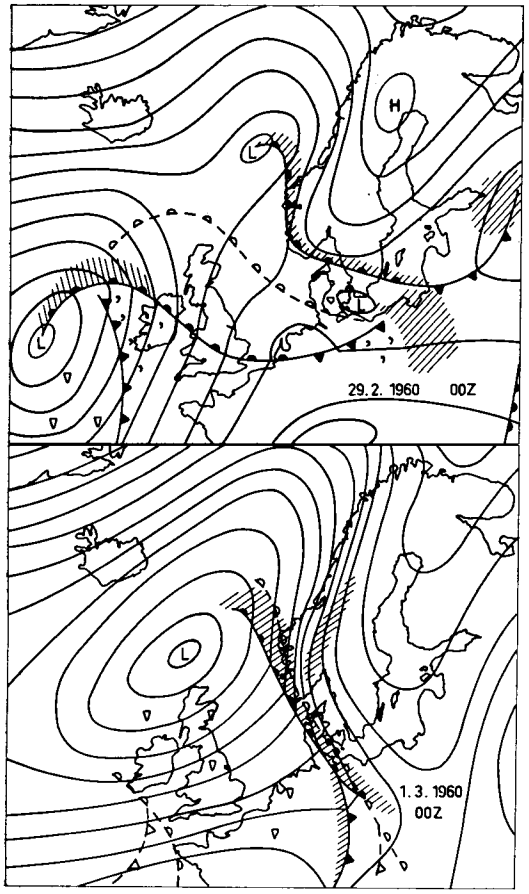

Fig. 1. Surface level weather maps. Dashed line indicates movement of front during next 12 hours.

precipitation falling initially in the form of freezing rain. Successive positions of the forward-edge of the precipitation area are shown in Figure 3.

Arrival and deposition of debris

The first trace of fresh radioactivity in the atmosphere was noticed in a precipitation sample from Gothenburg. This sample was collected with a large funnel (diameter 2 m), which was exposed for both dry deposition and precipitation for 24 hours. Only a few drops of the freezing rain had passed the ion-exchanger collecting unit when it was shifted on March 1st 06 GMT, and no other precipitation was reported during the collection period. Preliminary investigation showed fission product contents corresponding to a deposition about ten times as large as the maximum values obtained after periods with heavy precipitation during the last six months. The sampling period for the next filter was six days, and during the first half of this period 18 mm of rain was recorded. This sample was the most radioactive of all samples collected in Sweden since May 1959. After March 4th an almost dry period occurred over this part of Sweden, and no more radioactivity of recent origin was detected in the precipitation samples.

In ground level air samples from Gothenburg a comparatively smaller but identificable increase in fission product contents was reported for the period March 1st 00—12 GMT and was confirmed by an autoradiographical ex-

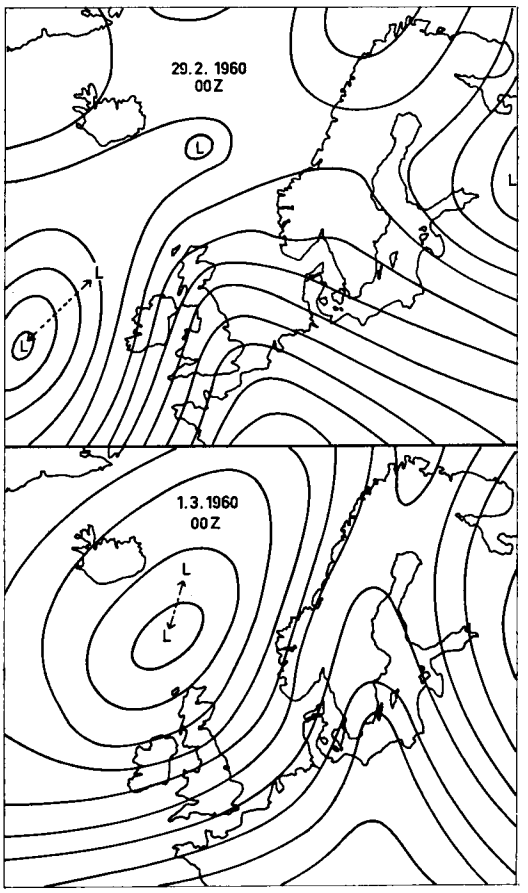

Fig. 2. 700 mb contours. Dashed line indicates movement of low pressure center during next 12 hours.

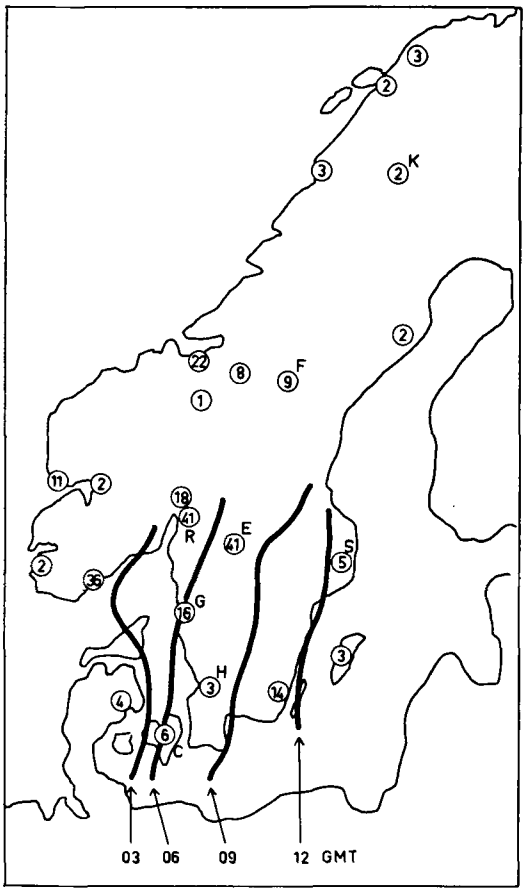

Fig. 3. Four successive positions (on March 1st) of the forward edge of the precipitation area. In the figure is also indicated the sampling stations and the preliminary total deposition values in mc km^{-2}. G = Gothenburg, E = Edsvalla, H = Högaryd, F = Östersund, K = Kiruna, R = Kjeller-Norway, C = Copenhagen-Denmark.

amination of the glass-fibre filters. No detectable amounts of fresh debris were observed before this period or during the four days following.

The positions of the Swedish and some Norwegian and Danish fallout collecting stations are indicated in Figure 3 together with total deposition values (mc km^{-2}) from the actual period corrected for decay to April 1st. The results from the Swedish measurements can be summarized as follows:

1. Radioactivity of an apparent age of about 20 days was deposited in Sweden during the passages of successives precipitation areas beginning March 1st.

2. In spite of the fact that there were no great differences in the measured (standard rain gauges) precipitation amounts in various parts of the country, the total deposition varied at least with a factor of 20. Practically no deposition of fresh debris was detected in samples from Kiruna in northern Sweden, whilst the highest values were observed in samples from Edsvalla and Gothenburg (cf. Figure 3).

3. Samples obtained from Gothenburg on March 1st showed a high concentration of radioactivity in the first few raindrops; at Edsvalla the larger part (75 %) of the total deposition was recorded during the first three hours of precipitation.

4. The increase of fission product radioactivity in ground level air was with the exception of Kiruna comparatively smaller than in precipitation. The first increase occurred in close connexion with the arrival over Sweden of the precipitation area on March 1st and was of short duration. It was followed by a low level period of activity until March 10th when a new increase occurred.

Analysis of individual samples

With the intention of studying the composition of the radioactivity a series of gamma-spectrometrical examinations was carried out with a 10 cm × 10 cm Na(Tl) I-crystal connected to a 70-channel kicksorter. The spectra of the most active precipitation samples showed a remarkable difference, indicating the occurrence of relatively strong fractionation effects in the scavenging processes. A further analysis therefore was undertaken with the aid of standard calibration spectra from reactor-produced isotopes of Te 132, Ce 141, I 131, Ru 103, Zr 95, and Ba-La 140. The contribution from each of these at the peaks of .15, .36, .50, .75, and 1.6 MeV was calculated, and fractionation factors were computed according to EDVARSON et al. (1959). The factors are defined in the following way:

$$f_{Ce\,141} = \frac{\left[\frac{Ce\,141}{(Zr+Nb)95}\right]_{experimental}}{\left[\frac{Ce\,141}{(Zr+Nb)95}\right]_{theoretical}}$$

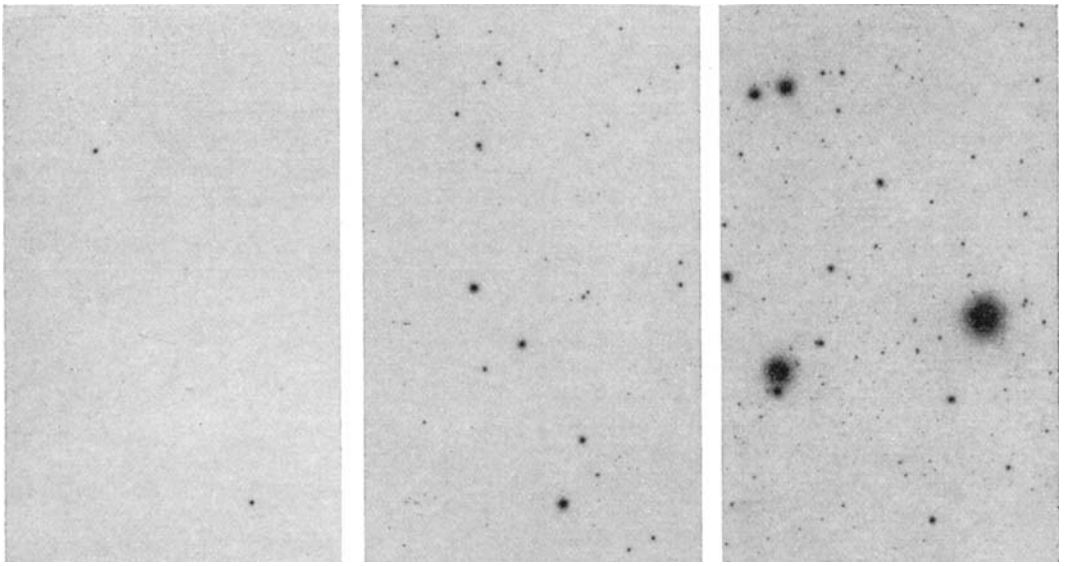

Fig. 4. Copies of autoradiographs from ground level air filter samples. Left: February 29th 1960 (30 days exposure). Middle: March 1st 1960 (10 days exposure). Right: November 16th 1958 (7 days exposure).

The result of the analysis (carried out by K Löw at this institute) is presented in Table 1. Yield values for fast fission of Uranium

Table 1

	Sample				
	Gothenburg		Stockholm		Edsvalla
	412	413	405	406	
$f_{Ce\ 141}$ ...	1.5	1.4	1.6	1.6	1.4
$f_{I\ 131}$	4.3	1.8	4.3	1.4	—
$f_{Ru\ 103}$ ...	5.5	1.3	2.2	0.8	1.4
$f_{Ba\ 140}$ ...	2.2	1.4	1.8	1.3	1.2
Sampling period ...	29/2—1/3	1/3—7/3	1/3—4/3	4/3—8/3	1/2—1/3

Fractionation factors from analysis of γ-spectra of precipitation samples normalized to 40 days after fission.

238 have been used. No significant difference would have occurred if instead values for fast fission of Plutonium 239 were applied.

The two samples from Gothenburg previously mentioned (called 412 and 413 respectively) especially seem to give interesting results. Sample 412 contained the activity from the very first few drops of the freezing rain of March 1st, while sample 413 would give a mean picture of the activity distribution after a considerably larger precipitation amount, either the activity came with the first millimetres or during the whole period. The excess of Ru 103 and I 131 seems to be significant in the first sample. Repeated gammaspectrometrical measurements have shown good agreements with the respective theoretical decay rates.

All precipitation samples were collected with the ion-exchanger method first proposed by EDVARSON (1957). The samples 412 and 413 therefore could be divided into three parts; one containing the plastic filter and the others containing the two ion-exchangers respectively. Each part was measured in the gammaspectrometer. It was found that a comparatively greater part of the total activity of sample 413 was concentrated on the plastic filter. This was especially the case for Ru 103, where only a minor part was found on the plastic filter of sample 412.

In order to make an autoradiographical examination of the particulate activity distribution possible, a careful ashing of the collecting parts had to be undertaken. The ash was spread out on a thin gummed paper and pressed together

with X-ray films on both sides. After exposure the spots of the films were measured and counted, and there appeared to be a clear tendency towards a relatively greater number of large spots on the filter parts of sample 412 when compared with sample 413.

When these findings are related to the total activity of the samples the conclusion seems to be that in the beginning of the precipitation there is an enrichment of particles smaller and larger than medium size.

From a study of the series of autoradiographs of air samples from Gothenburg it is evident that the sample from the period March 1st 00—12 GMT gave a sharp peak in the amount of active particulate matter in the lowest layer of the atmosphere. In Figure 4 copies of autoradiographs from February 29th and March 1st are reproduced together with one from November 16th, 1958, where the radioactivity originated from the Russian high yield test explosions at Novaja Zemlya during October 1958. From investigations by SISEFSKY (1960) the magnitude of the particle responsible for the largest spot of the older sample can be estimated to about 4μ (diameter).

Some high altitude air samples also have been taken during the period of arrival of the French debris, namely on February 24th, March 6th, and March 10th. On these occasions air has been collected above and below the tropopause, and all samples showed very weak radioactivity. The only conclusion to be drawn is that the contribution of fresh activity in these filters must have been very small.

Discussion

The available explosion data suggest that the top of the radioactive cloud from the first French explosion reached a height of about 13 km (200 mb). In an attempt to follow the wind transportation of the cloud one therefore has to have good information about winds in the upper troposphere (300 mb) during the first few days after the occurrence of the explosion. Then 500 mb-winds ought to give the best indication of the trajectory of the cloud. But even with access to all existing measurements of winds and temperatures in all levels in the Northern Hemisphere and a knowledge of time of arrival at the surface, it is hardly possible to reconstruct a trajectory for e.g. that part of the debris which was sampled in Sweden on March 1st. The uncertainties soon will become too great. In order to give a rough tentative idea of the magnitude of the air motions Figure 5 has been constructed on the basis of

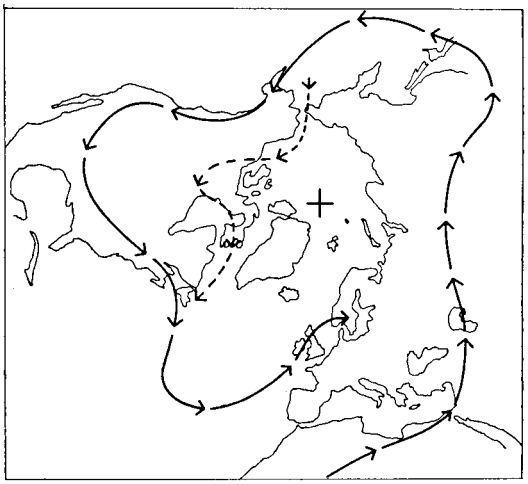

Figure 5. A tentative estimation of the magnitude of the 24 hour 500 mb air motions during February 13th to March 1st 1960. The resulting trajectory is of course subject to considerable uncertainty and applies only to that part of the debris which was sampled in Scandinavia. (Dotted line: alternative trajectory.)

information from the 500 mb contour maps and knowledge of the time of arrival of debris to Sweden.

Comparison with measurements from Norway and Denmark are of great interest. According to HVINDEN (1960) a peak in the radioactivity of the precipitation was measured during the period February 29th to March 1st at the Norwegian station Kjeller. This can be compared with the fact that Edsvalla and Gothenburg are the maximum-deposition stations in Sweden. These two stations are situated in the same part of Scandinavia as Kjeller. From Denmark results of precipitation measurements are available from two stations. The total deposition is smaller than at Kjeller, but the same time variation holds with a bigger part of the radioactivity in the samples of March 1st and a tendency to a drop in

specific and absolute activity during March 2nd.

The differences in the deposition pattern can be explained in many ways:

1. Uneven distribution of precipitation over the country.

2. Variations of the effectiveness of the samplers because of different sensitivity to wind direction and strength or different aggregate forms of the precipitation elements.

3. Differences in the scavenging effectiveness of different aggregate forms of the precipitation elements.

4. Uneven distribution of the radioactivity in the atmosphere.

A closer study of the pre- cipitation situation during the first days of March reveals a complex pattern with typical orographical influences partly capable of explaining the highest measured deposition valeus. Radiosonde measurements from Copenhagen and Gothenburg on March 1st show isothermal conditions of slightly above 0° C from the ground to about 750 mb. The temperatures were considerably lower in these layers at Stockholm and Östersund. In concordance herewith the precipitation fell as rain or freezing rain at stations Gothenburg and Högaryd and as snow at the others. Daily measurements also have been made of the amount of water passing the ion-exchanger column compared to precipitation amounts collected in the standard rain gauges at the same place, and differences according to wind and precipitation variations have been found.

It is clear that all factors mentioned above will contribute to make the measured deposition values uncertain. It seems however very probable that the atmospheric contents of fission products must have varied considerably from place to place during the deposition period, and that this was the primary cause to the reported differences.

During the period in question the amount of fission product radioactivity in ground level air in Sweden was lower than expected. This may be due to the fact that the winds in the friction layer of the atmosphere during the first days of March came from a direction between south and east, while winds above 800 mb (where the precipitation originated) had an predominantly west component. After the first short increase of the radioactivity at ground level several days passed before the next high-activity sample appeared.

In Norway there are two stations which show more activity in ground level air on March 1st than the others in Norway and Sweden. The stations are situated at the coast in middle Norway, and it is hardly possible to find a meteorological explanation for the activity differences. The low-level period which followed and the subsequent increase about ten days later are identified also in the Norwegian measurements.

It is wellknown from a number of measurements that the first part of a rain or snowfall generally contains the highest radioactive concentrations. The deposition measurements from Gothenburg and Edsvalla indicate that in this case the dependence on the first precipitation millimetre is unusually strong. At first sight this would point to "washout" (contrary to "rainout") as the dominating scavenging process. A tendency towards an increase of the relative number of larger particles in sample 412 seems to support this theory because according to GREENFIELD (1957) the washout process is most effective for larger particles. On the contrary the high concentrations of Ru 103 and I 131 in this sample and the examination of the ion-exchanger parts of the collecting units indicate that smaller particles ought to be responsible for most part of the first recorded radioactivity. It is possible that such an effect may be explained by a closer examination of the precipitation-formation process and the entrainment of air into the cloud system, e.g. that the first part of a precipitating warmfront cloud system appears at a comparatively high level, where turbulent or advective mixing of more radioactive air parcels from still higher levels are probable.

It is important to note that the decay analysis has shown that large deviations from the $t^{-1.2}$-law can be expected where individual samples are concerned, although they are collected at the same station and immediately after one another.

It is obvious that many interesting results can be obtained by comparing the meteorological situation with results from radioactive fallout measurements. Debris from a single nuclear explosion can be good tracer material

when the background is low enough to permit reliable detection. If ever another nuclear test will be conducted under good tracer conditions much more may be learned about the transport capabilities of the atmosphere. But then there will be a need for a short-time increase of the sampling arrangements in order to get an accurate picture of all processes involved.

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