On some results of measuring radioactivity of individual raindrops

By B. I. STYRA and B. K. VEBRIENE, *Institute of Physics and Mathematics, Academy of Sciences of Lithuanian SSR, Vilnius, USSR*

(Manuscript received January 10, 1968. Revised version April 14, 1968)

ABSTRACT

A method of measuring the radioactivity of separate raindrops and ice-crystals by capturing them on the sampling surface, with subsequent contact with nuclear emulsion A-2, is described.

The number of non-radioactive raindrops is defined. It is shown that for continuous rain the percentage of non-radioactive raindrops is near 30 %.

The average radioactivity value of individual raindrops increases with their size. The rise of this radioactivity value slows down as the size of droplets increases. The specific radioactivity of raindrops increases with the decrease of their size. Hence, the value of raindrop evaporation by fall from the cloud to the ground is obtained.

It is shown that the frequency maximum of raindrops having identical specific radioactivity takes place. On the assumption of a simplified activation scheme of raindrops in the cloud, it is shown that the processes there are quite non-stationary.

The possibility of measuring or evaluating the radioactivity of separate raindrops is presented in papers by Styra & Garbaliauskas (1955), Gerlach & Stierstadt (1960), Styra & Vebriene (1966) and Stierstadt & Kadereit (1961). Such measurement seems to be very perspective for it offers the opportunity of revealing new facts and enables attempts to be made to study the microphysical processes of the mechanism of the self-cleansing of the atmosphere from radioactive contamination.

The Radioactivity of single raindrops or ice-crystals was measured in the following way: A glass plate covered by filter paper or polished stainless steel plate was put in the open air for several seconds. Other materials (such as organic glass, brass, polyethylene or photo emulsion) proved to be less convenient (see Styra & Vebriene, 1966). A mixture of talc and erythrosin was rubbed into the filter. Being moistened, the erythrosin was coloured bright red. During evaporation raindrops left a visible stain on the polished plate. After evaporation of the raindrops and drying, the polished plate, with traces of droplets on it was put into contact with 50μ thickness of emulsion sensitive to α-particles and protons. Photoexposure lasted, depending on the task, from several hours up to scores of days. After this photoexposure the emulsion was developed and dried. Later, the plates were examined under the microscope at a magnification of $\times 500$–1.200. α-Tracks found in those places where the emulsion was put into contact with the raindrop traces on the paper or plate characterized the radioactivity value of the raindrops.

Raindrop size was determined according to the stain area on the filter or polished plate by means of calibration curves.

α-Particles came into the emulsion from above. Therefore, when measuring the radioactivity of raindrops, only those tracks should be taken into account which have their beginning in the upper part of the emulsion. The thickness of the emulsion after photo development decreased by 2.5 times. Hence, to preserve the first α-tracks' length in the emulsion, the emulsion should be restored to its original thickness. This was achieved by exposure of the emulsion in 20% aqueous solution of glycerine for an hour at room temperature.

The length of the α-tracks in the emulsion was measured and frequency histograms of the length of the α-particle traces were drawn up in order to determine the isotope composition of the

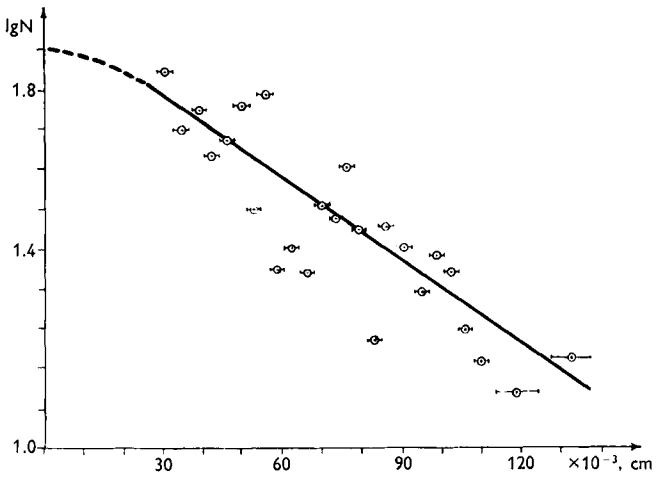

Fig. 1. Semilogarithmic dependence of non-radioactive droplets in continuous rain on their radius (in per cent).

radioactivity in a sample. Moreover, we made use of the data on stainless steel plates, since a partial absorption of energy in some α-particles, due to aerosol penetration into the filter paper pores, was observed by sampling on the filter.

The natural background was determined by the counting of α-tracks arising in those places where there was no contact of the emulsion with the traces of raindrops left on the sample plates or control plates. Relative error of measurement did not exceed 5–30%.

Before use, the photographic plates were exposured in 0.1% solution of potassium ferricyanide for 10–15 minutes and after one hour's washing were dried. This was done for the annihilation of the α-tracks' background.

The radioactivity of raindrops was defined using the contact method by means of photoexposure of emulsion with one side of the sample. This meant that the radioactivity of the sample was measured in the 2π angle. For definition of the values of radioactivity the obtained α-tracks values were multiplied by two, so the evalution at weak radioactivity was not accurate.

Conclusions were drawn from measurements of over 10.000 raindrops. First of all, an attempt to count the number of non-radioactive raindrops was made but while measuring in the 2π angle this number was obviously exceeded.

For this, short-lived radioactivity (defined by isotopes, mainly the short-lived Rn decay products) and long-lived radioactivity (determined by the other isotopes occurring in nature) should be distinguished. The short-lived radioactivity was defined by exposure up to 24 hours, the long-lived radioactivity after the first exposure some 10 days later. It was shown by histograms and by definition of the isotope composition of a sample that in the case of short-lived radioactivity the connection was with the nearest Rn decay products and to a lesser degree with Tn. In the case of long-lived radioactivity α-tracks were considerably shorter, which is characteristic of Po^{210}, U^{238}, U^{235}, Pu^{239} and some other isotopes.

While counting the number of non-radioactive raindrops it was found that their number decreased with the increase of raindrop size. This is shown in Fig. 1. In this and other figures, each dot is the mean value of several tens or even hundreds of raindrops. This dependence on raindrop size of more than $3{:}10^{-2}$ cm can be repre-

Table 1. Content of non-radioactive raindrops from measurements in the 4π angle

No. of raindrops	In 2π angle		In 2π angle		In 2π angle	
	No.	%	No.	%	No.	%
444	275	62	276	62	205	46

Table 2. *Number of non-radioactive droplets*

No. type of rain	Type of radioactivity	Droplets no.	Non-radioactive droplets, %	
			In 2π angle	Corrected for 4π angle
1 Continuous	Short-lived	954	43	27
2 Continuous	Long-lived	900	26	12
3 Shower	Short-lived	117	19	—
4 Snow	Short-lived	1175	77	—

sented in the form of exponential function. From this figure one can arrive at a preliminary conclusion that the number of non-radioactive raindrops in a given size range decreases with increasing radii. In addition one more conclusion can be drawn, namely: raindrops acquire activity mainly by being already formed, and active aerosols as a rule are never condensation nuclei.

For evaluation of percentage error in counting the number of non-radioactive droplets in the 2π angle the following control experiment was carried out: Raindrops were gathered on the cloth ΛΦC-1 with a thickness of 0.3–0.6 mg/cm² and after a little drying they were put between two emulsions, i.e. measurements were carried out in the 4π angle. Experimental results are given in Table 1.

Simple calculations show that a real number of non-radioactive raindrops (N_0) by measurements in the 2π angle can be calculated by the following formula:

$$N_0 = N(1-\alpha) - N_r^*, \qquad (1)$$

where N_r^* is the radioactive droplets number measured in the 2π angle, N is the total number of droplets, $\alpha = N_H/N$ is the coefficient defined by the control experiment and N_H is the number of fictitious non-radioactive droplets obtained additionally due to measurements in the 2π angle. The coefficient α has been obtained for continuous rain and proved to be equal to $\alpha \approx 0.16$.

The data of the non-radioactive droplets number corrected for continuous rain by means of formula (1) are given in Table 2.

Thus, the data of Tables 1 and 2 show that among raindrops and ice crystals a large percentage proved to be non-radioactive droplets and consequently the activation process of droplets in a cloud has a statistical character.

The graph of the dependence of the radioactivity of individual droplets on their masses is given in Fig. 2. Here and subsequently, the

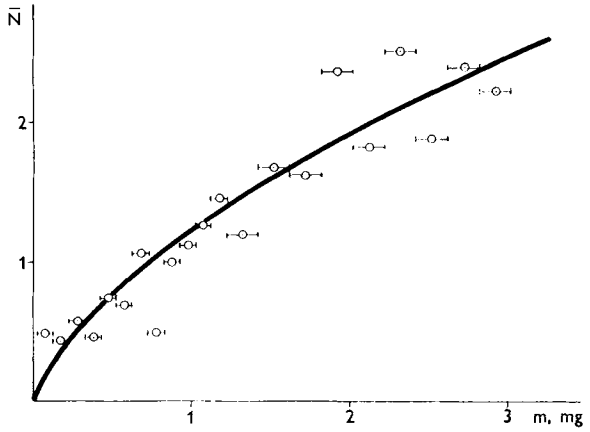

Fig. 2. Fallen droplet mass dependence on its radioactivity expressed in α-tracks' number in the contact region of the photo-emulsion with the droplet's trace.

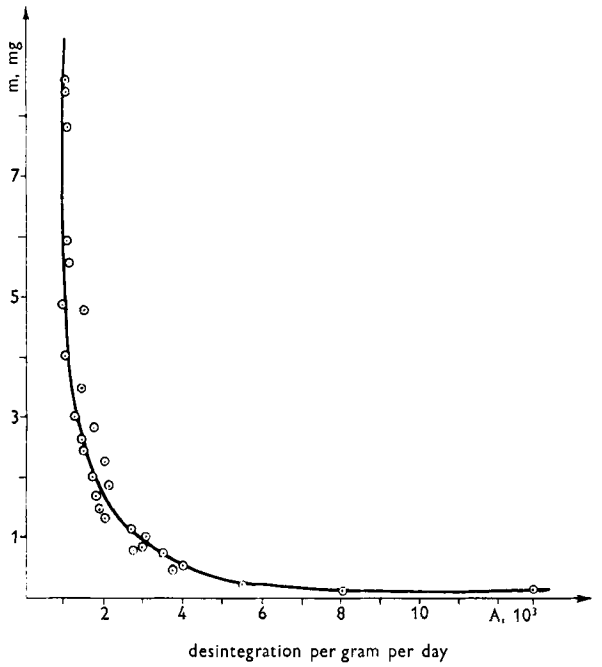

Fig. 3. Droplet's mass dependence upon its specific radioactivity value.

investigation was carried out only with short-lived Rn decay products. It is seen from Figure 2 that radioactivity increases with droplet size. However, this increase is not proportional to the droplet mass growth, that would have taken place if this radioactivity increase had been a consequence of only this coagulation of lesser raindrops. The observed regularity can reflect a number of processes: coagulation of radioactive droplets with non-radioactive capture by droplets during their fall, etc.

Fig. 3 gives the dependence of specific radioactivity on droplet mass. It is seen from this figure that the specific radioactivity increases with droplet decrease. Otherwise, when raindrops increased the specific radioactivity decreased approaching a conctant value. Hence, if it is assumed that the constant value characteristic of radioactivity is established in the cloud, there arises the possibility of calculating droplet evaporation intensity by fall, postulating that specific activity growth is conditioned only by evaporation of falling droplets. The relative decrease of droplet mass $\Delta m/m$ can be calculated by the formula:

$$\frac{\Delta m}{m} = 1 - \frac{A_0}{A_0 + \Delta A}, \tag{2}$$

where A_0 is the specific radioactivity in the cloud and ΔA is its increase by fall.

Table 3

No.	Droplet weight near the ground (mg)	Relative evaporation value	Droplet weight during leaving the cloud
1	0.12	0.83	0.7
2	1.5	0.75	2.0
3	1.0	0.58	2.4
4	1.25	0.50	2.5
5	2.0	0.30	3.0
6	3.0	0.23	3.6

Droplet evaporation values for averaged samples of continuous rain calculated by formula (2) are given in Table 3.

Finally, the frequency number (N%) of droplets with the given specific radioactivity value appears in Fig. 4. Each dot is a number of droplets expressed in per cent of all the droplets' specific radioactivity which is within $(n \pm 0.5) \cdot 10^3$ α-tracks per gram.

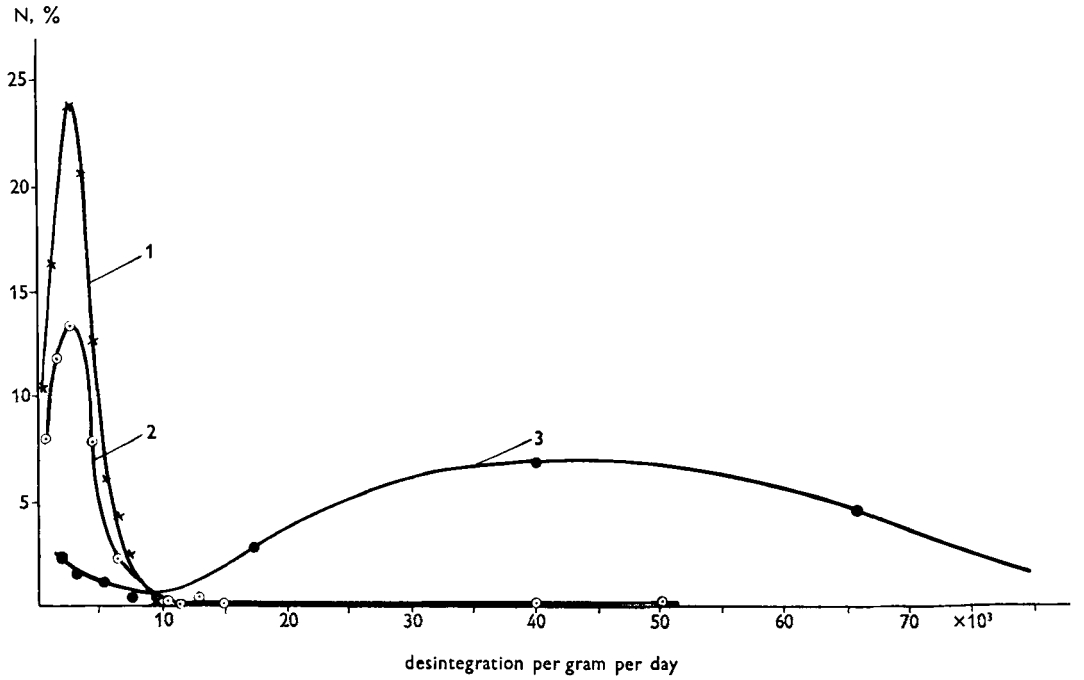

Fig. 4. Frequency (in per cent) of droplet number of the given value of specific radioactivity: *1*, shower *2*, continuous rain; *3*, snow.

The frequency maximum of droplets of certain specific radioactivity as well as the number of droplet decrease according to large and lesser activity values takes place as given in Fig. 4. During rain maximum it is at the value $2.5 \cdot 10^3$ α-tracks per gram per day, and during snow it is equal to $40 \cdot 10^3$ α-tracks per gram per day.

The presence of a probability maximim of the specific radioactivity of droplets is indicative of the tendency of droplets to satisfy a certain condition of dynamic equilibrium in the activation process, atom decay on droplets and droplet evaporation.

It follows from Fig. 4 that during showers specific radioactivity distribution on droplets is within a comparatively narrow range (curve 1). The latter is connected with the process of shower formation and it shows that raindrops are formed very quickly. The curve distribution of continuous precipitation to the part of high specific radioactivity indicates the age of droplets in the cloud, which have had enough time to activate up to high specific values. This fact demonstrates the possiblity of relatively large evaporation by fall that is characteristic of small droplets.

Thus a simple non-stationary equation of specific radioactivity accumulation on droplets has been solved. This accumulation is a result of two processes: (1) coagulation of radioactivity aerosols with droplets and (2) the aerosols' decay on droplets at various suppositions of Rn shortlived decay products distribution in a cloud.

The coagulation coefficient is taken from a paper by Styra *et al.* (1966). It has been established that the period of 150–200 min is necessary to set up equilibrium conditions. The same specific radioactivity corresponding to the maximum for rain given in Fig. 4 is obtained during 5 minutes. It is indicative of the sharp non-equilibrium of the activation processes of particles in a cloud. Equilibrium radioactivity of droplets (according to calculation) is equal to $\sim 3 \cdot 10^4$ α-tracks per gram per day. The last value occurs very seldom.

Thus, the supposition arises that the long accumulation of radioactive aerosols on cloud particles does not take place (especially in convective clouds).

It probably means that a continuous process of new droplet formation and growth as well as evaporation and repeated enrichment of a

cloud's air by radioactive aerosols (being on cloud particles) takes place in a cloud. The conditions in the experimental graphs (Fig. 4) are formed during this complex process. We see (Fig. 4) that the radioactivity maximum of cloud droplets occurs at a low specific radioactivity.

By artificial injection of Po^{210} (by means of special rockets) into convective clouds, some measurements of the radioactivity of individual raindrops were carried out by the α-tracks number. A new curve analogous to that of Fig. 4 has been obtained. It shows the activation processes' similarity. In addition the maximum here proved to be one order more and can be accounted for by physical and chemical properties of an injected isotope.

REFERENCES

Gerlach, W. & Stierstadt, K. 1960. Untersuchungen über radioaktive Niederschläge und die Aktivität der Luft. VI. *Atomkernenergie 5*, No. 9.

Stierstadt, K. & Kadereit, A. J. 1961. Die Verteilung der künstlichen Radioaktivität und der Menge des festen Rückstandes auf Grössenspektrum der Regentropfen. *Meteorol. Rundsch. 14*, No. 1.

Styra, B. I. & Garbaliauskas, Č. A. 1955. K voprosu ob izmerenii aktivnosti otdelnykh kapel doshdya. *Trudy AN Lit. SSR*, seriya B, 2 pp.

Styra, B. I. & Garbaliauskas, Č. A. 1955. O resultatakh izmerenija aktivnosti i zaryada otdelnykh kapel doshdya. *Trudy AN Lit. SSR*, seriya B, 2 pp.

Styra, B. I. & Vebriene, B. K. 1966. O metodike izmereniya radioaktivnosti otdelnykh chastic osadkov i nekotorye predvaritelnye resultaty takikh issledovanii. *Physika atm. i okeana. 2*, No. 10.

Styra, B. I., Vebra, E. I. & Shopauskas, K. K. 1966. Determination of some parametres of radioactive aerosol removal from the atmoshere. *Tellus 18*, No. 2, 541–548.

О НЕКОТОРЫХ РЕЗУЛЬТАТАХ ИЗМЕРЕНИЯ РАДИОАКТИВНОСТИ ОТДЕЛЬНЫХ КАПЕЛЬ ДОЖДЯ

Описана методика измерения радиоактивности отдельных капель дождя и снежинок путем захвата их на собирающую поверхность с последующим приведением в контакт с эмульсией типа А-2.

Определено число нерадиоактивных капель. Показано, что для обложных дождей процент нерадиоактивных капель близок к 30 %.

Величина радиоактивности отдельных капель в среднем возрастает с их размером, замедляясь по мере увеличения размеров. С уменьшением капель растет их удельная радиоактивность. Исходя из этого получена оценка величины испарения капель по мере их падения из облака на землю.

Показано, что имеет место максимум повторяемости капель, имеющих одинаковую удельную радиоактивность. Допуская упрощенную схему активации капель в облаке показано, что процессы там протекают резко не стационарно.