# Transport of radioactive aerosols across the trade wind inversion at Hawaii

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ABSTRACT

Radioactivity concentration measurements in aerosol and precipitation samples collected along the slopes of Mauna Loa volcano in Hawaii have been examined in relation to vertical profiles of temperature and humidity in the atmosphere across the trade wind inversion. The gradient of radioactive aerosol concentration in the air with height is related to the humidity profile and the character of the trade wind inversion. The concentrations increased with height across the trade wind inversion in contrast to aerosols of terrestrial origin whose concentrations in air decreased with height. The concentrations in rainfall generally decreased with height, except for an anomolous result at 2250 m, just below the trade wind inversion. At this altitude, the gross radioactivity concentration increased while the $Sr^{90}$ concentration decreased. The ratio of the gross beta concentration in air and rain gives a parameter with dimensions of mixing ratio, which may be of potential use in determining rainout efficiencies.

## Introduction

The effect of temperature inversions on the deposition of radioactive aerosols from the atmosphere has been examined by Kruger (1966), and the results of initial studies of transport of radioactivity in rain and air across the trade wind inversion during short periods in 1963 and 1964 were described by Kruger & Miller (1966). In these studies, the concentrations of radioactive aerosols were measured as a function of time and altitude along the north-east slope of Mauna Loa volcano (the direction of the prevailing trade winds) in both air and rainfall samples. The data were examined in relation to time sections of the humidity and temperature profiles obtained from radiosonde data at Hilo, Hawaii. From these studies it was noted that the vertical distribution of radioactivity concentration in the air oscillated in relation to the oscillations of the moisture mixing ratio. The drier air accompanying the lowering of the upper boundary of the moist layer generally contained greater radioactivity concentrations. These oscillations appeared to occur periodically and may be related to the generally-strong diurnal variation of the moisture content of the air along the slopes of Mauna Loa described by Price & Pales (1963).

The $Sr^{90}$ concentration in the rain collected along the slope generally decreased with altitude above the cloud base. Kruger & Miller (1966) attributed this to the continuous depletion of condensation nuclei by rain while remaining droplets in the air parcels continue to grow as they move upslope. This observation was in accord with measurements of other aerosol constituents in trade wind air, such as sea salt as noted by Woodcock (1957) and the halogens as noted by Duce, Winchester & Van Nahl (1965). However, sea salt, the halogens, and water vapor are of terrestrial (primarily oceanic) origin, while nuclear debris is of stratopheric origin, residing on aerosols of unknown character. Therefore it is not evident that the condensation nuclei of these divergent sources should have similar rainout or washout properties.

The massive size of Mauna Loa volcano affects the general circulation of the trade winds. Thus in study of the concentrations and transport of aerosol constituents in the atmosphere along the slopes, three "air motions" should be considered. These are the general circulation of the trade winds, the diurnal variations caused by the mountain, and the vertical oscillations of the trade wind inversion. The general characteristics of the north-east trade winds have been de-

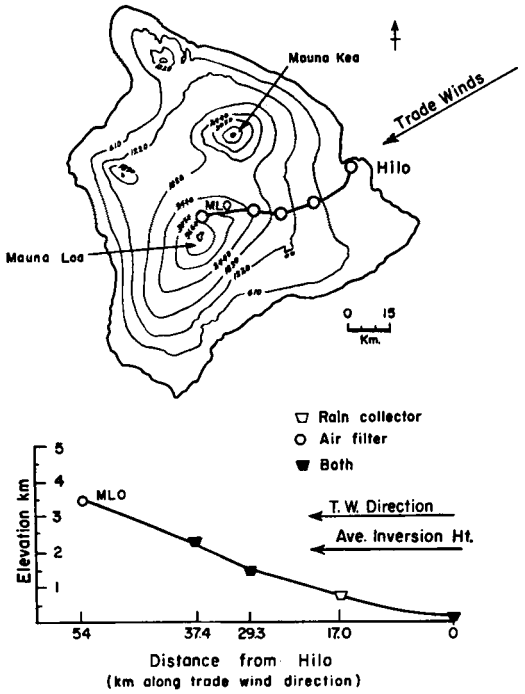



*Fig. 1.* The Mauna Loa volcano, Hawaii collection network.

scribed by Riehl *et al.* (1951). Downward transfer of mass through the inversion was shown to take place, presumably carrying with it the radioactive aerosols. Convective activity, through which cumuli penetrating the inversion layer carry moisture upward, was shown to be an adequate mechanism for the downstream rise and dissolution of the inversion. The diurnal circulation pattern of air along the slopes of Mauna Loa has been described by Price & Pales (1963). Moist air flows upslope during the day from the north and dry air descends during the night. Thus the moisture and radioactivity concentrations in the air near the slopes should be affected by this diurnal circulation. Finally, the extent of eddy diffusion and mixing across the inversion should depend upon the character of the inversion; such characteristics including the strength, height, and thickness of the inversion, and its continuity and lifting as the trade wind air moves inland.

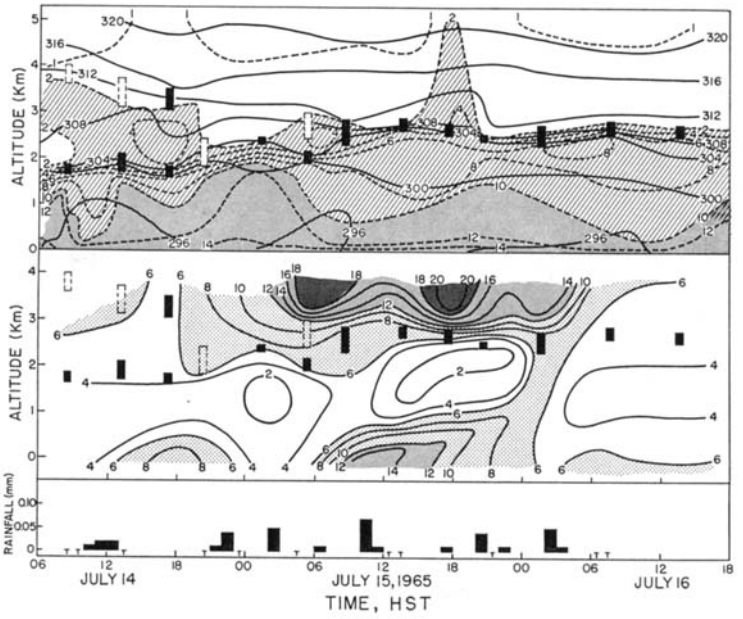
During a two-week period in July 1965 coincident with the period of the U.S.–Japan cooperative program on the Warm Rain Project in Hawaii (Lavoie, 1966) additional sampling of aerosols and precipitation was made along the network of sites on the north-east slope of Mauna Loa. The radiochemical data of these samples were examined in conjunction with the meteorological data obtained during the study period to investigate the concentration changes and transport of the radioactive aerosols.

## Results

The network of air and rain sampling sites along the Mauna Loa volcano is shown in Figure 1. This network is identical to that described by Kruger (1966) except that an additional air sampler was included at the 2250 m site. Thus, the 3.62 $m^2$ stainless steel rain funnels were located at altitudes of 10, 750, 1500, and 2250 m; and the Staplex air samplers (with flow rates of about 1 $m^3$/min through 8″ × 10″ No. 41 filter paper) were located at 10, 1500, 2250, and 3400 m. The air samplers were calibrated by a method given by Harding & Hendrickson (1964). During the sampling period from July 14–28, 1965, a total of 205 aerosol samples was collected at the four sites, and during the same period, the only rainfall under normal trade wind conditions occurred during a 24-hour period on July 15–16. A total of 43 rainshower samples was collected at the four sites. A small aliquot of each of these samples was chemically analyzed for sodium and chloride concentrations at the University of Hawaii in Hilo[1] by flame photometry and Mohr titration, respectively. The aerosol samples were analyzed for gross beta radioactivity and the rain samples for gross beta radioactivity and for $Sr^{90}$ content by the standard low-level radiochemical techniques summarized by Kruger & Miller (1966).

A time section of the temperature and humidity profiles, to a height of about 5 km, was prepared from the 47 radiosonde ascent data compiled during the 15-day collection period. The gross beta radioactivity concentration of the filtered aerosols was also prepared as time sections. These two sets of time sections were divided for convenience into four periods, shown in Figures 2–5. In each of these figures, the upper time section shows the potential temperature (°K), the mixing ratio (g/kg), and the height of the base and top of the temperature inversions and isothermals. The lower time section shows

[1] Through the courtesy of R. A. Duce and W. Zoller of the Hawaii Institute of Geophysics.




*Figs. 2–5.* Time sections of the meteorological and radioactive aerosol concentrations in the atmosphere during July 14–28, 1965, in Hawaii. The *upper time* sections show: ——, potential temperature, °K; – – – –, mixing ratio, g/kg; ■, heights of base and top of inversions; □, heights of base and top of isothermals. The *lower time* sections show: gross beta radioactivity concentration, pC/100 SCM; inversion and isothermals, as above.

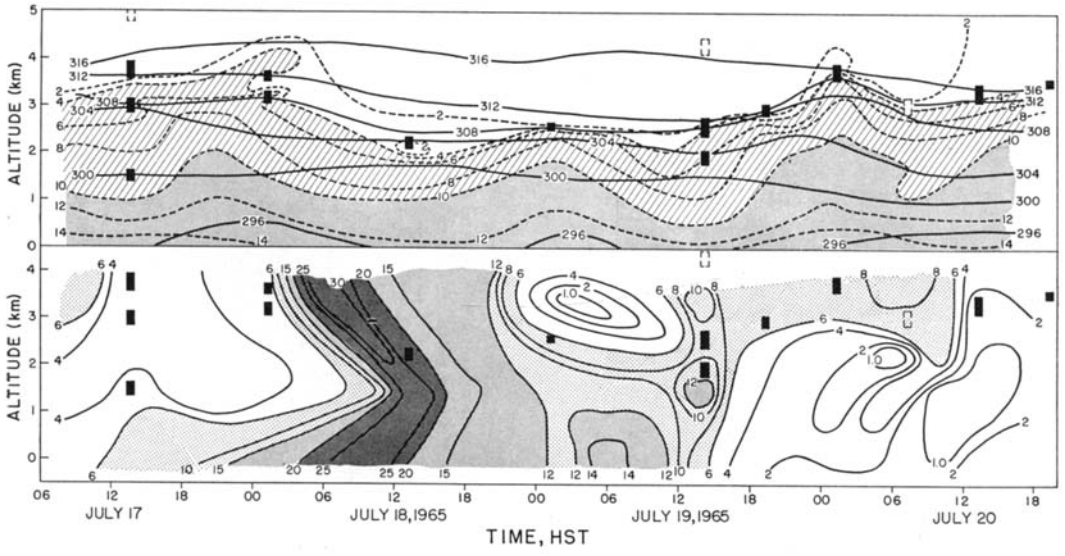



Fig. 3.

the gross beta radioactivity concentration (pC/100 SCM) and the same inversion data. Shadings are used to accentuate the moisture profiles in the upper part and the radioactivity concentration gradients in the lower part. Figure 2 shows the time sections for July 14–16, Figure 3 for July 17–20, Figure 4 for July 20–22, and Figure 5 for July 26–28, 1965.

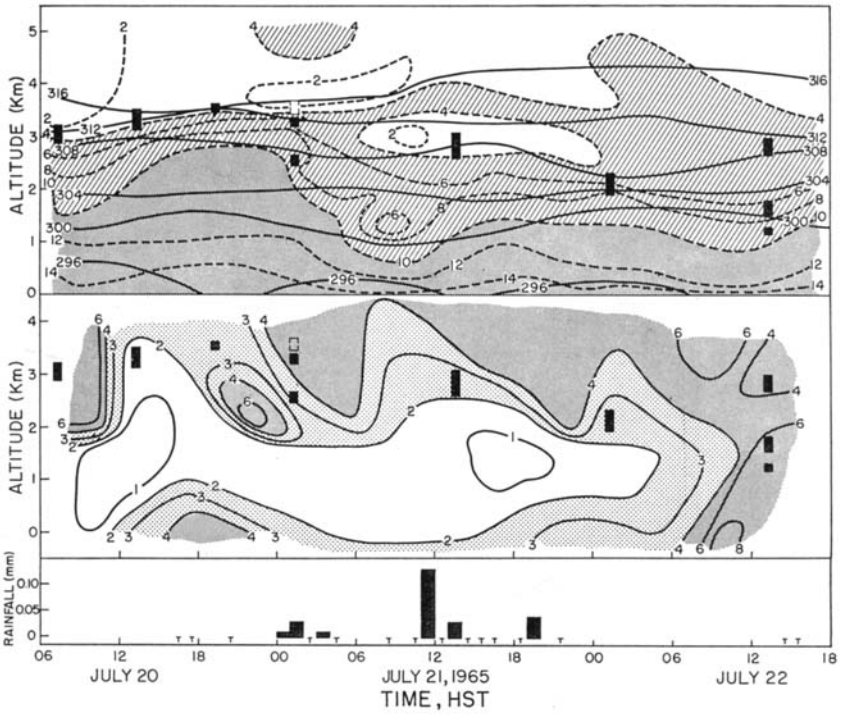



Fig. 4.

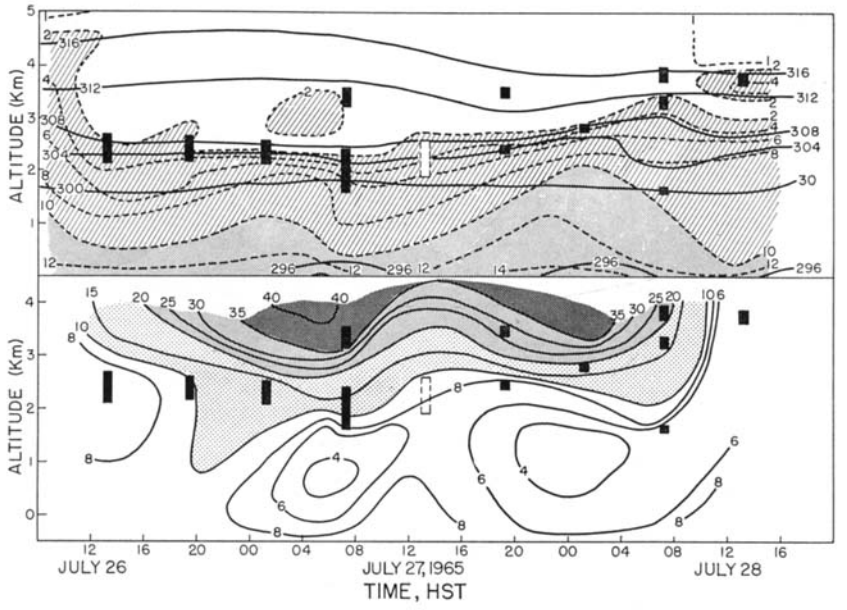



Fig. 5.

Where significant, the hourly rainfall recorded at the Hilo weather station is shown at the bottom of the figures. For the collection period of July 17–20, traces were recorded for the hours 0000–0100 HST on July 19, and 1600–1700, 1700–1800, and 2000–2100 HST on July 20.

Table 1. *Collection and radiochemical data trade wind showers, Hawaii, July 15–16, 1965*

| Altitude (meters) | Collection period (HST) Day | From | To | Precipitation (mm) | Concentrations[a] $Na^+$ (ppm) | $Cl^-$ (ppm) | $\beta$ (pC/l) | $Sr^{90}$ (pC/l) | Ratios $Sr^{90}/Na^+$ | $Sr^{90}/\beta$ |
|---|---|---|---|---|---|---|---|---|---|---|
| 10 | 15 | 0615 | 0618 | 0.95 | 1.9 | 2.1 | 51 | 2.7 | 1.4 | 0.05 |
| | | 0618 | 0621 | 0.98 | 1.1 | 2.5 | 24 | 2.4 | 2.2 | 0.10 |
| | | 0639 | 0643 | 0.15 | 2.1 | 5.0 | 128 | 6.9 | 3.3 | 0.05 |
| | | 1034 | 1114 | 0.92 | 5.8 | 0.7 | 38 | 2.3 | 0.4 | 0.06 |
| | | 1311 | 1319 | 0.11 | 15.0 | 23.4 | 278 | 24.7 | 1.6 | 0.09 |
| | | 1716 | 1722 | 1.08 | 4.0 | 6.0 | 115 | 6.8 | 1.7 | 0.06 |
| | | 1722 | 1724 | 0.26 | 3.0 | 4.6 | 76 | 6.1 | 2.0 | 0.08 |
| | | 1743 | 1747 | 0.03 | 8.9 | — | 116 | 14.3 | 1.6 | 0.12 |
| | | 2058 | 2117 | 0.82 | 3.0 | 4.3 | 87 | 7.3 | 2.4 | 0.08 |
| | | 2221 | 0324 | 0.80 | 4.3 | 6.0 | 135 | 11.4 | 2.7 | 0.08 |
| | 16 | 0636 | 0713 | 0.72 | 2.4 | 3.2 | 50 | 5.0 | 2.1 | 0.10 |
| 750 | 15 | 0559 | 0729 | 0.42 | 1.3 | 1.4 | 49 | 5.1 | 3.9 | 0.10 |
| | | 0729 | 0802 | 0.24 | 0.9 | 0.7 | 39 | 3.9 | 4.4 | 0.10 |
| | | 0802 | 0920 | 1.19 | 0.7 | 0.7 | 44 | 3.2 | 4.6 | 0.07 |
| | | 1034 | 1044 | 0.26 | 3.0 | 5.3 | 111 | 10.3 | 3.4 | 0.09 |
| | | 1050 | 1204 | 1.54 | 0.4 | 0.3 | 43 | 3.9 | 9.6 | 0.09 |
| | | 1247 | 1311 | 1.07 | 2.1 | 3.9 | 72 | 8.3 | 3.9 | 0.12 |
| | | 1404 | 1524 | 0.92 | 2.0 | 3.9 | 67 | 7.9 | 3.9 | 0.12 |
| | | 1554 | 1626 | 0.16 | 5.2 | 8.5 | 159 | 16.0 | 3.1 | 0.10 |
| | | 1811 | 1843 | 0.33 | 4.2 | 7.8 | 99 | 10.8 | 2.6 | 0.11 |
| | | 2200 | 2230 | 0.19 | 3.4 | 5.0 | 68 | 9.9 | 2.9 | 0.15 |
| | | 2230 | 2330 | 0.94 | 1.2 | 1.1 | 39 | 5.6 | 4.7 | 0.15 |
| | | 2330 | 0430 | 0.52 | 1.7 | 1.8 | 46 | 5.9 | 3.5 | 0.13 |
| | 16 | 0430 | 0500 | 0.60 | 1.0 | 0.7 | 41 | 5.7 | 5.7 | 0.14 |
| 1500 | 15 | 1105 | 1205 | 0.10 | 1.6 | 0.7 | 141 | 10.1 | 6.3 | 0.07 |
| | | 1205 | 1234 | 0.54 | 0.3 | 0.3 | 37 | 2.3 | 7.7 | 0.06 |
| | | 1234 | 1407 | 0.11 | 0.3 | 0.3 | 66 | 5.1 | 17 | 0.08 |
| | | 1407 | 1504 | 0.39 | 0.2 | 0. | 32 | 2.1 | 10 | 0.07 |
| | | 1504 | 1605 | 0.30 | 0.3 | 0.3 | 29 | 3.2 | 11 | 0.11 |
| | | 1605 | 1702 | 0.14 | 0.6 | 1.0 | 25 | 5.9 | 9.8 | 0.23 |
| | | 1702 | 1802 | 0.35 | 0.6 | 0.7 | 37 | 4.2 | 6.9 | 0.11 |
| | | 1802 | 1900 | 0.22 | 0.8 | 0.3 | 28 | 3.5 | 4.3 | 0.12 |
| | | 1900 | 2004 | 0.53 | 0.3 | 0.3 | 24 | 3.0 | 9.9 | 0.12 |
| | | 2004 | 2058 | 0.24 | 0.5 | 0.3 | 31 | 5.3 | 11 | 0.17 |
| | | 2059 | 2136 | 0.10 | 0.6 | 0.7 | 30 | 6.0 | 10 | 0.20 |
| | | 2300 | 2357 | 0.02 | 0.7 | — | 41 | 7.5 | 11 | 0.18 |
| | | 2358 | 0101 | 0.38 | 0.1 | 0 | 17 | 2.3 | 23 | 0.14 |
| | | 0101 | 0620 | 0.35 | 0.1 | 0.3 | 17 | 2.5 | 25 | 0.15 |
| 2250 | 15 | 1855 | 1930 | 0.12 | 0.4 | 0.3 | 176 | 2.3 | 5.8 | 0.01 |
| | | 1930 | 2030 | 0.23 | 0.3 | 0.3 | 24 | 0.7 | 2.5 | 0.03 |
| | | 2030 | 2145 | 0.12 | 0.1 | 0 | 11 | 0.8 | 8.2 | 0.07 |
| | | 2330 | 0030 | 0.08 | 0.6 | 0.3 | 87 | 4.2 | 7.0 | 0.05 |

[a] The sodium and chloride analyses were made by R. A. Duce and W. Zoller, Hawaii Institute of Geophysics, at the Hilo Campus.

During the period July 26–28, the only rainfall recorded at Hilo was 0.76 mm for 0800–0900 HST on July 26 and 0.25 mm for 2200–2300 and a trace for 2300–2400 HST on July 27.

The collection and radiochemical data for the rainfall samples of the July 15–16, 1965, rain-shower period are given in Table 1. From repeated analysis of standards, the probable error of the individual gross beta concentrations is estimated to be $\pm 15\%$, and of the $Sr^{90}$ concentrations, $\pm 10\%$.

## Radioactive aerosol concentration in air

Figures 2–5 exhibit the same general correlation between changes in the vertical profile of

moisture and aerosol concentrations noted in the earlier studies. The heavier shading of mixing ratios of 10 or more g/kg points out the depletion of radioactive aerosols at the mid-altitudes at almost every occasion when the 10 g/kg isopleth rose to mid-altitudes. It is noted that the height of the 2 g/kg isopleth may be taken as the approximate upper boundary of the moist layer, and is generally in a zone of the steep mixing-ratio gradient characterized by being in or near the inversion zone, except for the period around July 21, when the high-level storm occurred. In general, the oscillations of the mixing ratio gradient reflect the diurnal variations of moisture in the air (e.g. as also shown by Lavoie (1966) for the period July–August, 1965). However, examination of specific 24-hour periods indicate that the observed oscillations in the gradient of the radioactive aerosol concentration in the air are not due solely to diurnal variations.

The July 14–16 period (Figure 2) is of special interest because it included the 24-hour period of rainfall sampled during the two-week collection program. The sea-level values for the radioactive aerosol concentrations do not correlate with the amount of rainfall; e.g. rainfall occurred during both the hours coinciding with the low concentration value of 4 pC/100 SCM and the high value of 14 pC/100 SCM. However, the 3 high value periods at sea-level do coincide with the relative drying of the lower levels, as seen by the lowering of the 10 g/kg isopleth at 1800 HST on July 14, at 1000 HST on July 15, and the morning of July 16. The two peaks of the 10 g/kg isopleth at about 0000 HST and 1800 HST on July 15 coincide with the two lows in aerosol concentration of 2 pC/100 SCM at about 2 km. Finally, the two rises of the 2 g/kg isopleth above the trade wind inversion, moderate at about 0600 HST and significant at about 1800 HST on July 15, coincide with the two highs in concentration of 18 to 20 pC/100 SCM at the Mauna Loa Observatory (3400 m).

The July 17–20 period (Figure 3) did not include any significant rainfall at Hilo. Yet the structure of the trade winds as noted by changes in the temperature inversion characteristics was very complicated compared to the earlier period of July 14–16. The rise of the 10 g/kg isopleth at about 2000 HST on July 17 appears to be accompanied by the depletion of aerosol to below 4 pC/100 SCM at mid-altitudes, but the drying of the upper air after 0400 HST on July 18 was accompanied by what appears to be a narrow tongue of air, containing a radioactive aerosol concentration of over 30 pC/100 SCM, all the way down to sea-level. The rise in moisture profile at about 0200 HST on July 19 shows a continuously negative gradient of the radioactive aerosol with altitude from a high of over 14 pC/100 SCM at sea-level to less than 1 pC/100 SCM at 3 km. The drying period at about 1400 HST on July 19 suggests turbulent mixing across the double inversion at that time with generally uniform concentrations of 10–12 pC/100 SCM at all altitudes. Accompanying the rising moist period at 0000 HST on July 20 is the reestablishment of the low aerosol concentration of 1–2 pC/100 SCM at mid-altitudes, which extends down to sea-level.

The July 20–22 period (Figure 4) is difficult to characterize definitively because of the severe disturbances above the trade wind inversions. The rise of the 10 g/kg isopleth to a height of over 2.5 km around 2000 HST on July 20 was accompanied by the almost complete removal of radioactive aerosol at mid-altitudes. Several arrivals of air containing radioactive aerosol with concentrations greater than 4 pC/100 SCM seems to have occurred during this period. It is possible that the convective activity associated with the upper-level air is responsible for these changes. The larger rainfall at Hilo during the hour 1100–1200 HST on July 21 corresponds with the lowering of the 10 g/kg isopleth to under 1 km and the aerosol concentration to less than 4 pC/100 SCM at all altitudes.

The July 26–28 period (Figure 5) was characterized by a return to normal trade wind conditions with a strong steady inversion averaging between 2200 and 2600 meters. The two waves of the 10 g/kg isopleth peaking at about 0200 HST both on July 27 and July 28 were accompanied by the two lows in radioactivity concentration of less than 4 pC/100 SCM down to sea-level, but above the inversion, the air contained concentrations of radioactive aerosols in excess of 40 pC/100 SCM, the largest concentrations observed during the entire collection period. The lack of rainfall during this 48-hour period and stability of the trade wind inversion allowed an examination of the validity of the observation by Kruger & Miller (1966) that a gradient of radioactivity concentration existed across the inversion. The data for the period July 10–13, 1963 (Kruger & Miller, 1966) are given in

Table 2. *Comparison of vertical gradient of radioactive aerosol across the trade-wind inversion*

| Collection altitude (m) | Gross Radioactivity Concentration in Air (pC/100 SCM) | | |
|---|---|---|---|
| | July 10–13, 1963 (trace rainfall) | July 15–16, 1965 (showers) | July 26–28, 1965 (no rainfall) |
| 10 | 23 | 9.4 | 7.7 |
| 1500 | 47 | 3.8 | 6.3 |
| 2250 | N.A. | 4.9 | 9.2 |
| 3400 | 107 | 16.4 | 26.5 |

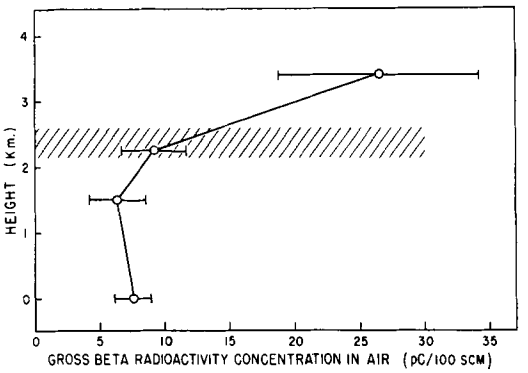



*Fig. 6.* Gross beta radioactivity profile as a function of altitude during July 26–28, 1965, at Hawaii. The average base and top of the trade wind inversion are given by the hatched lines across the figure.

Table 2, together with the data for the periods of July 15–16, and July 26–28, 1965. The data for the shower period of July 15–16 are discussed in the next section; the data for the period of July 26–28 are shown in Figure 6. These data show a steep gradient across the trade wind inversion. The air filter site at 2250 m was not available in 1963. The ratio of concentrations at 3400 m (at a height above the inversion) to that at 1500 m (at a height below the inversion) was about 2.4. The corresponding ratio for July 26–28, 1965, was 4.2, while the ratio of the 3400 m to the 2250 m site (at the base of the inversion) was still 2.9. The ratios for the shower period of July 15–16 are in remarkable agreement with the dry period of July 26–28, the respective values being 4.3 and 3.3.

The two steady inversion conditions during the sampling periods in 1963 and 1965 enabled an estimate to be made of the change in radioactivity concentrations in the air at Hawaii. The data obtained at the 3400 m site, which generally is above the average height of the top of the trade wind inversion, may be considered to be representative of the free air in the lower to middle troposphere. The radioactive aerosol at this altitude is usually assumed to be nuclear weapons debris of stratospheric origin. Feely *et al.* (1966) have shown that the stratospheric burden of $Sr^{90}$ and other particulate nuclear debris from the 1961–62 low altitude nuclear weapons tests has decreased during the period from 1963 to 1965 with a half-residence time of about 10 months. The average concentration of radioactive aerosols at the 3400 m site during July 10–13, 1963, was 107 pC/100 SCM. The concentration for the two periods July 14–16, and July 26–28, 1965, when the trade wind inversion was strong and persistent, had an average value of 18.1 pC/100 SCM. These two values, taken 24 months apart, may be used to estimate the half-residence time, assuming that the contribution of gross radioactivity from other sources (e.g. decay products of $Rn^{222}$, regenerated fallout, etc.) are small. The combined removal from the stratosphere and decay of the radioactivities is approximated as a first order process in which the half-residence time $R_{\frac{1}{2}}$ is given by

$$R_{\frac{1}{2}} = \frac{t \ln 2}{\ln (C_0/C)} = \frac{24 \times 0.693}{\ln (107/18.1)}, \quad R_{\frac{1}{2}} = 9.4 \text{ months},$$

in agreement with the measurements of Feely *et al.* (1966), and in support of the assumption that the radioactive aerosol at 3400 m is indeed associated with nuclear weapons debris from the stratosphere.

## Aerosol constituents in the rain showers of July 15–16, 1965

$Sr^{90}$ concentrations were measured by Kruger & Miller (1966) for the 1963 and 1964 rainfall collections and by grouping the shower periods by time as the showers moved upslope, they noted that the mean $Sr^{90}$ concentration in the rain decreased with altitude, even though the radioactive aerosol concentration in the air increased with altitude. The $Sr^{90}$ concentration values for the total rainfall of August 8–9, 1964, were 2.7 pC/l at 10 m, 1.5 pC/l at 750 m, 0.7 pC/l at 1500 m, and 0.4 pC/l at 2250 m. This decrease in concentration was observed to be

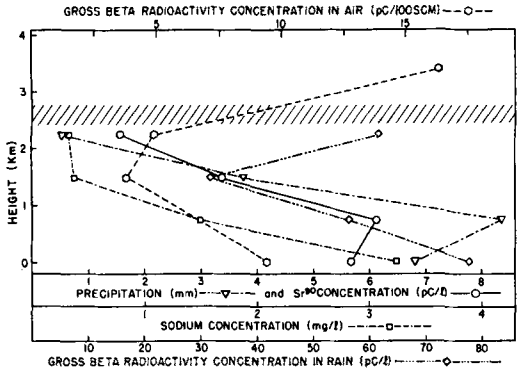



*Fig. 7.* Precipitation and aerosol data for the shower period of July 15–16, 1965, along the slopes of Mauna Loa volcano in Hawaii. The average base and top of the trade wind inversion are given by the hatched lines across the figure.

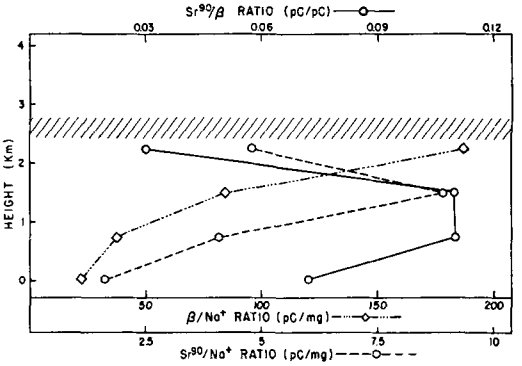



*Fig. 8.* The vertical profiles of aerosol concentration ratios in the rain showers of July 15–16, 1965, along the slopes of Mauna Loa volcano in Hawaii. The hatched lines represent the trade wind inversion.

consistent with the observations of others for terrestrial-origin aerosols (e.g. the chloride concentration as given by Duce *et al.*, 1965).

During the 2-week collection period from July 14 to July 28, 1965, orographic rainfall occurred solely during the period July 15–16. The hourly rainfall record at Hilo, Hawaii, was given in Figure 2; the total rainfall collected was only 6.8 mm. The data for the individual samples (Table 1) show the marked showery nature of the rainfall, several total rain showers at Hilo over 1 mm occurring in rainfall durations of less than 10 minutes. The rainfall at the upper locations were of somewhat more steady rates, although much of the time, the rainfall was hardly more than a drizzle. The periods of rainfall collections at the four altitudes appear to be sufficiently scattered in time, that it has not been possible to follow any of the showers upslope.

In order to compare the rainout of the terrestrial and radioactive aerosols, the data for the showers at each altitude were combined into average values. The total precipitation and the average concentrations of sodium (also indicative of chlorine), of gross beta radioactivity, and of $Sr^{90}$ (listed in Table 3), are shown in Figure 7 together with the gross beta radioactivity concentration in the air and the heights of the base and top of the trade wind inversion during the precipitation period.

The precipitation shows a maximum amount at the 750 m collection site. Rainfall records for Hawaii show that 750 m is at about the altitude of the greatest annual rainfall. The sodium concentration decreases from its maximum value of 3.5 ppm at sea level to about 0.3 ppm at the 2250 m site, also as expected. However, the gross beta concentration and the $Sr^{90}$ concentration show divergent behavior at the upper altitudes, the $Sr^{90}$ decreasing continuously from its maximum value at 750 m to the highest collection site at 2250 m, while the gross beta radioactivity concentration decreases from sea level to the 1500 m collection site, and then increases at the 2250 m site. The $Sr^{90}$ concentration changes in a manner similar to that observed in the 1963 and 1964 studies (Kruger & Miller, 1966) in that in about half of the shower periods, the maximum concentration occurred at 750 m (when it was not at sea level) and in all cases decreased steadily from the maximum up to the 2250 m site. It was suggested in these studies, that the mean $Sr^{90}$ concentration in rain should decrease with altitude, even though the radioactive aerosol concentration increased with altitude. The explanation offered was that the concentration of cloud droplets decreases while the average size of the droplets increases with height above the cloud base. However, this explanation assumed that little or no condensation nuclei from above the inversion were entrained to supplement the gradually-decreasing original condensation nuclei concentrations. The gross beta radioactivity in the rain (which was not measured during the prior studies) did not

Table 3. *Data for the trade wind showers of July 15–16, 1965, at Hawaii by collection altitude*

| Collection altitude (m) | Precipitation (mm) | Concentrations | | | Ratios | | |
|---|---|---|---|---|---|---|---|
| | | $Na^+$ (ppm) | $\beta$ (pC/l) | $Sr^{90}$ (pC/l) | $\beta/Na^+$ (pC/mg) | $Sr^{90}/Na^+$ (pC/mg) | $Sr^{90}/\beta$ (pC/pC) |
| 10 | 6.82 | 3.49 | 77.9 | 5.68 | 21.8 | 1.63 | 0.07 |
| 750 | 8.38 | 1.5 | 56.3 | 6.13 | 37.5 | 4.09 | 0.11 |
| 1500 | 3.77 | 0.38 | 31.9 | 3.38 | 83.9 | 8.89 | 0.11 |
| 2250 | 0.55 | 0.33 | 61.7 | 1.58 | 187 | 4.80 | 0.03 |

follow this trend. The ratios of the concentrations of the three aerosols, given in Table 3, are shown as a function of altitude in Figure 8.

On the basis of sodium being an aerosol of terrestrial origin and the gross beta radioactivity primarily of stratospheric origin, the steadily increasing ratio of $\beta/Na^+$ appears to be reasonable. However, the $Sr^{90}/Na^+$ ratio at the 2250 m site indicates a much steeper negative gradient of $Sr^{90}$ with altitude from 1500 m to 2250 m compared to that of sodium. Even more unusual is the change in ratio of $Sr^{90}/\beta$, which suggests that the rainfall at the two mid-altitude sites is enriched in $Sr^{90}$ aerosol compared to the sea level site and especially to the 2250 m site. These data are difficult to reconcile.

Table 1 indicates that the rain collected at 2250 m was very sparse, consisting of only 4 samples, totalling a rainfall of only 0.55 mm. This by itself might make these data suspect, since a significant amount of evaporation of the water falling on the dry rain collectors may occur before the surface becomes sufficiently wet. Yet the concentrations of the aerosols do not appear to be high, except for the beta activity of the first aliquot, which gives a $Sr^{90}/\beta$ ratio of only 0.01, the lowest value observed in Table 1. This high beta concentration cannot be attributed to evaporation, because the sodium and $Sr^{90}$ in the same aliquot would have become equally concentrated. Further, the $Sr^{90}/Na^+$ ratio looks reasonable, so that the very low $Sr^{90}/\beta$ ratio does not appear to be due to a low $Sr^{90}$ determination. The possibility exists that the aliquot became contaminated in the laboratory with non-$Sr^{90}$ radioactivity, but no evidence for this exists. Several other samples in Table 1 exhibited gross beta concentrations in excess of 100 pC/l. Furthermore, eliminating this sample from the average reduces the beta concentration to a value close to that at 1500 m, but does not eliminate the generally smaller $Sr^{90}/\beta$ ratios observed in the other three aliquots. Thus, the explanation for these data may need to come from physical or chemical processes occurring in the trade winds. Aerosols containing the gross beta activity are generally of unknown character. The activity itself consists to a great degree of refractory elements, such as the rare earths and the transition metals, and their compounds, which are generally insoluble materials. $Sr^{90}$, on the other hand, an alkaline-earth element, is generally soluble in almost all chemical compounds expected to exist in the atmosphere. The transport of $Sr^{90}$ aerosol in the trade wind air, thus, may be different than the transport of the general aerosol carying the gross beta radioactivity, due in part to the differences of particle-size distributions or solubility in condensed cloud water. The data from Table 1 do not allow a definite observation to be made, but it will be of interest in future studies to examine this possible fractionation more fully.

## Ratio of radioactivity concentrations in air to rain

During the July 15–16 rainshower period, simultaneous collections of aerosols and rain were made at several of the sites along the slope. The ratio of the radioactivity concentration in air to the concentration in rain has the dimensions of a mixing ratio. With the appropriate conversion factors, the ratio, $R^1$, in units of (pC/100 SCM)/pC/l can be expressed as a ratio, $R$, in units of g/kg where

$$R\ (\text{in g/kg}) = 7.75\ R'\left(\text{in } \frac{\text{liters}}{100\ \text{SCM}}\right).$$

Table 4. *Mixing-ratio data for the showers of July 15–16, 1965, in Hawaii*

| Altitude (m) | Radioactivity concentration in air (pC/100 SCM) | Average radioactivity concentration in air to 2440 m (pC/100 SCM) | Radioactivity concentration in rain (pC/l) | Calculated ratio, R (g/kg) | Observed mixing ratio, W (g/kg) | Average mixing ratio to 2440 m (g/kg) | Estimated fraction of water vapor condensed (%) |
|---|---|---|---|---|---|---|---|
| 2440 | 6.8 | | | | 4.9 | | |
| | | 5.8 | 61.7 | 0.73 | | 5.6 | 13 |
| 2250 | 4.9 | | | | 6.2 | | |
| | | 5.2 | 31.9 | 1.26 | | 6.6 | 19 |
| 1500 | 3.8 | | | | 8.8 | | |
| | | 5.6 | 56.3 | 0.77 | | 7.8 | 10 |
| 750 | 6.5 | | | | 11.2 | | |

For the relatively thin cloud layers associated with strong trade-wind conditions, this ratio may be related to the removal efficiencies of the radioactive aerosol and the fraction of water vapor precipitated. Hicks (1966) has shown from calculations of supersaturations of radioactive aerosols of stratospheric origin (i.e. nuclear weapons debris) that such aerosols are likely to be efficient nuclei in the production of cloud droplets. However, Elliott & Hovind (1964) have shown that orographic precipitation along the mountain range of southern California during Pacific cyclones may account for only about 25 per cent of the amount of water vapor condensed. For orographic rainfall in Hawaii, it might be expected that the radioactive aerosol is very effective as condensation nuclei, but that only some fraction of the condensed water actually deposits. If the rain water collected is a representative sample of the original cloud water, the ratio, $R$, may represent the amount of water vapor (in grams) that must have condensed to incorporate the amount of radioactive aerosol in 1 kg of air. The ratio would thus represent the liquid water content within the cloud at that time. Further condensation before rainout would lower the radioactivity concentration in the rain, and washout or evaporation below the cloud layer would increase the concentration. An examination of the data for the orographic rainfall of July 15–16 has been made with the assumption that the rainfall collected at the upper elevations was representative of the original cloud-water.

For below-cloud collection sites, the effective cloud water layer may be considered to lie between the lifting condensation level (LCL) and the base of the trade wind inversion. For collection sites above the LCL, the layer may be considered to be between the site and the inversion base. For the July 15–16 shower period, the LCL was at about 400 m and the inversion base was at 2440 m. The data in Figure 7 show that the gross beta concentration between these levels was reasonably constant, the average value was estimated to be about 5.5 pC/100 SCM.

For the calculation, it was assumed that the average value of the radioactivity concentrations in the air between each site and the base of the inversion at 2440 m represented the average concentration over the precipitation layer and that the filtering efficiency and condensation nuclei utilization was close to 100 per cent. The rainfall at each site represents the precipitation from the cloud tops down to the site. The mixing ratio at each site was calculated from the seven radiosondes taken over the shower period, and the average mixing ratio between each site and the inversion base was calculated from these values. The data for these calculations are given in Table 4.

The ratio, $R$, shows values of 0.73, 1.26, and 0.77 g/kg for the respective layers from 750, 1500, and 2250 m up to 2440 m. These values may be compared with the data of Squires & Warner (1957) who showed that the liquid water content of the orographic clouds in Hawaii was generally less than 1 g/kg unless there was rain in the cloud, and that the vertical profile through the cloud showed a peak at mid-cloud altitudes. Jiusto (1967)[1] has also shown

[1] See Jiusto manuscript, this issue of *Tellus*.

liquid water content of about 1 g/kg during the 1965 study period.

If the calculated ratio, $R$, is indicative of the liquid water content of the cloud layer, then the estimated fraction of water condensed ranged from 10 to 19 per cent, the peak occurring at the mid-layer site. These values imply that almost twice as much water vapor condensed at the mid altitudes of the moist layer than at either near the top or the bottom. This implication is also seen in the data in Table 4 which shows that the average radioactive aerosol concentration in the air was essentially uniform in the whole cloud layer, and since the average mixing ratio did not decrease too rapidly with height, the decrease in radioactivity concentration in the rainfall at the middle of the cloud layer reflects the increased fraction of water vapor condensed. The data in Table 4 also imply, if the estimate of Elliott & Hovind (1964) that about 25 per cent deposition of the orographic condensation in California applies to the orographic condensation in Hawaii, that about 2 to 4 per cent of the precipitable water in each layer was deposited as rainfall.

Further measurements of aerosol concentrations in air and rain at varying elevations are suggested to investigate further the potential of using the ratio of concentrations in thin-cloud systems to study the rainout process. Such additional measurements made under more steady precipitation periods may lead to indications of the droplet growth processes and drop size distributions of the rain with height in the cloud layer. An independent method of determining the fraction of water vapor deposited as rain, e.g. the adiabatic method of Fulks (1935), may enable estimates to be made of the rainout efficiencies, that is the fraction of aerosol removed as condensation nuclei. It is anticipated that such examinations will be made in future investigations of radioactive and stable aerosol deposition by warm rain along the slopes of Mauna Loa.

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## ПЕРЕНОС РАДИОАКТИВНОГО АЭРОЗОЛЯ ЧЕРЕЗ ИНВЕРСИЮ В ПАССАТАХ НА ГАВАЙЯХ

Результаты измерений концентрации радиоактивности в аэрозоле и в пробах осадках, собранных на склонах вулкана Мауна Лоа на острове Гавайи рассматривались в связи с вертикальными профилями температуры и влажности в атмосфере при наличии инверсии в пассатах. Вертикальный градиент концентрации радиоактивного аэрозоля связывается с профилем влажности и с характером инверсии. Значения концентрации увеличиваются с высотой при прохождении через инверсию в пассатах в противоположность аэрозолю земного происхождения, концентрация которого падает с высотой. Концентрация в дождях в общем уменьшается с высотой, за исключением аномального результата на 2250 м как раз под инверсией. На этой высоте полная концентрация радиоактивности увеличивается, в то время как концентрация $S^{90}$ уменьшается. Отношение полной $\beta$-концентрации в воздухе и дожде дает параметр с размерностью отношения смеси, который может дать потенциальную возможность для определения эффективности вымывания.