Meteorological Fractionation of Nuclear Bomb Debris

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Abstract

Radioactive particles are produced by nuclear bomb explosions which are widely different in size and nature. Systematic differences in properties are expected for particles formed under different conditions. Differences in particle properties may cause sorting during deposition because of systematic differences in weather trends at different geographical locations and thus bring to light information about the particles.

Deposits at a mountain station in Norway appear to have contained more short-lived fission products than those at lowland stations, especially during periods when considerable fresh fission products have been present in the atmosphere. This could be due to a substantial chemical fractionation on particle sizes taking place during formation, or to the general formation of particles somewhat larger than 0.5 micron diameter during high-yield explosions.

1. Introduction

Nuclear bomb explosions produce radioactive particles which are measured all over the world. The explosions are widely different in nature and size, and for each explosion, particle formation occurs over an interval of time during which physical conditions are drastically changed. Differences in particle size and composition are, therefore, to be expected.

If the world-wide distributions of radioactive deposition and air concentration are properly analyzed, influences of these differences might turn up. This question will be further examined.

2. Formation of particles

Part of the energy liberated in an explosion is used in evaporation of the bomb material and material from the ground. The resulting warm mass rises rapidly through the atmosphere, is cooled, and particles are formed.

STEWART ET AL. (1956) have microscopically examined bomb debris particles present in samples from the atmosphere and they found that most of the particles bore resemblance to condensation particles.

The conditions under which the particles are formed vary widely. Early substances in some decay chains are gaseous or very volatile, and particles starting to grow very early or very late in the condensation phase may be enriched or diminished in content of certain isotopes. Systematic differences may also arise between particles from larger and smaller bombs due to the slower cooling and later condensation for the former.

After formation, the particles may agglomerate. Some debris clouds may be held tightly together for a long time while others may be torn to pieces at once. The behaviour might be quite different for particles inserted into the dry and stable stratosphere than for those in the much more moist and turbulent troposphere.

Ground deposition of the particles is influenced by the weather, and because weather varies systematically from place to place, the particles may, to a certain degree, be sorted during deposition in such a way that different locations may get particles with different

physical and chemical properties. Such effects may appear between different climates or due to variations within climatic regions, between low and high level ground or between the leeward and windward side of mountain ridges where a predominant wind direction exists.

3. Representativity and analysis of samples

It seems to be plausible that in a climate with a reasonable amount of precipitation, pot samples of bomb debris are fairly representative for the content in precipitation. Such samples are taken regularly in Norway and Figure 1 shows a map over the southern part of the country with 4 sampling stations indicated.

The western part of Norway between the west coast and the top of a south-north mountain ridge forms an area where orographic precipitation very often occurs due to the mean westerly wind bringing moist air over the country. The air current crosses the mountain ridge and is forced to rise as it moves towards the top. This will strengthen precipitation systems and sometimes form precipitation in air masses where it otherwise would not occur. When streaming from the coast towards the mountain ridge, more and more precipitation is exhausted from the same air elements. New dry air entrained into the system tends to decrease the precipitation formed in that part of the clouds, and most precipitation might be expected to be formed in air which existed in the clouds when they crossed the coast. This effect will be most obvious when the air masses are stably stratified, which is often the case for the large precipitation systems giving the bulk of the precipitation in the area.

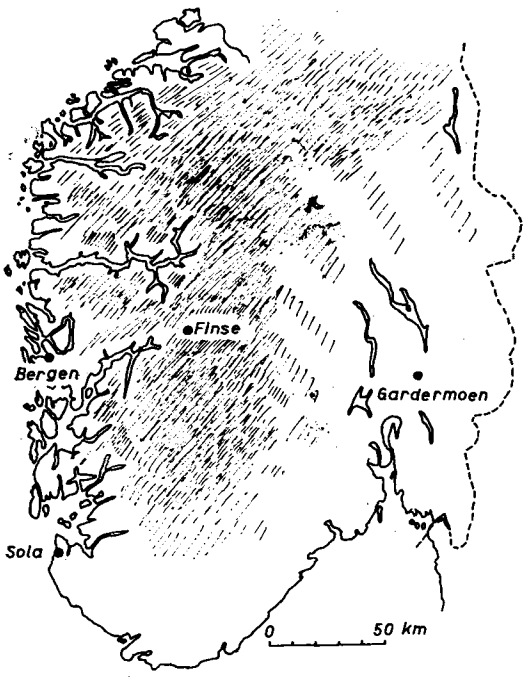

Fig. 1. Map of Southern Norway showing measurement stations. Mountain areas are shaded.

Most particles brought down with rain must for some time have resided in the lower air layers, where precipitation is more or less evenly or arbitrarily distributed. Air concentration of different particles in the cloudbearing layers will then be adjusted to conform with the quasi-constant removal fraction of particles and the rate of incorporation of new particles. When precipitation is suddenly strengthened orographically, a more rapid removal of particles sets in, and a new and lower air concentration of particles tends to be formed in accordance with the removal rate and the incorporation rate now existing. The adjustment will take some time, and one might expect an excess of particles most easily brought down at the start of the orographic area, at Bergen and Sola, and an excess of the more slowly captured particles towards the end of the area, at Finse.

The sampling periods for radioactive deposition are too long to make it possible to select convenient weather situations. The next best thing might be to add observations for such a long time that one may have a fair confidence that the mean conditions prevail. One month is regarded as a lower limit for Norwegian samples.

In this way some masking of an orographic fractionation effect must be expected because of the different origins of precipitation systems. Heavy precipitation may occur partly or all over the area with wind direction inside a large angle, nearly 180 degrees. Showers may occur, and although they may be orographically strengthened, the incorporation of new air is probably much greater than in the more stable precipitation systems. In the upper part of western Norway, precipitation also occurs when the wind comes from the east. The

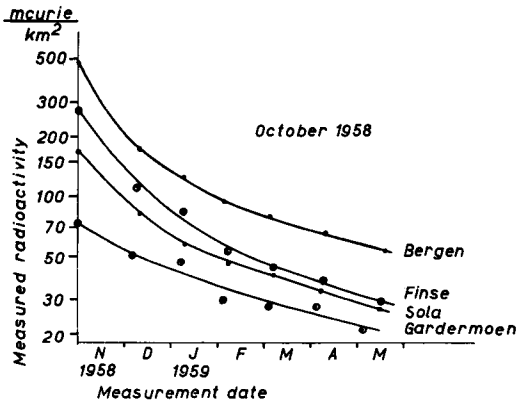

Fig. 2. Decay of October 1958 samples.

mountain station, Finse, lies however at the end of the orographical precipitation area also in this case, while Gardermoen has a position somewhat similar to that usually held by Bergen and Sola.

In one sampling period, the bulk of radioactivity in a sample from one station need not be deposited simultaneously with the bulk of the material at another station.

For meteorological fractionation processes time elapsed since explosion might be decisive, and equal weighting of all deposition with regard to time of deposition is desirable. This meets difficulties.

If t is time since explosion, fission product activity varies roughly as $t^{-1.2}$. If, for instance, products four days old are deposited at the beginning of the month, they will be weighted only $^1/_{10}$ compared with similar products deposited at the end of the period, provided the samples are measured just after the end of the period. Measured one month after the end of the sampling period the former will be weighted about $^1/_2$ of the latter. For older products the weighting factor will be more nearly one, but a discrimination will always exist.

If particles of different ages exist in the samples, the older particles gain more weight the longer time elapses before measurements. A compromise must be sought if comparatively fresh fission products are to be measured. The author feels that if monthly samples are measured one month after the end of the sampling period, fair weight is given to both old and new particles.

From the actual decay curves of Figure 2 it appears that the Finse sample has a more rapid decay than the other samples. Due to the unique position of the Finse site, this is very interesting. For a closer inspection, a decay index has been used. This index is the ratio between the radioactivity of a sample measured six months after end of the sampling period and the radioactivity of the same sample measured one month after end of the period. A relatively high decay index indicates a relatively high content of isotopes with half-life longer than about 2 months.

The decay indices have been computed from smoothed decay curves covering 8—10

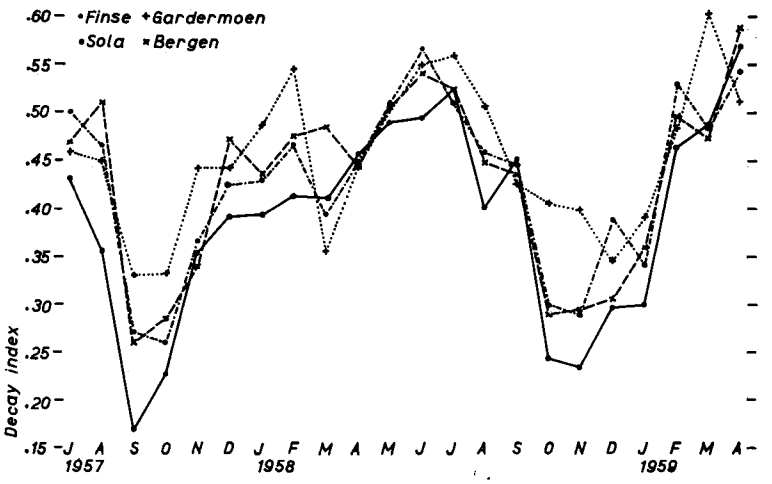

Fig. 3. Decay indices for monthly samples from July 1957 to April 1959.

months of measurements, and are assumed to be correct within 5 per cent. The variation with time has been plotted on Figure 3 for the four stations in question. The general impression is that the same pattern is roughly reproduced in all curves. The Finse decay index has however a tendency to be lower than the others, especially during periods when much fresh debris is present in the air, this appears as a dip on the curves. This feature might point to some sort of selection of particles in the deposits, the reason for which might be found in the mechanism of orographical precipitation.

4. Discussion of measurements

It was earlier mentioned that in an orographic precipitation region the particles most easily brought down by precipitation should be found in excess at the geographical beginning of the precipitation area, and other particles should be found in relatively greater amounts near the end of the precipitation area. It seems logical to associate the less rapid decay of radioactivity at Sola, Bergen and Gardermoen with the particles most easily brought down. The washout by precipitation may be due to the physical or chemical properties of the particles, but it is at present difficult to pursue the last possibility.

Particle capture by water droplets has been examined by GREENFIELD (1957). For larger particles capture by coalescence is predominant, for smaller particles capture by Brownian motion. The border between these two effects is found when the particle diameter is about 0.5 micron, and this corresponds to minimum capture efficiency.

Capture by coalescence will favour large droplets, and from LANGMUIR'S (1948) or MASON'S (1957) tables for the coalescence collection efficiency of droplets, it can be deduced that larger droplets have a higher probability for reaching the raindrop stage. Confronted with Greenfield's results, this means that larger particles should be more easily transported downward by rain than smaller particles. Deposition in precipitation may therefore cause more of the larger particles to be deposited at the lowland stations than at the mountain station. In the periods when the decay index at Finse is lower than at the other stations, it may be that the air pollution either consists of many large particles containing more medium and long-lived fission products, or many small particles containing more short-lived fission products, or both.

The enrichment of large particles with medium and long-lived fission products may be due to chemical fractionation during particle formation. Such an effect is generally dealt with by RAND (1953) and is used for explaining specific problems by STOREBÖ (1959) and EDVARSON ET AL. (1959). It may be mentioned that the authors of the last paper by use of gamma spectrometry on stratospheric samples have found enrichment of a certain mass number in large particles.

If the effect on the decay index on Figure 3 shall be most pronounced when the debris is young, the bulk of the large particles must be cleaned out of the atmosphere within some months after an explosion. For debris from small explosions this agrees with present opinion about residence time in the atmosphere before deposition. However if the large particles are identical with those found in stratospheric air samples (EDVARSON ET AL., 1959), they must have significant velocity of fall if they shall selectively disappear within some months. Calculations show (RAND, 1953, page 86) that a particle diameter about 5—7 micron should be in the right size range.

The other possible explanation of the meteorological fractionation effect is the presence of many small particles when the debris is young. During a test series of mixed small and high yield explosions, most radioactivity in the lower air layers is expected to originate from small bombs, because the bomb clouds do not rise through the tropopause. Most debris from high yield explosions is inserted in the stratosphere, and will generally remain there some time before being deposited. During this period much of the short-lived fission products will decay to a low level, and the particles, will appear as being composed of more long-lived fission products when they reach the ground. The described effect might appear if these particles are generally larger than particles from low yield explosions.

Particles from high yield explosions are generally spoken of as "very small" or "in submicron range". However, if the particles in worldwide fallout are condensation particles, the particle size in high yield explosions

might be larger than for low yield explosions. The rate of cooling of the debris vapour is much slower for the former, leading to a longer period of condensation phase. It is generally known in physics that condensation occurring under such circumstances tend to give larger condensation elements. This is counteracted by the lower atmospheric pressure at the heights at which condensation takes place, and by the greater dilution of the materials which may have taken place. On the other hand, more rapid agglomeration of the particles formed may occur in a particle cloud in the stratosphere due to less turbulent spreading and dilution afterwards.

It thus seems possible that the orographic effect examined here could be due to generally larger particles being formed in high yield explosions than in low yield explosions. The fact that differences in decay index are most pronounced when new debris is present in the troposphere, must then be explained by a lack of younger smaller particles at other times. This must also in this case be due to the rapid disappearance of debris from smaller bombs inserted only into the troposphere.

5. Conclusion

The meteorological fractionation of bomb debris in Norway indicates that a substantial part of the radioactivity might be associated with particles somewhat larger than 0.5 micron diameter, at least during periods when considerable fresh fission products are present. These particles contain, in the mean, less fission products with half-lives shorter than 1—2 months than is the case for the gross activity in the air.

Particles of this size have an appreciable rate of fall if times of years are considered, and they may have an heavy influence on the residence time of the bulk of the debris in the atmosphere. For estimates of bomb debris behaviour in the atmosphere, it might be necessary to take this into consideration. The exchange of air between stratosphere and troposphere may not play such an important part as thought earlier.

Acknowledgments

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