Seasonal variation of atmospheric radioactivity at Ibadan

By A. O. SANNI, *Department of Physics, University of Ibadan, Ibadan, Nigeria*

(Manuscript received March 20; revised version September 21, 1972)

ABSTRACT

The results of regular monitoring of air samples for radioactivity at Ibadan for the period July 1966 to February 1967 are presented here. The results show a general increase of atmospheric specific radioactivity and particular periods of high atmospheric specific radioactivity during the Harmattan season. The general increase in atmospheric specific activity is not correlated with nuclear weapons tests and is explained in terms of the enhanced exhalation of emanation from the earth's crust. The prominent peaks of atmospheric specific radioactivity are shown to correlate with severe Harmattan days, characterized by high specific volume of water vapour in the atmosphere. The prominent peaks of atmospheric specific radioactivity during the Harmattan months are attributed to the continental north-easterly winds which transport radioactive dusts from the radioactive fields of Niger Republic and of the plateau regions of Nigeria to the station of observation.

1. Introduction

During the French weapons tests in the Sahara in 1960, monitoring of air for radioactivity was undertaken in order to estimate the radioactive fall-outs from the tests. The results of this monitoring have been published by Agu (1965). Since then routine monitoring of air samples at Ibadan has been continued for the prime purpose of establishing a baseline of radioactive contaminants of the atmosphere due to natural sources as well as to nuclear devices. The result of this monitoring for the period July 1966 to February 1967 which covers both the wet and the dry seasons in Nigeria is presented here. This period July 1966 to February 1967 was the only period for which continuous data were available, since monitoring was disrupted by the civil war in Nigeria. Nevertheless the results for this period were found interesting enough to analyse.

A statistical analysis of the results is discussed. The seasonal changes in the specific radioactivity of the atmosphere are explained in terms of meteorological factors.

2. Observations and data reduction

(a) *Observations*

Air was drawn in through a filter paper by means of the Fleming Air Sampler, Type 1355B, which has a built-in anemometer for measuring the suction speed. The filter head was mounted at a height of 2.2 m above ground level. The air sampler was operated continuously with short breaks at 8 a.m. each day for changing the filter paper. At the end of a daily run the sample collected was dried and then counted for 5 min with a thin window, low background Geiger-counter which has an anti-coincidence guard counter. As this Geiger-counter was only 0.025 % efficient to gamma radiation and approximately 70 % efficient to both alpha- and low energy beta-particles, the radiation detected was due mainly to alpha- and low energy beta-particles. This was confirmed by the simple absorption test.

Other data employed for the interpretation of the results of atmospheric radioactivity are, the hourly dew point, hourly pressure, hourly temperature, and the record of dates of reported nuclear weapons tests.

(b) *Data reduction*

From the count rate of the dust sample collected and the volume of air sample monitored, the daily specific radioactivity (in pCi per kgm of air) of the atmosphere was determined.

Starting from Clausius-Claperyon equation for unit mass

$$\frac{d\varepsilon_s}{dT} = \frac{L}{T(v_s - v_L)} \qquad (1)$$

where ε_s = the saturation vapour pressure, T = the temperature of vapour in the Kelvin temperature, L = the latent heat of vaporisation, v_s = the specific volume of saturated vapour, v_L = the specific volume of saturated liquid; and assuming that the water vapour obeys gas laws, that L varies linearly with temperature, i.e. $L_T = L_a(1 + \alpha T)$ and that $v_s \gg v_L$, it can be shown that

$$\log_e \varrho_w = \frac{L_a}{R_w}\left(\frac{1}{T_{100}} - \frac{1}{T_d}\right) + \alpha \log_e \frac{T_d}{T_{100}} + \log_e \left(\frac{\varepsilon_{s100}}{R_w T_A}\right) \qquad (2)$$

where L_a = the latent heat of vaporisation at 273.15 K, R_w = gas constant for water vapour, $T_{100} = 373.15$ K, T_d = temperature at which water vapour in air at temperature T_A when cooled at constant pressure will just be saturated, ε_{s100} = saturation vapour pressure at 100°C, and ϱ_w = density of water vapour in air at T_A.

The following values of L_a, α, R_w were assumed:

$$L_a,\ 3166 \times 10^3 \text{ Joule kg}^{-1}$$
$$\alpha,\ -7.7 \times 10^{-4} \text{ K}^{-1}$$
$$R_w,\ 4.62 \times 10^{-4} \text{ Joule K}^{-1} \text{ kg}^{-1}$$

By employing eq. (2), and the hourly dew point and air temperature, the hourly density of water vapour in kg m^{-3} of air was calculated. From these data the daily means were obtained.

Since the relative humidity varies in the same manner with density of water vapour in atmosphere, specific volume of water vapour rather than its density is employed as an indication of dryness of the atmosphere and consequently of the severity of the Harmattan. Hence density of water vapour is converted to specific volume of water vapour.

(c) *Results of data reduction*

The detection limit of specific radioactivity by the low background Geiger-counter is determined mainly by the background count rate which is about 0.6 c.p.m. This count rate corresponds to a specific radioactivity of 0.05 pCi kg^{-1}. For the comparison of day to day specific radioactivity, the uncertainty in measurement is more important than the detection limit, since uncertainty in measurement is much greater than the detection limit.

Fig. 1*a* shows daily values of atmospheric specific radioactivity. From the period mid-July to mid-October, 1966, the atmospheric specific radioactivity was fairly constant at about 1 pCi kg^{-1} of air, except for statistical fluctuations. The uncertainty in measurement for these low specific radioactivity is equivalent to a relative standard deviation of 15 %. At the onset of Harmattan (Oct.–Nov.), a general increase of specific radioactivity set in, which continued until Feb. 1967. Between Nov. 1966 and Jan. 1967, pronounced peaks of radioactivity superimposed on an enhanced background level of activity were registered. The uncertainty in measurement for the prominent peaks is equivalent to a relative standard deviation of 5 %. These peaks labelled (i) ... (v) in Fig. 1*a* correlated well with severe Harmattan days characterised by high values of specific volume of water vapour in Fig. 1*b*.

3. Correlation of atmospheric specific volume of water vapour with atmospheric specific radioactivity

(a) *Method of analysis*

The period between July 1966 to February 1967 was divided into 15 intervals each comprising of 14 days. These intervals were selected such that each of the four prominent peaks exhibited in Fig. 1*b* fell within an interval. For each interval, the daily values of atmospheric specific volume of water vapour and atmospheric specific radioactivity were cross-correlated. The cross-correlation coefficient, r, for each interval was calculated using different time lags, l, of 0, 1, 2, 3, 4 and 5 days.

The advantage of this method of analysis lies in the fact that other relationships, apart from the obvious ones which are not discernable from Figs. 1*a* and *b*, would be revealed. Also the relative importance of the deduced relationships can easily be assessed by testing for the significance of r. In testing for the significance of r, the Student's t distribution is assumed since only 14 pairs of data constituting a small sample, are available in each interval. If n is the number of pairs of data on

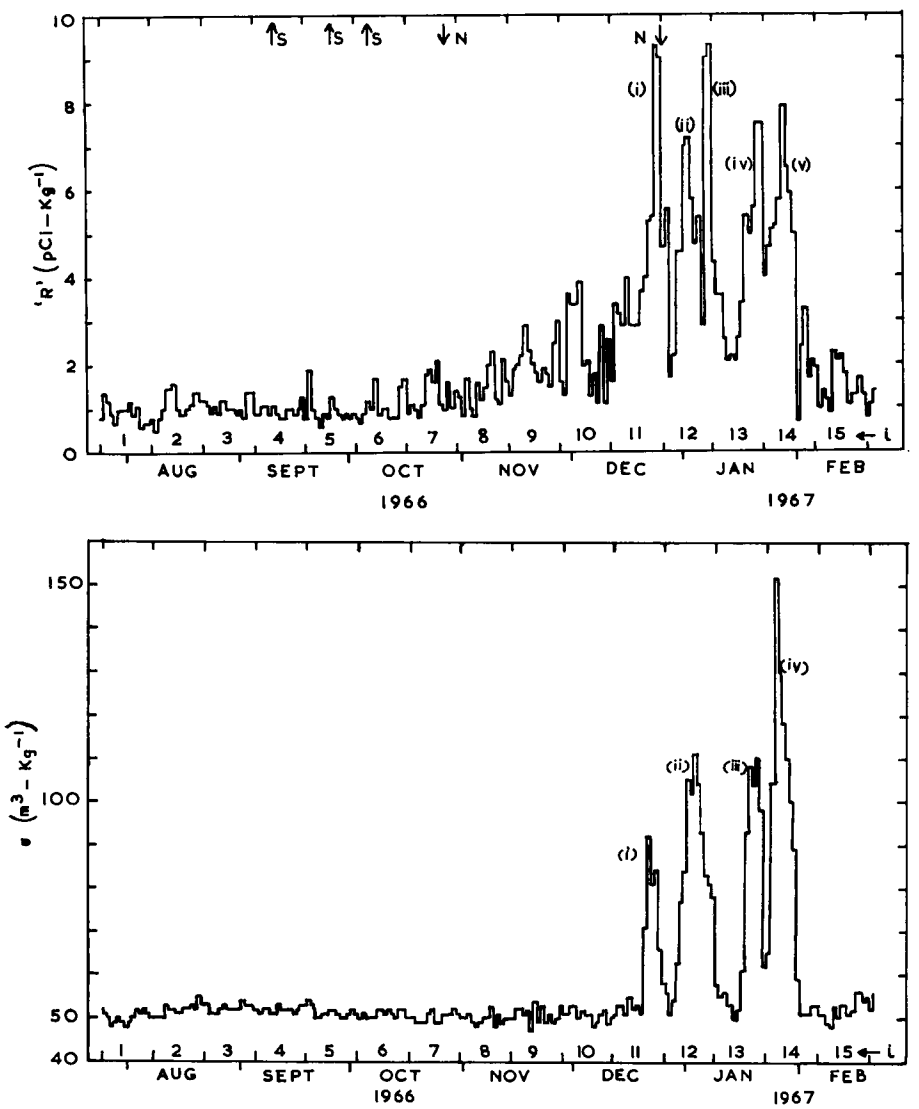

Fig. 1a. The daily values of atmospheric specific radioactivity R. Arrows on the top of the diagram indicate approximate dates of nuclear tests and the hemisphere in which tests were conducted. N, northern hemisphere; S, southern hemisphere.
Fig. 1b. The daily values of specific volume of water vapour v in the atmosphere. Prominent peaks of specific volume of water vapour correspond to severe Harmattan days.

which the correlation is based, and r the correlation coefficient obtained, then t is given by

$$t = \frac{r}{\sqrt{1-r^2}}\sqrt{n-2}, \qquad (3)$$

when Sheppard's correction is neglected. Hence the probability, P, that such a correlation should arise, by random sampling, from uncorrelated data was deduced (Fisher, 1954). If P is less than 0.02, the correlation is regarded as significant.

Also the variation of r with time lag for significant correlations would lead to a realistic assessment of time lags.

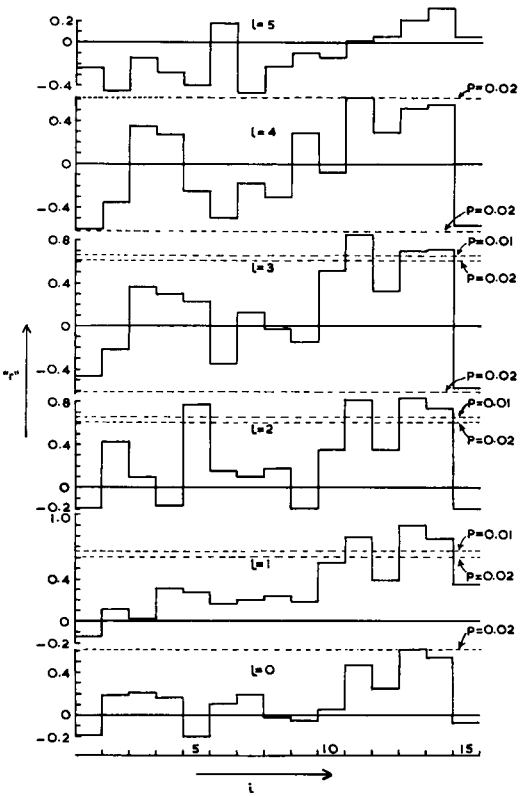

Fig. 2. The variation of cross-correlation coefficient r with time lag, l, for the different intervals (i). These intervals were selected such that each of the four prominent peaks exhibited in Fig. 1*b* fell with an interval.

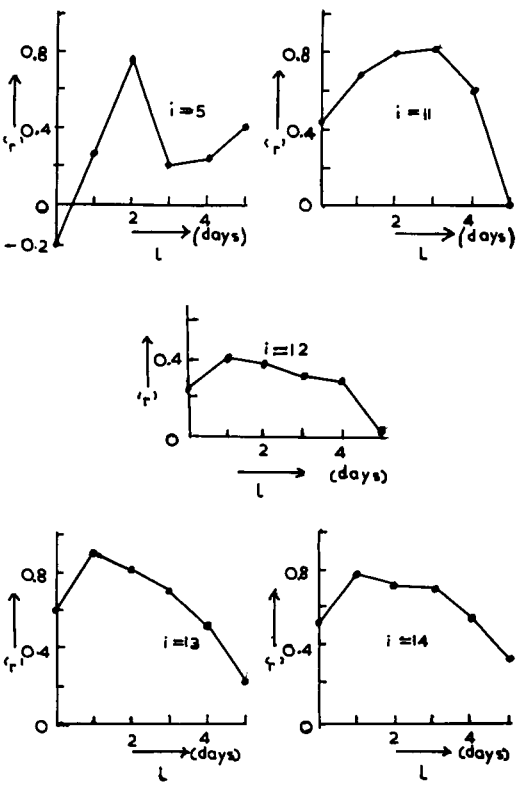

Fig. 3. The plots of cross-correlation coefficient r against time lag for the 5th, 11th, 12th, 13th and 14th intervals for which r was significant at a probability of 0.02 of accidental correlation.

(b) *Results for r*

The cross-correlation coefficient for the different intervals have been calculated. The results obtained for time lag, l, of 0, 1, 2, 3, 4 and 5 days are shown in Fig. 2.

When no time lag is introduced, correlation of specific volume of water vapour with specific radioactivity is just significant for the 13th interval, but for time lags between 1 and 4 days, correlation is significant at a formal probability of 0.02 that correlation is accidental for 5th, 11th, 13th and 14th intervals.

(c) *Results for time lags*

In order to deduce the time lags for significant correlation, the cross correlation coefficient r is plotted as a function of time lag, l, and the value of l when r is maximum is taken as the time lag. Hence from Fig. 3, time lag for the 5th interval is 2 days, 11th interval 2–3 days, 13th interval 1 day, and the 14th interval about 2 days. The result for the 12th interval is included for comparison with other intervals which contain peaks of specific radioactivity. The shape of the curve is similar to that of the 14th interval though r is not significant.

4. Discussion

(a) *General increase of specific radioactivity*

The dates of nuclear weapons tests are indicated in Fig. 1*a*. The French tests were carried out in the South Pacific (Southern Hemisphere) on the 11th and 24th of September,[1] and the

[1] Reported Nuclear Detonations, Sept. 1966, *Radiological Health Data*, vol. 7, no. 10, US Dept. of Health, Education and Welfare.

4th of October, 1966[1] while the Chinese tests were carried out in China (Northern Hemisphere) on the 27th October[1] and the 28th December 1966[2]. These tests which are of low yields were estimated to contribute not more than 1 pCi kg^{-1} to the measured specific radioactivity because of the distances of the sites of tests from Ibadan. Moreover, sharp increases of specific radioactivity would be expected within two weeks of the tests. From Fig. 1*a*, it can be seen that there is no obvious association between nuclear weapons tests and the general increase in atmospheric radioactivity during the period July 1966 to February 1967.

The general increase of specific radioactivity of the atmosphere during Oct.–Feb. can be explained in terms of exhalation of emanation from the ground. The earth's crust contains the radioactive nuclides, U-238 (1.3 μCi m^{-3}), U-235 and Th-232 (2.2 μCi m^{-3}) which decay to produce isotopes of Radon. After formation in the ground, Radon (Rn-222) and Thoron (Rn-220) which are gases, diffuse into the atmosphere. The decay products which are heavy metals then become attached to natural aerosol particles. Israel (1958) has proposed a simple theory for such a diffusion process.

(b) *Particular periods of high specific radioactivity*

The results of statistical analysis of data presented in Section 3(b) indicate that when the appropriate time lag is introduced, the correlation of periods of high specific radioactivity and those of specific volume of water vapour is significant, except for the 12th interval corresponding to peaks (ii) and (iii) of Fig. 1*a* and (ii) of Fig. 1*b*. This implies a dependence of high specific radioactivity on specific volume of water vapour or the dependence of both parameters on a common factor.

Taking into consideration that (1) the cross-correlation coefficient, r, obtained in Fig. 3 are moderate in values and (2) that despite the one to one correspondence in the peaks of the two parameters for the 11th, 13th and 14th intervals, the coefficient of correlation was insignificant for the 12th interval, one can conclude, contrary to the case of the general increase in specific radioactivity, that the dryness of the air could not be directly responsible for the high values of specific radioactivity recorded during the Harmattan. Additionally, one would expect to record the same time lags for the peaks (i), (ii) and (v) of Fig. 1*a* for which moderate correlation exist, if the converse were the case.

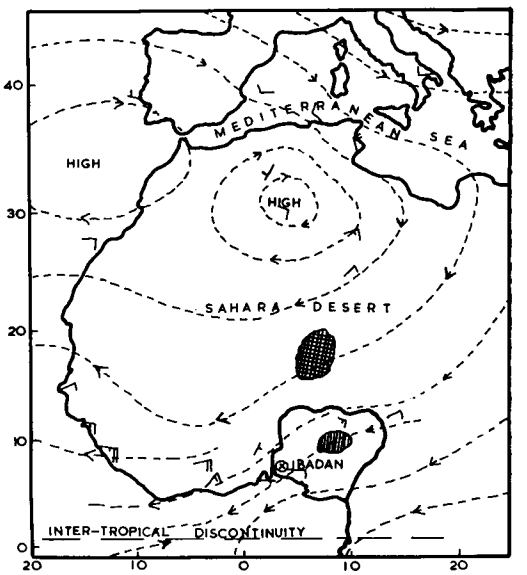

Fig. 4. Streamlines at 850 mbars for the 2nd of Jan. 1967—a typical severe Harmattan day at Ibadan. ⟋, wind of 20 knots blowing from N.E. ⟋, wind of 10 knots blowing from N.E. ⟋, wind of 5 knots blowing from S.W. Recognised areas of high radioactivity are shaded. Crosshatched area, the radioactive fields of Basin of Agada in the Niger Republic; striped area, the radioactive fields of plateau regions of Northern Nigeria.

In interpreting the results of the strong correlation of periods of high specific volume of water vapour and high specific radioactivity, an important fact which must be taken into consideration is that periods of high specific volume of water vapour correspond to the presence of continental north-easterly wind at a station on the West Coast of Africa. The north-easterlies blow around the subtropical high pressure cells which are virtually stagnant over the Sahara Desert during the Harmattan season (Fig. 4). These north-easterlies were

[1] Reported Nuclear Detonations, Oct. 1966, *Radiological Health Data*, vol. 7, no. 11, US Dept. of Health, Education and Welfare.

[2] Reported Nuclear Detonations, Dec. 1966, *Radiological Health Data*, vol. 8, no. 1, US Dept. of Health, Education and Welfare.

originally north-westerlies and these blow in from the higher latitudes. They are cold and fairly moist. But in their usually slow transit over the desert, they lose their moisture to higher levels of the atmosphere by convection. When they arrive over Northern Nigeria their relative humidity would have dropped to as low as 10–15 %. On the other hand, the maritime south-westerly wind blows from over the Atlantic Ocean, so that it is laden with moisture and its relative humidity is generally over 70 % corresponding to low specific volume of water vapour. The positions where these two winds meet form the intertropical line of discontinuity of humidity or of specific volume of water vapour. Hence the specific volume of water vapour can be employed to distinguish between winds which are predominantly north-easterly and south-westerly, a low specific volume of water vapour will then correspond to the south-westerly wind while a high specific volume of water vapour to the north-easterly wind.

(c) *Origin of peaks of radioactivity and their transportation to Ibadan*

Since from Section 4(b) it was concluded that the dryness of the wind was only indirectly responsible for periods of high specific radioactivity during the Harmattan, the strong correlation of high specific volume of water vapour and high specific radioactivity can now be interpreted as simultaneous presence of high specific radioactivity and the north-easterly wind. Hence the north-easterly wind might be responsible for transporting radioadtive dust to Ibadan. The plateau regions of Northern Nigeria, and Azelik, Arlit, Modovela and the Basin of Agada in the Niger Republic (Fig. 4) are recognised areas of high natural radioactivity due to the presence of Uranium, Thorium and other radioactive minerals. These might be sources of the radioactive dust measured at Ibadan. Convectional currents, sandstorm, and turbulence would play an important role in raising radioactive dusts to heights of 3 km which is the ceiling of Harmattan dust haze observed by aircrafts (Aina, 1970). In the plateau regions of Nigeria, where monazite and columbite mines are on the northern edge of the plateuau, orography might be important also. These dust particles mostly in the diameter range of 0.1–0.4 μm, once air-borne are transported southwards by the north-easterly wind during the Haramattan.

The highest correlation values from Fig. 3 are for time lags of 1–3 days. It is difficult to find an acceptable theory which can explain these time lags. This is a problem worthy of further investigation and maybe, the answer will enhance our understanding of the role of meteorology on atmospheric radioactivity.

Acknowledgement

The meteorological data for the analysis of these results were provided by the kind permission of the Director of the Meteorological Services of Nigeria.

REFERENCES

Agu, B. N. C. 1965. Observation of Radioactive fallout in Nigeria up to 1961. *Nature 205*, 649–651.

Aina, J. O. 1970. Private communication.

Fisher, Sir R. A. 1954. *Statistical methods for research workers*, Chap. VI, 12th ed. Oliver and Boyd, Edinburgh.

Israel, H. 1958. Die natürliche Radioaktivität in Boden, Wasser und Luft. *Beitr. Phys. Atmosphäre 30*, 177–188.

СЕЗОННЫЕ ВАРИАЦИИ АТМОСФЕРНОЙ РАДИОАКТИВНОСТИ В ИБАДАНЕ

Представлены результаты регулярного исследования проб воздуха на радиоактивность в Ибадане за период с июля 1966 г. по февраль 1967 г. Результаты указывают на общее увеличение удельной атмосферной радиоактивности и на специальные периоды высокой удельной радиоактивности в течение сезона Харматтана. Общее увеличение удельной атмосферной радиоактивности не коррелирует с испытаниями ядерного оружия и объясняется усилением эксхаляции эманации из земной коры. Показано что заметные пики удельной активности коррелируют с тяжелыми днями Харматтана, которые характеризуются высоким удельным содержанием водяного пара в атмосфере. Заметные пики удельной атмосферной радиоактивности в месяцы Харматтана приписываются континентальным северо-восточным ветрам, которые переносят радиоактивную пыль с радиоактивных полей республики Нигер и с областей плато Нигерии к станции наблюдения.