

# Reports

## Air Concentrations of Twelve Radionuclides from 1962 through Mid-1964

*Abstract. New spectrometric techniques for the multidimensional counting of gamma rays permit the direct measurement of twelve radionuclides collected on air filters. Observed concentrations of Be<sup>7</sup>, Na<sup>22</sup>, Mn<sup>54</sup>, Co<sup>60</sup>, Y<sup>88</sup>, Zr<sup>95</sup>-Nb<sup>95</sup>, Ru<sup>106</sup>, Sb<sup>124</sup>, Sb<sup>125</sup>, Cs<sup>134</sup>, Cs<sup>137</sup>, and Ce<sup>144</sup> during the past 2½ years help to explain the origin and fallout rates of the trace radionuclides in air.*

The absolute and relative concentrations of all the various airborne radionuclides are of interest in studying their fallout rates and origin as well as in determining their availability for uptake by man. The tracing of some of the minor radionuclides in fallout, as well as the making of more precise measurements of the major radionuclides, has recently been made possible by use of multidimensional gamma-ray spectrometric counting methods (1). By the direct counting of a sample obtained on an air filter, it is now possible to make reasonably precise measurements of the 12 radionuclides Be<sup>7</sup>, Na<sup>22</sup>, Mn<sup>54</sup>, Co<sup>60</sup>, Zr<sup>95</sup>-Nb<sup>95</sup>, Y<sup>88</sup>, Ru<sup>106</sup>, Sb<sup>124</sup>, Sb<sup>125</sup>, Cs<sup>134</sup>, Cs<sup>137</sup>, and Ce<sup>144</sup>. Such measurements are especially valuable since they permit a direct comparison, on the same sample, of the relative concentrations of this large group of radionuclides and thus provide information on both fallout rates and origin.

The spectrometer system consists of a gamma-ray detector containing three NaI(Tl) crystals, which is used in conjunction with a 4096-channel multi-parameter analyzer. Use is made of the gamma-ray decay characteristic of each radionuclide for its identification and measurement by counting the sample between two detectors, each 10 cm thick and 15 cm in diameter, and cancelling with an anticoincidence annulus those events which are not totally absorbed in the two principal detectors. Coincidence counts are stored according to the two photon energies responsible for the event, while noncoincidence events are stored in the normal

manner. The effect of background radiation is reduced by the anticoincidence shielding, and the large, efficient detectors allow the sample to be viewed in a nearly 4 $\pi$  arrangement; thus the system is both very sensitive and selective. The radionuclides Na<sup>22</sup>, Co<sup>60</sup>, Y<sup>88</sup>, Ru<sup>106</sup>, Sb<sup>124</sup>, Cs<sup>134</sup>, and Ce<sup>144</sup> are measured from their characteristic coincidence gamma-ray spectra, while the remaining radionuclides are measured from their single photon gamma-ray spectra. The selective measurement of mono-gamma emitters is improved by use of a difference spectra, obtained from two measurements taken 3 to 6 weeks apart.

Air sampling (2) is now performed with a continuously operating vacuum pump which pulls air at 2.8 m<sup>3</sup>/min through a membrane filter of 5  $\mu$  pore-size. These membrane filters have been shown to be essentially absolute for fallout radionuclides (2). The filters are composited on a monthly or semi-monthly basis, pressed into a standard geometry of 1.25 cm thick by 2.50 cm in diameter, and counted on the multidimensional gamma-ray spectrometer. The air samples were obtained at a point 4.5 m above the ground, 9.5 km north of Richland, Washington. The annual precipitation in this area is only about 20 cm and dry deposition does account for a significant fraction of the fallout. The concentration in air of each radionuclide being studied is recorded in Fig. 1 (3).

The radionuclides listed in Fig. 1 result from fission and neutron activation during atomic tests, and also from cosmic ray interactions with the at-

mosphere. The radionuclides Zr<sup>95</sup>-Nb<sup>95</sup>, Ru<sup>106</sup>, Sb<sup>125</sup>, Cs<sup>137</sup>, and Ce<sup>144</sup> are fission products and presumably this is their main source in the atmosphere; Mn<sup>54</sup>, Co<sup>60</sup>, Y<sup>88</sup>, Sb<sup>124</sup>, and Cs<sup>134</sup> are formed as neutron activation products during testing, while Be<sup>7</sup> and Na<sup>22</sup> are produced continuously in the atmosphere by interactions of cosmic rays. The fallout rates of many of the fission products are measured from sampling locations the world over (4) and they serve as a base line for comparing the concentrations of less abundant radionuclides in the atmosphere.

The radionuclide Y<sup>88</sup> (105 days) was recently reported to be present on samples of grass collected in August 1962 near the Euratom Nuclear Center at Ispra, Italy (5). It was in the form of insoluble particles containing much larger amounts of Zr<sup>95</sup>-Nb<sup>95</sup> and, although its source was not known, it did not come from the Nuclear Center and presumably came from either nuclear bombs or an uncontrolled release from a nuclear establishment. More recent studies by high altitude sampling techniques (4) have shown that large amounts of Y<sup>88</sup> were produced during the U.S.S.R. tests in late 1962. We have detected the arrival of this Y<sup>88</sup> and measured its fallout rate. The Y<sup>88</sup> was not detectable in our air samples prior to November 1962. This may have been due, in part, to the fact that during 1962 smaller air volumes were collected and also, considerable decay occurred before the multidimensional spectrometric measurements were made in 1963. Yttrium-88 has been easily measurable during 1963 and 1964. The radionuclide Na<sup>22</sup> was first reported to be present in the atmosphere by Marquez (6) in 1957, who found it in rain water at Rio de Janeiro in a concentration of 0.017 disintegration min<sup>-1</sup> liter<sup>-1</sup>. Its current concentration in air, foods, and people will be reported later (7). It is produced naturally by cosmic ray spallation of argon in the atmosphere but also results from the reaction Na<sup>23</sup>(n,2n) Na<sup>22</sup> during tests of nuclear weapons. Its potential value as a tracer of atmospheric circulation has been recognized, and recently reported measurements for this purpose by Bhandari and Rama (8) have shown air concentrations ranging from 1 disintegration min<sup>-1</sup> 10<sup>-6</sup> ft<sup>-3</sup> below the tropopause to 460 disintegration min<sup>-1</sup> 10<sup>-6</sup> ft<sup>-3</sup> above the tropopause. It is evident from Fig. 1 that the Na<sup>22</sup> concentrations in air were higher by about a factor of 2 dur-

ing 1963 and 1964 than in 1962. Also, the 1963-64 values are about an order of magnitude higher than those reported for troposphere measurements prior to the atomic tests conducted in 1961 (8). This suggests that a large amount of  $\text{Na}^{22}$  was produced during the past series of tests.

The  $\text{Cs}^{137}$  appears to be produced in nuclear detonations mainly by an n,  $\gamma$  reaction on  $\text{Cs}^{135}$  but is also produced directly by fission (9). The ratio of

$\text{Cs}^{134}$  to  $\text{Cs}^{137}$  during the past 2½ years has not shown a pronounced drop; however, our biological studies (7) have shown that the ratio of these radionuclides in Alaskan caribou flesh dropped by a factor of 4 during the period from just prior to the last test series (October 1961) through July 1963. This suggests that a major injection of  $\text{Cs}^{134}$  into the atmosphere occurred prior to this last test series.

The radionuclides of  $\text{Co}^{60}$  and  $\text{Mn}^{54}$

were reported to be present in the atmosphere in 1958 by Marquez *et al.* (10) who found them in rain water collected between June 1957 and March 1958. Their presence was attributed to thermonuclear testing. It was reported recently (4) that a number of activation products including  $\text{Mn}^{54}$ ,  $\text{Fe}^{55}$ , and  $\text{Sb}^{124}$  were produced in relatively high abundance in the high yield detonations carried out by the U.S.S.R. at Novaya Zemlya in the fall of 1961 and that  $\text{Y}^{89}$

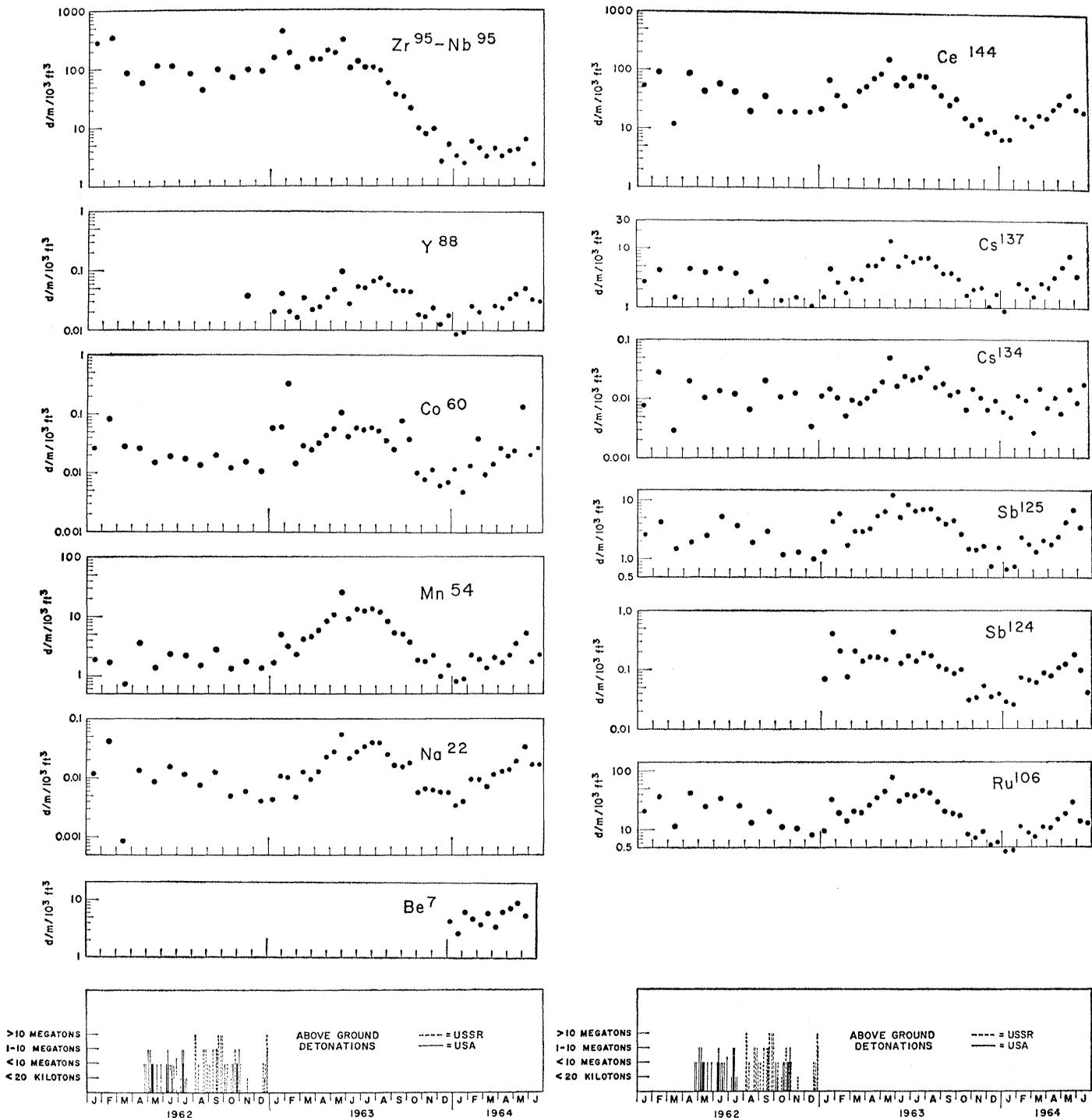


Fig. 1. Concentration of radionuclides in the atmosphere at Richland, Washington, during 1962-64.

and  $\text{Sb}^{124}$  were produced in U.S.S.R. tests in late 1962. These tests appear to be the major source of the  $\text{Mn}^{54}$ ,  $\text{Co}^{60}$ ,  $\text{Y}^{88}$ ,  $\text{Sb}^{124}$ , and some of the other activation products which are included in the study described here.

The  $\text{Be}^7$  is produced by cosmic spallation reactions in the atmosphere and its presence was first reported in the atmosphere by Arnold and Al-Salih in 1955 (11) who chemically separated it from rain water samples. Measurements of its concentration in the atmosphere at various altitudes and latitudes have since been reported (8, 12, 13). Only recently have we begun to study the concentration of  $\text{Be}^7$  in the atmosphere. It is interesting that, unlike other fallout radionuclides,  $\text{Be}^7$  did not show a large increase in concentration during the spring.

It is extremely interesting and significant that the activation products  $\text{Y}^{88}$  (105 days) and  $\text{Sb}^{124}$  (60 days) are at concentrations this year which are comparable with those 1 year ago, while the fission product  $\text{Zr}^{95}\text{-Nb}^{95}$  (65 days) is lower by about a factor of 20. Also, the fission product  $\text{Cs}^{137}$  (30 years) is at a concentration comparable with that last year and the  $\text{Ru}^{106}$  (1 year) and  $\text{Ce}^{144}$  (285 days) are at about half their last year's concentrations. Since there have been no atomic detonations above ground during 1963 and 1964 (14), all of these radionuclides entered the atmosphere prior to 1963 and their reservoir in the atmosphere has decreased with their respective half-lives since that time. This points to an obvious difference in the source of  $\text{Y}^{88}$  and  $\text{Sb}^{124}$  relative to most of the  $\text{Zr}^{95}\text{-Nb}^{95}$ , and the other fission products, and also allows one to estimate the rate of fallout of the radioactive material which was generated at the time  $\text{Y}^{88}$  and  $\text{Sb}^{124}$  were formed. A continuation of these measurements is expected to establish any other differences in the fallout rates and origin of the activation products.

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#### References and Notes

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### Component Shapes in Double Radio Sources

Abstract. *High-resolution interferometry at a wavelength of 10.6 centimeters shows that several well-separated double radio sources have small, bright regions near the outward ends of their components. This is consistent with an expanding model for the sources.*

One of the first results to emerge from the study of discrete sources with the Caltech interferometer was a recognition of the predominance of two-component sources (1). An initial survey of brightness distributions at 31-cm wavelength gave information about the approximate component sizes and spacings in a number of these objects (2, 3). More recent observations, made with the higher resolution available when the Caltech interferometer is operated at 10.6-cm wavelength, have revealed some additional properties which seem to be shared by the well-separated double sources. As is true in many other respects, Cygnus A seems to be a good prototype.

Lequeux (4), in 1962, reported high-resolution interferometric observations of Cygnus A which showed that the two components of the source were elongated in the direction of their separation and that they were more or less sharply peaked at their outward ex-

tremes. This peaking was evident from a decrease, at large antenna spacings, of the period of the oscillations in the observed visibility amplitude. Thus the effective separation of the two components seemed to increase with increasing resolution. This feature has been found common to three additional sources in the work reported here.

In Cygnus A the situation is further complicated because the relative importance of the bright peaks and the less-bright, bridging emission varies with wavelength, as was pointed out by Lequeux (4) and emphasized by Swarup, Thompson, and Bracewell (5). Thus the effective separation of the components is a function of degree of resolution and of wavelength, whether measured interferometrically (2, Table 3) or with a narrow fan beam (5, 6). The only invariant dimension for Cygnus A is the limiting apparent separation, of 115 seconds of arc along the major axis, which must correspond to the separation of the sharp peaks at the outward edges of the two components.

I have recently observed Cygnus A and several additional double sources using interferometer spacings of up to 6500 wavelengths, with baseline azimuths of  $0^\circ$ ,  $90^\circ$ , and  $135^\circ$  (Table 1). In each case the components are found to be elongated in the direction of the major axis, and in each case the effective separation increases with higher resolution (7).

Because of the small size of its com-

Table 1. Properties of double radio sources.

| Position angle of major axis (deg) | Apparent separation of components (sec) |                    | Dimensions of component (sec) |
|------------------------------------|---|--------------------|-------------------------------|
|                                    | At low resolution                       | At high resolution |                               |
| 19                                 | Source: 3C 33                           |                    | 16 by 8                       |
|                                    | 215                                     | 250                |                               |
| 175                                | Source: 3C 134*                         |                    | 60 by 30                      |
|                                    | 120                                     |                    |                               |
| 101                                | Source: Hercules A                      |                    | 51 by 27                      |
|                                    | 118                                     | 145                |                               |
| 84                                 | Source: 3C 353                          |                    | 84 by 42                      |
|                                    | 150                                     | 230                |                               |
| 109                                | Source: Cygnus A                        |                    | 25 by 15                      |
|                                    | 85-100†                                 | 115                |                               |

\* Data taken from Maltby and Moffet (2).  
† Varies with wavelength.