The Dependence of Artificial Radioactivity in Rain on Rainfall Rate

By J. F. BLEICHRODT, JOH. BLOK, R. H. DEKKER and C. J. H. LOCK

Medical Biological Laboratory of the National Defence Research Council T. N. O., Rijswijk, Netherlands

(Manuscript received February 18, 1959)

Abstract

The artificial radioactivity in rain was measured as a function of rainfall rate. As a rule an inverse relationship was found between the specific activity and the rate of rainfall. A tentative explanation is given.

Introduction

Radioactive material released by nuclear explosions is returned to earth in several ways. If the particles are relatively large, gravity fall will be an important factor. On small particles, however, the influence of gravity will be negligible. The deposition of these particles is mainly accomplished by rain.

When no radioactive dust from recent atom bomb explosions is present in the troposphere the rain will contain only small particles of about 5 μ diameter and smaller, because bigger particles will have been deposited by gravity already. Little is known about the mechanism by which these particles are collected by rain or cloud drops.

Falling raindrops will collect radioactive dust particles by impaction. CHAMBERLAIN (1953) and GREENFIELD (1957) made some calculations on this "scrubbing" effect of raindrops. However, these calculations are based on assumptions which are difficult to justify. First the so-called "collision-efficiencies" are used as computed by LANGMUIR (1948) who incorrectly treated the particles as points. MASON (1957) published more exact values which differ appreciably from those given by Langmuir. Both Langmuir's and Mason's data refer to particles of a specific gravity of 1 g cm^{-3}.

Secondly Chamberlain and Greenfield assumed that each collision between a drop and a dust particle results in coalescence. If, however, the particle is not wetted easily it will only enter the drop when it penetrates deeply enough to be enclosed by the drop. Moreover, a nonwettable particle may not be retained for long because it will possibly be expelled as soon as it reaches the drop surface.

Finally Chamberlain and Greenfield use the size distributions of raindrops mentioned by BEST (1950). These distributions, however, apply only to raindrops at ground level. The raindrop spectrum will undoubtedly change with the altitude as the evaporation rate varies with the size of the drop and drops of different size will coalesce. Information on this subject is scarce.

Besides radioactivity collected by impaction, the rain will contain radioactivity picked up in the cloud. Radioactive particles may act as nuclei of condensation and, moreover, the cloud droplets will collect atomic debris by diffusion. GREENFIELD (1957) also made some calculations on this pickup in the cloud.

It will be clear that the scavenging action of rain is a very complex mechanism, which will require extensive experimentation to be fully understood. Greenfield arrives at the conclusion that the fraction of airborne radioactivity scavenged by impaction is proportional to the product of rainfall rate and time. The present paper describes an investigation of the dependence of the radioactivity in rain on rainfall rate.

Experimental

Rain was collected in a galvanized tray of 5 m^2 which was placed with a slight slope on the flat roof of the laboratory. To the lowest part of the tray an outlet-pipe was attached through which the rainwater could be collected in a vessel.

Shortly before the collection of rainwater started, the tray was thoroughly cleaned with tap water in order to remove fall-out material. This method of cleaning proved to be sufficient.

A measuring cylinder was then placed under the outlet-pipe. As soon as the measuring cylinder contained about half a liter of rainwater the remaining water film in the tray was swept down as fast as possible by means of a screen wiper. The total volume collected in this way, usually amounting to about one liter, and the time in which it was obtained were noted. From these data the average rainfall rate (mm h^{-1}) was calculated. The measuring cylinder used was immediately replaced by another.

The bulk of the beta-activity in the rainwater fraction was isolated by adding about 4 g of active coal (Norit C, Norit Verkoopmaatschappij, Amsterdam) per liter, stirring it from 15 to 30 minutes and filtering off the coal. In this way less than 25 % of the beta-activity remained in the water. The filtrate was then evaporated and the residue together with the coal containing filter was ashed at 500° C. This procedure was followed to minimize loss of radioactivity.

At least three days after sampling, to allow the natural radioactivity to decay, a weighed sample of ash was put in a brass tray under an end-window Geiger-Müller tube (Philips 18506) and counted for betaactivity (standard error < 3 %).

The beta-activity per liter of rainwater could then be computed from the measured activity, the sample weight, the total ash weight and the volume of rainwater collected.

Results and conclusions

The results of the experiments lead to the conclusion that the specific activity is dependent on the rate of rainfall, in such a way that the specific activity increases when the rain intensity decreases and conversely. In Figure 1 is plotted a representative part of eleven showers which were analysed.

By means of the measured decay rates of the radioactivity in the rainwater and the approximate relation (HUNTER and BALLOU, 1951; HOLTER and GLASSCOCK, 1952)

$$A = A_1\, t^{-1.2}$$

where A is the activity at time t and A_1 is the activity at unit time after fission, it was possible to demonstrate that the fission products in the collected rain were "older" than two months (once a value of 51 days was found). As mentioned earlier this implies that the presence of large particles is very improbable. Further BLOK (1957) could show by experiments using a cascade impactor that radioactive dust of such "age" contains largely particles smaller than 1.5 μ in diameter.

On the other hand it is shown by extrapolating Mason's "collision efficiencies" (MASON, 1957) that such small particles will be washed out only in minute amounts. Therefore, it is not likely that the dependence of the specific activity on rainfall rate is due to impaction effects. Moreover, experiments made in this laboratory on the hourly fluctuations in airborne activity have never shown a decrease in activity during showers. In interpreting this finding it should be kept in mind, however, that air streams continually bring fresh radioactivity into the rain zone.

If the impact mechanism plays a minor rôle there are two other processes by which the observed phenomenon may be explained. First the evaporation of raindrops will effect an increase in specific activity which is larger when the rainfall rate is lower. Observations on drop size spectra show the mean drop diameter to increase with rainfall rate (BEST,

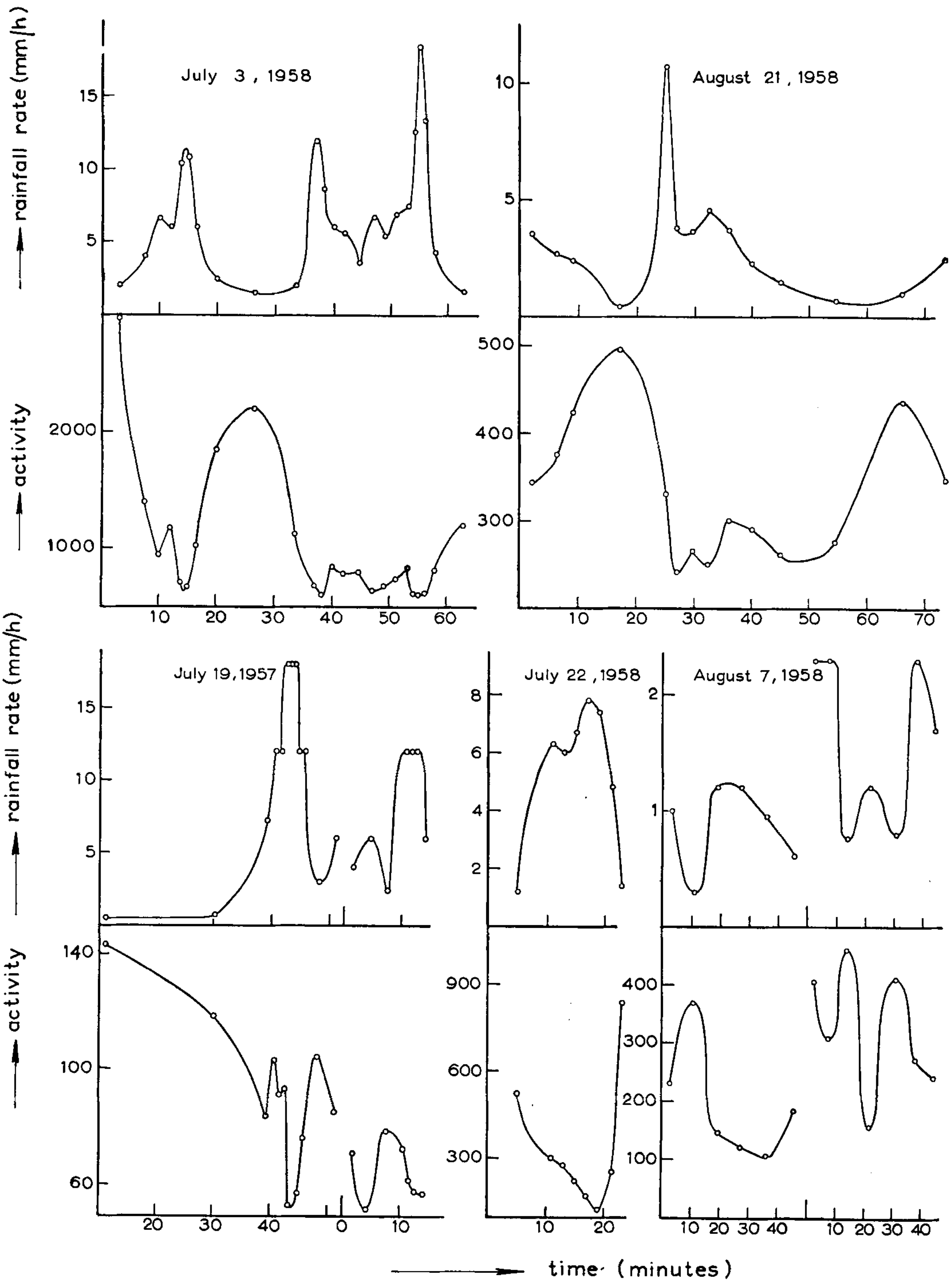

Fig. 1. Fluctuations in rainfall rate and the corresponding fluctuations in specific activity. The upper parts of the graphs show the fluctuations in rainfall rate versus time, the lower parts the specific activity (arbitrary units) versus time. The first point of a curve usually does not coincide with the beginning of the shower.

1950; MARSHALL and PALMER, 1948; MASON and RAMANADHAM, 1953). Large falling drops evaporate relatively slower than small ones (KINZER and GUNN, 1951) so that at low rainfall rates a larger percentage of rainwater will evaporate giving a higher specific activity at ground level.

Secondly the observed fluctuations may be the result of the pickup mechanism in the cloud. The physics of rain formation is not yet sufficiently understood. Therefore it is impossible at the present time to predict the dependence of the specific radioactivity of the rain on rain intensity just below the cloud base.

Recently COWAN and STEIMERS (1958) reported measurements of the fluctuations of radioactivity in rainwater. By collecting hourly samples and evaluating gross activity they could discern two types of behaviour:

1. The gross activity is usually proportional to the volume of rainwater in the sample.

2. The concentration of activity tends to vary inversely with the volume of rainwater so that high concentrations are observed during periods of low rainfall rate and low concentrations when heavy rainfall occurs.

In the light of the results reported in this article this is understandable. By taking hourly samples the fluctuations in specific radioactivity will, for the greater part, be masked. This explains the first type of behaviour. A variation of activity with rainfall rate as reported by these authors will be found only when the rainfall rate differs slightly from the mean during an hour of sampling and the mean rainfall rate varies sufficiently from hour to hour.

Acknowledgement

The authors are indebted to Prof. Dr. W. Bleeker and Dr. F. H. Schmidt of the Royal Netherlands Meteorological Institute for valuable discussions.

REFERENCES

BEST, A. C., 1950: The size distribution of raindrops, *Quart. J. R. Meteor. Soc.*, **76**, p. 16.

BLOK, JOH., 1957: *Radioactieve besmetting van de biosfeer in Nederland*, Thesis Vrije Universiteit, Amsterdam, H. A. van Bottenburg N. V.

CHAMBERLAIN, A. C., 1953: Aspects of travel and deposition of aerosol and vapour clouds, *A.E.R.E. HP/R 1261*, Harwell.

COWAN, F. P., and STEIMERS, J., 1958: The distribution of fallout activity in rainfall at Brookhaven National Laboratory, June to September 1957, *BNL 496(T-117)* Upton, N.Y.

GREENFIELD, S. M., 1957: Rain scavenging of radioactive particulate matter from the atmosphere, *J. Meteor*, **14**, p. 115.

HOLTER, N. J., and GLASSCOCK, W. R., 1952: Tracing nuclear explosions, *Nucleonics*, **10**, No. 8., p. 10.

HUNTER, H. F., and BALLOU, N. E., 1951: Fission product decay rates, *Nucleonics*, **9**, No. 5, p. C-2.

KINZER, G. D., and GUNN, R., 1951: The evaporation, temperature and thermal relaxation-time of freely falling waterdrops, *J. Meteor.*, **8**, p. 71.

LANGMUIR, I., 1948: The production of rain by a chain reaction in cumulus clouds at temperatures above freezing, *J. Meteor.*, **5**, p. 175.

MARSHALL, J. S., and PALMER, W. M., 1948: The distribution of raindrops with size., *J. Meteor.* **5**, p. 165.

MASON, B. J., 1957: *The physics of clouds*, Oxford, Clarendon Press, p. 424.

MASON, B. J., and RAMANADHAM, R., 1953: A photoelectric raindrop spectrometer, *Quart. J. R. Meteor. Soc.*, **79**, p. 490.