Radon in the Upper Atmosphere

Radon measurements near the equatorial and polar tropopause suggest the nature of atmospheric transport.

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Radon is a decay product of uranium-238 in the earth's surface. As the first gas in the uranium decay chain, it can breathe out of the soil and rocks. Further, its 3.8-day half-life permits it to be carried upward without undue loss while participating in normal atmospheric transport processes. It has a series of short-lived daughters (with half-lives less than 30 minutes) before decaying to long-lived (22-year) lead-210.

There have been numerous theoretical studies computing the vertical distribution of radon (or its daughters) from an equilibrium theory in which the divergence of the flux due to vertical mixing balances the loss due to its radioactive decay (*1*). An early and simple derivation of Schmidt's (*2*), predicts a simple exponential decrease of concentration with altitude by assuming a constant coefficient of vertical mixing.

A few measurements in the troposphere have confirmed a decreasing concentration with altitude (3–5) but with the same kinds of irregularities characteristic of humidity mixing ratio. The troposphere, on the average, is well mixed and the observed decrease in radon concentration—which at about 30,000 feet is about one-tenth of that measured at ground level—is consistent with well-established mixing intensities in this layer. Special conditions within a few tens of feet of the ground on nights with little wind and strong nocturnal thermal stability have been frequently noted by Moses *et al.* (*6*) but do not affect the broader picture to be described in this article.

The stratosphere, on the other hand, is thought to be characterized by slow vertical exchange such that one might expect little or no radon to be found in this upper layer of the atmosphere; it should have decayed before being transported deeply into the stratosphere. It was therefore felt that measurements of radon below and above the tropopause might prove to be a means of identifying the top of the well-mixed troposphere.

Two attempts were made to measure the short-lived radon daughters (which are a direct measure of the radon itself) by filtration aboard a B-57 aircraft above and below the tropopause. The first took place over Alaska in midwinter. It revealed a radon daughter concentration 5000 feet above the 33,000-foot tropopause at least 20 times smaller than that 5000 feet below the tropopause on the same day. Due to possible contamination of the filter paper during descent of the stratospheric flight through the troposphere it was possible that the decrease might have been far in excess of the factor of 20. The second attempt took place over Albuquerque, New Mexico, in the late winter. Again the radon daughter concentration just above the tropopause was roughly 20-fold smaller than below the tropopause and again the difference might have been larger had the sampling been conducted without contamination of the stratospheric sample.

Because of the difficulty of sampling for the radon daughters in the stratosphere, advantage was taken of the United States' world-wide sampling operations in May–June 1961 to measure the radon gas itself. Two of the geographically extreme stations were selected: one at 70°N (roughly, over Point Barrow, Alaska) and a second at 12° to 20°N (south of Hawaii). Steel bottles containing about 70 cubic feet of air at standard temperature and pressure (STP) were shipped by air freight to Argonne National Laboratory, where the radon content was determined.

The radon concentration was obtained by collecting the radon from 500- to 1000-liter samples of air on activated cocoanut charcoal at Dry Ice temperatures with subsequent transfer to an alpha scintillation radon counter (*7*). This system has a sensitivity of 5.45 count/min per 1×10^{-12} curie (picocurie) and a background counting rate of about 0.1 count/min.

Complete removal of carbon dioxide and water vapor is required for quantitative retention of radon on the charcoal (*8*). This was accomplished by passing the air through two spray-type gas washing towers in series, each filled with 380 grams of potassium hydroxide in 1.5 liters of distilled water, and then through a water-cooled condenser and six dip-type water freeze-out traps immersed in Dry Ice and carbon tetrachloride-chloroform slurry (1 : 1) (*9*). Warming of the air between each water trap was required to prevent "snow" carry-over.

Duplicate 500-liter samples were analyzed from 3 to 6 days after collection. The volume of each sample was recorded on a wet test meter (*10*) and corrections to standard conditions (STP) were made. The volume of each sample was obtained to 0.3 percent or better. Additional, 600- to 1000-liter, samples were taken 4 to 8 days later, and except for samples with less than 10^{-16} c/liter, agreement of replicate samples was within 10 percent of reported statistical error. All samples were counted for 900 minutes, with a repeat 400-minute counting interval for all except a few of them. Counting rates varied from a few tenths to 10 count/min. The statistical counting error, 0.9 count/min, was calculated on 0.35 of the net (radon + daughter) counts to compensate in part for the effect of the very short half-life of the radon daughters on the counting statistics (*11*).

Corrections were made for decay of radon to mid-collection, radon contamination from 2.3 pc radium-226 in the potassium hydroxide, the 0.1 pc radium-226 in the charcoal, and the 0.002 pc radon introduced through the rubber of the radon transfer system. No correction was made for contamination by radium within the sample bottle.

One such sample bottle, aged for 42

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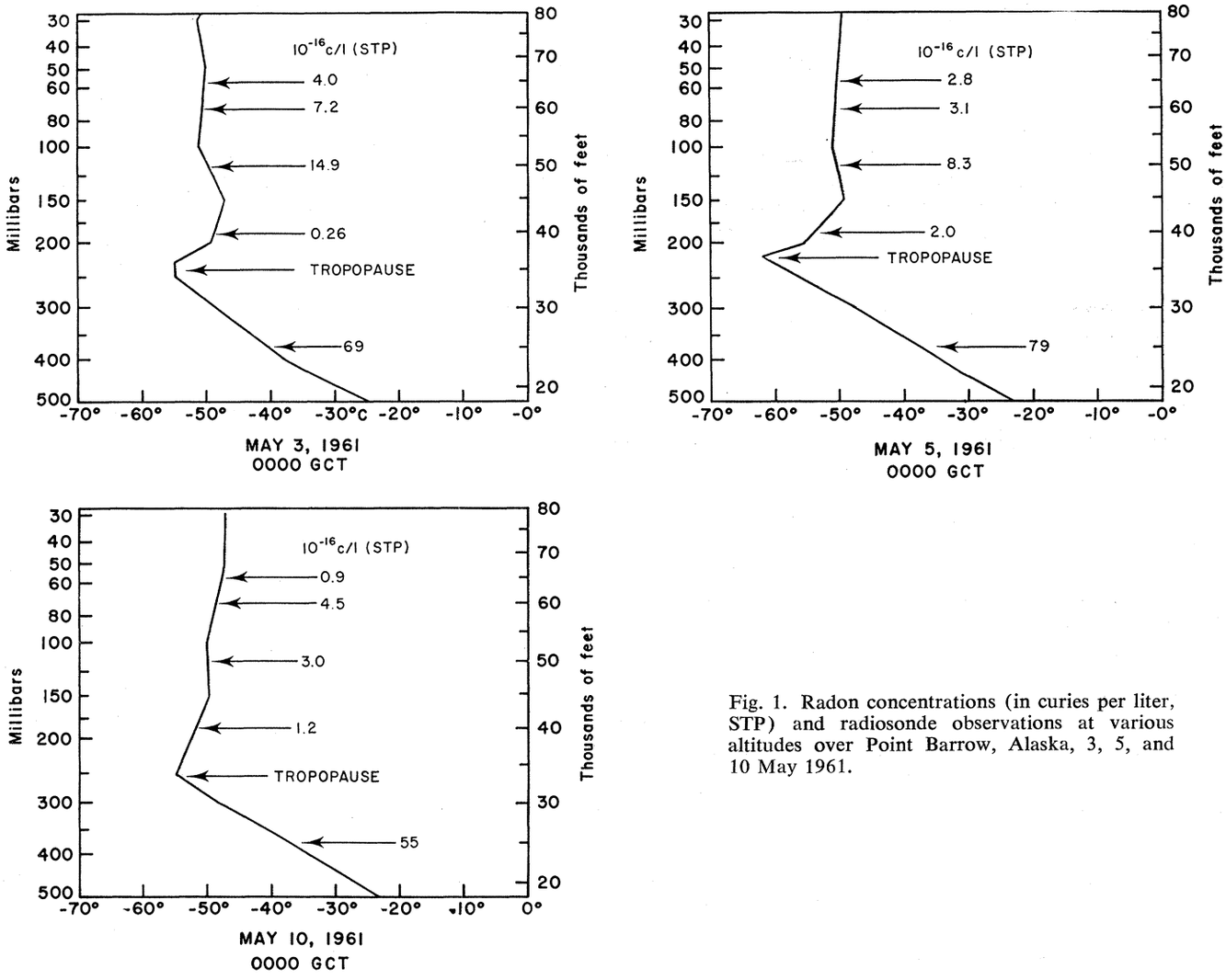

Fig. 1. Radon concentrations (in curies per liter, STP) and radiosonde observations at various altitudes over Point Barrow, Alaska, 3, 5, and 10 May 1961.

days at 2000 lb/in.^{2} (gage), was found to have a radon concentration of 0.4×10^{-16} c/liter, which would correspond to 0.2×10^{-16} c/liter at time of analysis. This is very much less than the 5×10^{-15} to 5×10^{-16} c/liter found previously for larger compressed air cylinders (*8*). Since variations in the radium content of the various sample bottles and in the pressure and the effectiveness of purging at time of sample collection have not been evaluated, the accuracy of values of less than 1×10^{-16} c/liter is limited.

The resulting concentration in 10^{-16} c/liter (STP) is summarized in Table 1. Concentrations from less than 1×10^{-16} to 90×10^{-16} c/liter were found. These can be compared to measurements of normal ground-level air at Argonne National Laboratory, measured by a similar technique in which an annual average of about 3×10^{-13} c/liter was found (*12*). Under certain weather conditions, the radon concentration was found to vary from 0.5×10^{-13} to 17×10^{-12} c/liter. Over the oceans, far removed from land, the concentration may decrease to about 1×10^{-15} c/liter (*13*). The radon concentration is also likely to be smaller over frozen ground (*14*). For these reasons, it is difficult to assess a mean hemispheric ground-level concentration for comparison with samples collected at altitude but very likely will be within 1×10^{-13} to 1×10^{-14} c/liter.

Alaska Series

The temperature profiles and the radon concentrations are shown in Fig. 1 for the three Point Barrow, Alaska, observations. The three series have common features both with regard to the temperature structure and the radon concentration. The 25,000-foot samples lie in the troposphere and uniformly have the highest values. The 40,000-foot samples lie just above the tropopause in a layer characterized by temperature increasing with height. The radon content of these samples are the lowest observed at any level on two of the three days. On all three days, the 50,000-foot radon content increases over the 40,000-foot value, the amount of the increase appearing to correspond rather closely with the rate of temperature decrease with altitude in the layer in which the sample was collected. Above 50,000 feet the radon concentrations decrease with altitude on all but the last run.

It is evident that the layer of air in which the 50,000-foot sample was collected has had a more recent tropospheric history than the 40,000-foot layer. Thus separate layers with more or less recent tropospheric history may readily overlay one another within the

Table 1. Radon concentrations of high-altitude air. The statistical counting error or average deviation of the mean of replicate samples, whichever was greater, was 0.9. The Alaskan samples were collected at approximately 1900 to 2300 GMT, and the Hawaiian samples at 0000 to 0200 GMT, on the indicated days.

Altitude (feet)	Counts ($\times 10^{-16}$) per liter (STP)				Average
	Alaska (between 67.5° to 71.5°N, 143.5° to 146°W)				
	2 May	*4 May*	*9 May*		
25,000	69. ± 3	79. ± 6	55. ± 1		68
40,000	0.26 ± 0.2*	2.0 ± 0.4	1.2 ± 0.4		1.2
50,000	14.9 ± 0.2	8.3 ± 0.3	3.0 ± 0.4		8.7
60,000	7.2 ± 0.2	3.1 ± 0.3	4.5 ± 0.4		4.9
65,000	4.0 ± 0.2	2.8 ± 0.3	0.9 ± 0.1		2.6
	South of Hawaii (12° to 20°N, 159°W)				
	6 June†	*8 June*	*15 June*	*20 June*	
50,000	12.2 ± 0.4	21.7 ± 1.2	19.3 ± 1.0	18.7 ± 0.2	18.0
60,000	6.1 ± 0.4		3.0 ± 0.4	7.1 ± 1.1	5.4
65,000	2.7 ± 0.4		0.5 ± 0.2*	4. ± 3.	2.4

*These values cannot be differentiated from zero values; see text. †This collection was made at 16° to 20°N and 159°W.

stratosphere. This conclusion is also evident from the detailed chemical ozone measurements (*15*). Radon, however, having a 3.8-day half-life, places a time scale on the process, although some qualifying assumptions must be made in deriving quantitative time intervals. Thus, if one compares the 8.7×10^{-16} c/liter at 50,000 feet with the 1.2×10^{-16} c/liter at 40,000 feet, one can argue that the 40,000-foot sample is 11 days older. Or, if the 8.7×10^{-16} c/liter was derived from air which had, say, 25×10^{-16} c/liter farther south where the 50,000-foot level is in the troposphere, then there has been about a 6-day quasi-horizontal transit from this southern latitude to Point Barrow. While there may be an uncertainty in assuming 25×10^{-16} c/liter the true value is likely to be larger than the 18×10^{-16} c/liter found at 12° to 20°N and smaller than the 68×10^{-16} c/liter found at 25,000 feet over Alaska. Thus, the transit time might be as long as about 2 weeks or as short as 2.5 days rather than 6 days.

The horizontal airflow pattern over the Point Barrow area was dominated at least up to 40,000 feet by an anticyclone located to the west or northwest of the sampling point. This weather system weakened considerably toward the end of the collection period. Airflow at the 25,000-foot level arrived from the west to northwest quadrant. At 40,000 feet the air came from the northwest on the 2nd and 4th of May, then from the south on the 9th. At 50,000 feet, however, the air appeared to originate from the north on all three days, despite its more tropospheric nature. The winds at 60,000 and 65,000 feet blew from an easterly quadrant on all occasions.

One may interpret the changes of radon concentration either in terms of transport by organized circulations or by turbulent mixing. Present meteorological thinking by Machta does not suggest that there is organized rising motion in the polar regions in the spring season of the year. Such rising motion would be required, in the absence of vertical mixing, between all layers except the 40,000- and 50,000-foot layers. If present, it would amount to about 1 cm sec^{-1}, assuming the concentrations in the 50,000- to 65,000-foot layer to be representative. But, if the vertical flux between 50,000 and 65,000 feet is due to vertical diffusion and a steady state is assumed, then there is an average diffusion coefficient of 1×10^5 to 6×10^5 cm^2 sec^{-1} in this layer. This value (as well as the computed rising current) is rather large for the stratosphere and probably reflects the lack of reality of the assumptions. On the other hand, many meteorologists have suggested that vertical mixing is enhanced over its normal stratospheric value of 1×10^3 to 1×10^4 cm^2 sec^{-1} during the polar spring season.

The correspondence between the high radon concentration, a measure of "tropospheric" air, and the decrease of temperature with altitude in the "stratosphere" may require a reevaluation of the ease with which exchange of air between the two layers takes place (*16*). Further, air at 40,000 feet may be more "stratospheric" than air at 65,000 feet, despite the lower altitude and apparent lesser remoteness from the troposphere.

Hawaiian Series

Comparative temperature and radon profiles for the equatorial observations are not given because of the long interval over which the collections were made and the absence of weather data at all but the northern end point. Extrapolating from nearby radiosonde observations, the 50,000-foot sample collected on 8 June, the highest value, appears to have been obtained entirely within the troposphere, and the 20 June sample, entirely in the stratosphere, but the three remaining 50,000-foot samples are mixtures of air from both layers. All of the 60,000- and 65,000-foot samples were collected above the tropopause.

The surprising feature of this picture is the large amount of radon at 60,000 and 65,000 feet compared with that at 50,000 feet. If a rising current is invoked to account for this transport into the stratosphere, and the radon concentrations at sampling location are considered to be those at the points at which the upward motion began, one needs rising motions of the order of 0.5 cm sec^{-1}. It is likely that the upward currents originate over continental areas associated, in part, with convection. For this reason, the radon concentration just below the tropopause is likely to be considerably greater than that reported at 50,000 feet in the mid-Pacific. Thus, the magnitude of the rising current just computed, 0.5 cm sec^{-1}, is probably an overestimate by perhaps as much as an order of magnitude.

On the other hand, if the radon reaches 60,000 and 65,000 feet by turbulent mixing, and if, again, the mean concentrations in Table 1 represent the steady state, one computes a coefficient of vertical mixing of between 0.7×10^5 to 1.3×10^5 cm^2 sec^{-1}. This exceeds the expected value in the lower equatorial stratosphere by perhaps an order of magnitude. It may be in error, again, since the main source of radon is over equatorial continental areas, but in this case it can be either too small or too large. If about 1×10^5 cm^2 sec^{-1} is accepted as correct, the ozone and fission products (after a few months of the injection anywhere in the lower equatorial stratosphere), both of which are tracers of stratospheric air, should show virtually no vertical gradient between the tropopause and 65,000 feet. In fact, there is a deficiency of ozone over that expected from photochemical

equilibrium theory, witn concentrations not unlike those of the upper troposphere, and except during periods of equatorial test operations there is a very marked increase in fission product concentration from the tropopause to at least 65,000 feet. It is therefore suggested that the radon which is found above the equatorial tropopause has been carried there by a slow rising (on the average) current, through the tropopause to at least 65,000 feet. Further, this mode of transport dominates the flux that results from turbulent vertical exchange. The history of the radiotungsten introduced during the United States 1958 equatorial tests suggests, however, that the rising current does not reach above 65,000 feet (*17*).

The mean stratospheric radon concentrations at 60,000 and 65,000 feet at both the polar and equatorial locations are very similar. It is unlikely that intense horizontal mixing in this part of the stratosphere could have produced the similarity, because even the most extreme coefficients of horizontal mixing would result in significant differences if injection into the stratosphere occurred at either location, or if injection occurred in the temperate zone.

The initial collections of radon gas in two extreme geographical locations have proven to be far more interesting than expected. In the case of the Alaskan results, the reversals in concentration with height are associated with features of the thermal structure and indicate that slices of recent tropospheric air may readily interleaf the air normally considered stratospheric. Further, semiquantitative estimates of the time scale can be given, since radon has a comparatively short half-life of 3.8 days and its presence in observable amounts would probably not allow transit times to the lower stratosphere from the troposphere longer than about a month. More radon was also found in the lower equatorial stratosphere than expected. These observations, combined with the vertical profiles of ozone and fission products whose origin is the stratosphere, suggest that a rising motion, rather than turbulent mixing, is the more likely mode of transfer. The magnitude of such rising currents of about 10^{-2} to 10^{-1} cm sec^{-1} is about the magnitude predicted as upper limits by Murgatroyd (*18*) and expected by Machta (*19*).

It is expected that additional profiles can be obtained in future aircraft operations at these locations and in the southern United States. The usefulness of radon to measure the coefficient of vertical turbulent mixing has been demonstrated for the troposphere (*4*) and in this article its potential value in the lower stratosphere is equally evident (*20*).

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News and Comment

Playing with Numbers: The Public Wants a Balanced Budget, So the Public Gets a Balanced Budget

With the submission this week of the Economic Report, the President has completed his first full presentation of the series of three messages that begin each congressional session. The State of the Union and Budget messages were reviewed here last week. In the Economic Message, and its accompanying Report of the Council of Economic Advisers, the Administration offers its view of the state of the economy, its estimates of the economic significance of the President's program, and a rationale of the basis for the budget.

As reported last year in a review of Kennedy's and Eisenhower's approaches to economics (10 Feb. 1961) the two administrations are in basic agreement on any number of general principles (that greater investment in science and education is necessary for economic growth, that a major goal of economic policy should be to limit inflation, that deficits are useful in combatting recessions, and so forth), but are in substantial disagreement over how these general principles should be applied in practice. Kennedy obviously believes in spending more money (as reported last week, Kennedy's new budget increases spending by over $3 billion at the same point in the business cycle that Eisenhower's fiscal 1960 budget reduced spending by over $3 billion); he does not emphasize, as Eisenhower did strongly, a reduction in taxes as even a long-range goal; and he does not share Eisenhower's concern over the question of whether the budget is balanced.

Nothing contrasts so sharply between the Eisenhower Budget and Economic