# Fission Product Gamma Activity with Northerly Winds and with Southerly Winds

Abstract. *The gamma ray activity of particulate matter filtered from air samples of northerly winds and of southerly winds has been studied during the period October 1961 to May 1963 with a scintillation spectrometer. It was found that the gamma activity at 0.75, 0.49, and 0.145 Mev was greater when the wind was southerly than when the wind was northerly during the months December through May; the opposite was true from June through September. The gamma ray activity was least during the summer of 1962, the decrease being more pronounced for southerly winds than for northerly winds. These results can be accounted for by modifying the Brewer-Dobson theory of stratospheric circulation so that during most of the year stratospheric debris first reaches the ground south of latitude 40°N. No diurnal effect on the gamma ray activity of the samples could be detected.*

It has been known for many years that the concentration of ozone in surface air varies seasonally, maximum concentrations being observed in early spring, minimum values in late fall, and the amplitude of the variation increasing with latitude (*1*, p. 279). More recently it has been observed that fission product radioactivity shows a similar springtime maximum (*2*). In addition to the seasonal variation, there are other ways in which the concentration of ozone varies with meteorological conditions. Paneth (*3*) and Dobson (*1*, p. 284) observed that invasions of polar air (northwesterly winds) are accompanied by an increase in ozone concentration, whereas the concentration is slight in the case of tropical air (southwesterly winds). Kroenig and Ney (*4*) have observed a strong diurnal variation of ozone near the ground, the night-time concentration being approximately one-thirtieth that observed during the day. The close similarity between the seasonal variations of ozone and fission product radioactivity suggested that the variation in the latter with wind direction, as well as its diurnal variation, should be investigated.

Samples of particulate matter in a measured volume of air were obtained by drawing air through HV-70 (Hollingsworth-Vose) filters. The air pump was rated at 10 ft$^3$ min (approximately 400 m$^3$ in 24 hours). Twenty-four hour samples were obtained on days of unusually high temperature (southerly surface winds) and of unusually low temperature (northerly surface winds). Samples were taken at irregular intervals from October 1961 to May 1963, with an average of about two samples per month. Care was taken to avoid periods of active precipitation and no sample was started until at least 24 hours had elapsed since active precipitation occurred locally. The filter paper was folded and inserted in a small test tube (6 mm inside diameter) which in turn was placed in the well of a sodium iodide crystal 3 inches (7.62 cm) in diameter by 3 inches thick. Scintillations produced by gamma rays and detected by a photomultiplier tube were analyzed for 200 minutes with a multichannel analyzer (*5*).

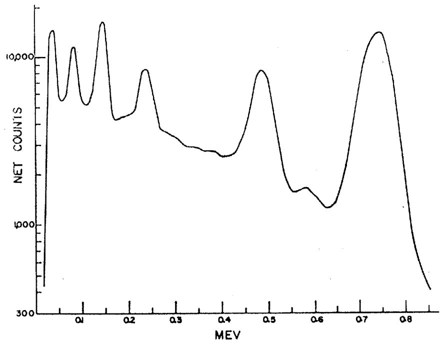

Fig. 1. Gamma ray spectrum of particulate matter in an air sample collected 27–28 March 1962.

Figure 1 shows the gamma ray spectrum of a sample collected 27–28 March 1962, with southerly winds. The peak at 0.24 Mev is due to thorium B and the peak at 0.08 Mev is the associated bismuth x-ray. These peaks can be eliminated if the analysis is delayed for several days until the thorium B has a chance to decay. The peaks at 0.75, 0.49, and 0.145 Mev are due to $Zr^{95}$–$Nb^{95}$, $Ru^{103}$, and $Ce^{141}$, respectively. The peak at 0.03 Mev is due possibly to the praseodymium x-ray associated with $Ce^{141}$. The spectra of samples collected during the period 15 September to 15 October 1962 showed additional peaks at 0.06, 0.10, 0.21, and 0.265 Mev (all unidentified and all with half lives of 6 to 9 days) as well as a peak at 0.36 Mev due to $I^{131}-Xe^{131}$. All of the unidentified peaks except the one at 0.10 Mev became very weak by the end of October 1962. The peak at 0.36 Mev due to $I^{131}-Xe^{131}$ was present in the spectra of samples taken during the fall of 1961 but was absent during the period 1 December 1961 to 1 August 1962 (except for one sample taken 9 July 1962). The peak reappeared in the spectrum of a sample taken 18 September 1962 and continued to appear during the fall of 1962 with moderate intensity when the wind was northerly and low intensity when the wind was southerly. According to a report by Isotopes, Inc. (*6*) the U.S.S.R. was responsible for a series of nuclear detonations during the period 1 September 1961 to 4 November 1961 and again from 5 August 1962 to 27 September 1962. It is interesting that the period during which the peak due to $I^{131}$ was not observed (1 December 1961 to 1 August 1962) coincides with the quiet period in the Russian detonations, and that the detonations initiated by the United States during the period 25 April 1962 to 17 July 1962 contributed no $I^{131}$ to the air samples here reported. This latter observation is consistent with the finding of Staley (*7*) that "debris in the troposphere from United States testing at 2°N spread mostly into the southern hemisphere."

During June and July 1962, on three occasions (two with northerly winds and one with southerly winds) under generally clear skies, two consecutive 12-hour samples were taken from 8 A.M. to 8 P.M. and from 8 P.M. to 8 A.M. No diurnal effect on the gamma ray activity of the samples could be detected.

A record was kept of the heights (above the Compton background) of the three prominent peaks at 0.145, 0.49, and 0.75 Mev. These values should be proportional to the concentration of $Ce^{141}$, $Ru^{103}$, and $Zr^{95}$, respectively (*8*). The data for $Ru^{103}$ are

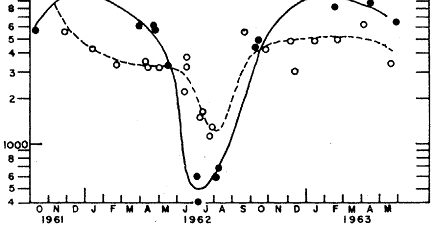

Fig. 2. Concentration of $Ru^{103}$ in northerly winds (dashed line) and in southerly winds (solid line).

shown in Fig. 2. Similar curves were obtained for $Ce^{141}$. The curves for $Zr^{95}$ differ from those in Fig. 2 in two respects: (i) the summer minimum for northerly winds occurs late in September rather than late in July, and (ii) the spring maximum for southerly winds occurs in April rather than in January. It would appear from Fig. 2 that during the summer months, the gamma ray activity of fission products varies with wind direction in a way similar to that observed for ozone by Paneth and by Dobson. It would be interesting to determine whether, during the winter months, the ozone concentration in tropical air was greater than that in polar air. The data of Fig. 2 are also consistent with results obtained by Damon and Kuroda (*9*) who observed that during June and July, 1953, the fission product radioactivity of polar air was considerably greater than that of tropical air.

It would appear that the seasonal variation in fission product radioactivity, previously discussed (*2*), is due primarily to the marked seasonal variation in the activity of southerly winds; this variation could be accounted for if it could be shown that air from the south was washed by precipitation more thoroughly during summer months. A study of precipitation patterns along a trajectory extending 700 miles (roughly 1100 km) upwind from Carbondale, Illinois (lat. 38°N) at the time these samples were taken indicates that rainfall pecularities cannot account for the variations shown in Fig. 2. A more plausible explanation is to be found in the stratospheric circulation model of Brewer and Dobson, as presented by Machta (*10*), with two modifications: (i) the equatorial updraft, spanning perhaps 40° of latitude, is assumed to move north and south with the sun, and (ii) an appreciable portion of the downdraft is assumed to be just north and south of the updraft. According to this picture, during the winter, when the sun is south of the equator, stratospheric air would reach latitude 40°N most directly from the south. During the summer, however, the center of the updraft will be appreciably north of the equator, so that the associated downdraft will be north of latitude 40°N, and the most active air will reach Carbondale, Illinois, from the north.

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### References And Notes

1. *Compendium of Meteorology* (American Meteorological Soc., Boston, 1951).
2. N. G. Stewart, R. G. D. Osmond, R. N. Crocks, E. M. Fisher, *At. Energy Res. Estab. (Gt. Brit.) Rept. AERE HP/R2354* (1957); P. K. Kuroda, *Argonne Natl. Lab. Publ. No. ANL-5920* (Oct. 1958), pp. 1–40; E. A. Martell, *Science* **129**, 1197 (1959); W. F. Libby, *Proc. Natl. Acad. Sci. U.S.* **45**, 959 (1959); P. K. Kiroda, H. L. Hodges, L. M. Fry, *Science* **132**, 742 (1960); P. F. Gustafson, S. S. Brar, M. A. Kerrigan, *ibid.* **133**, 460 (1961); J. F. Bleichrodt, J. Blok, R. H. Dekker, *J. Geophys. Res.* **66**, 135 (1961).
3. F. A. Paneth and E. Gluckauf, *Nature* **147**, 614 (1941).
4. J. L. Kroenig and E. P. Ney, *J. Geophys. Res.* **67**, 1867 (1962).
5. Dr. J. A. Eisele (now at Texas A&M College), J. D. Thompson, and H. D. Belt assisted in obtaining the gamma ray spectra reported here.
6. A. Walton and M. W. Leo, *Isotopes, Inc. NYO-9533,* pp. 45–50 (Nov. 15, 1962).

→ D. O. Staley, *Science* **140**, 667 (1963).

8. Dr. P. F. Gustafson of Argonne National Laboratory has kindly carried out the computations which show that, for these experiments, a peak height of 10,000 counts corresponds to an activity of $Ce^{141}$, $Ru^{103}$, and $Zr^{95}$ of $0.8 \times 10^{-12}$, $4.4 \times 10^{-12}$, and $5.3 \times 10^{-12}$ curies per cubic meter of air, respectively.
9. P. E. Damon and P. K. Kuroda, *Nucleonics* **11**, 59 (Dec. 1953).
10. L. Machta, *Advan. Geophys.* **6**, 276 (1959).

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## Permselective Membranes: Spectroscopic and Conductivity Effects

Abstract. *The absorption spectrum of permselective ion-exchange membranes containing positively charged groups shows specific effects which depend on the nature of the negative gegenion used. Such colored permselective membranes with positive or negative functional groups show increased conductivity after illumination with light flashes. The increased conductivity returns slowly to the original value.*

The nature of the anion influences the absorption spectrum of ion-pair complexes in homogeneous solution. There is a correlation for such complexes of one cation, between the properties of the anion—such as electron affinity or ionic radius—and the resulting spectrum in aqueous solution (*1*). Permselective ion-exchange membranes contain as their functional groups charged ions. There arises therefore the possibility to investigate through spectroscopic effects, the interaction between these charged functional groups located in the membrane, and gegenions adsorbed from solutions. We have chosen for our experiments permselective membranes (*2*) in which the active groups are positive quaternary ammonium groups produced by amination of polyvinyl chloride plastic membranes followed by quaternization. These membranes show deep reddish colors. We found that the absorption spectrum of these membranes in aqueous solutions is specifically affected by the nature of the anion used as the gegenion. For example, when the anion was changed in the series—fluoride, chloride, bromide, iodide—and, for comparison, perchlorate, the absorption edge in the red region of the spectrum changed with the change of halide in a regular manner. The membranes were permitted to come into equilibrium with saturated aqueous solutions of alkali halides, then rinsed thoroughly with distilled water, and the spectra were taken while the membranes were immersed in distilled water at *p*H 7. Thus only the halide gegenions bound to the membranes were retained. The wave length, $\lambda_{\frac{1}{2}}$, at which the transmittance of the membrane reaches 50 percent of its maximum, is 655, 648, 632, 622, and 640 m$\mu$ for the gegenions $I^-$, $Br^-$, $Cl^-$, $F^-$, and $ClO_4^-$, respectively. After equilibration in saturated aqueous NaCl, when the membranes were rinsed with distilled water and equilibrated with different solvents, a regular change in the absorption edge was observed, correlated with the polar and nonpolar nature of the solvent.

To examine the effect of illumination on the conductivity, we used membranes, in aqueous KCl, 0.1-mm thick with a surface area of approximately 0.5 by 3 cm. They were introduced into a quartz spectrophotometer cell and contact to either side of the membrane was through 0.02*M* KCl in capillary agar bridges. The bridges led to silver/silver chloride electrodes which were well protected against illumination. A bias of 0.5 volt (direct current) was applied to the circuit and the current was measured with a Metrohm Polarecord E 261 R instrument. As

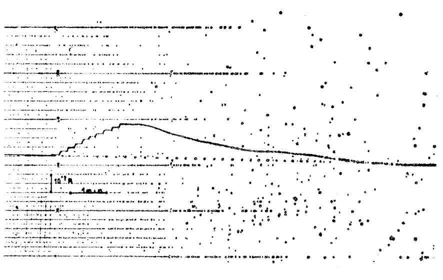

Fig. 1. Change in the current through the permselective membrane as a result of flash illumination.