METEOROLOGICAL EFFECTS ON ATMOSPHERIC CONCENTRATIONS OF RADON (Rn^{222}), RaB (Pb^{214}), AND RaC (Bi^{214}) NEAR THE GROUND[1]

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ABSTRACT

Atmospheric concentrations of radon were calculated from filtered radon daughter β activity, obtained from 1- and 15-m. elevations at a rural site, and from 1- and 91-m. elevations at a semi-rural site near Washington, D.C. Measurements were obtained from the successive filtration of air over 20-min. periods for durations of several hours, during various meteorological conditions. The extent of secular equilibrium between radon and its daughter products in the atmosphere near the ground ($\leq$ 15 m.) was determined from a calculation of the RaC/RaB atom ratio (ρ) in the air, derived from the measured rate of decay of the filtered β activity. Estimates of an effective residence time (τ) in the atmosphere of the radon conglomerate, based on either a continuous or single emission of radon into the air, are presented. The relationship between ρ and τ for various weather conditions is discussed, as well as possible effects of (a) wind, (b) thermal stability, (c) atmospheric scavenging, and (d) precipitation on measured levels of natural radioactivity. Radon concentrations measured during an "air pollution potential" episode and during fumigation conditions are presented. In general, the response of measured β activity in the air to changes in the turbulent state of the atmosphere near the ground gives evidence of the feasibility of using naturally occurring radon and radon daughter products as tracers for determining atmospheric diffusivity.

1. INTRODUCTION

From measurements at selected times of the rate of decay of RaB+RaC β activity collected on a filter, a determination of the extent of secular equilibrium in the air between radon and its daughter products can be obtained; this is expressed by the derived atom concentration ratio (ρ) in the air, defined as ρ=RaC/RaB (Lockhart et al. [12]). To calibrate this analytical method of determining radon concentration from measured radon decay product activity, measurements of RaB+RaC β activity were made at elevations of 1 and 91 m. during various weather conditions (Hosler and Lockhart [3]). From these experiments information was obtained of meteorological effects on the temporal and vertical distribution of radon and its daughter products in the air near the ground. These data, supplemented by other radon concentration measurements including those at a height of 15 m., are the subject of this discussion. The radon decay series and a discussion of the meaning of low and high ρ-values are presented in the Appendix.

Most of the radon measurements documented herein were obtained at Sterling, Va., about 25 mi. west of Washington, D.C. This site is on flat terrain where the grounds are mowed in summer. There are woods to the south and east about 400 m. from the sampling apparatus; relatively unobstructed fetches to a distance of 1000 m. lie to the west and north. Alternate woods, cultivated fields, and pasture land comprise the surrounding land Dulles International Airport adjoins the site to the southeast; the north-south runway is about 1200 m. southeast of the site. Only one filtration unit was employed at Sterling, which required alternate air sampling at 1- and 15-m. heights to obtain vertical profiles of radon concentration. This was accomplished by drawing air through a 5.08-cm. (inside diameter) polyethylene tube to a filter housed inside the laboratory at the site. The tube was attached to a 15-m. tower and a pulley arrangement permitted alternate air sampling at two levels. The tower was instrumented with thermocouples at 1 and 15 m. to provide temperature gradient data; a Beckman & Whitley anemometer measured wind at a height of 10 m.

Two air sampling units installed at a 91-m. tower facility at Tysons Corner, Va. permitted simultaneous air filtration at 1- and 91-m. elevations. This site is about 15 mi. west of Washington, D.C., at the edge of the suburbs. Alternate open fields, woods, and housing developments on gently rolling terrain characterize the area. This tower is meteorologically instrumented with aspirated thermocouples and dew cells at 2-, 30-, 60-, and 91-m. elevations; Aerovane anemometers are mounted at 30, 60, and 91 m. All meteorological data are recorded on charts.

Radon concentrations derived from filtered β activities are corrected for secular equilibrium departure on the basis of derived ρ-values. An added correction factor of 1.25 was applied to 1- and 15-m. concentrations; this correction was based on previous calibrations (Hosler and Lockhart [3]). Most of the experiments were conducted

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under dry soil, non-precipitation conditions; filtrations made during other than "normal" weather conditions are specified. All runs were made for a period of 20 min. which provided filtration of about 15 m.^{3} of air per run. The counting technique, sampling apparatus and calibration procedures employed for these experiments have been described previously (Lockhart et al. [12]; Hosler and Lockhart [3]).

2. METEOROLOGICAL EFFECTS

At any point in the atmosphere the radon concentration is a function of (a) exhalation rate from the soil, (b) rate of vertical mixing, (c) rate of horizontal transport, (d) radioactive decay, (e) elevation, and (f) depletion due to fallout, washout, absorption, and other scavenging effects. The meteorological influences at near-ground levels on the first three factors have been discussed by Moses et al. [15, 16] in the studies of the hourly variation of radon concentration at different levels below 40 m. at Argonne National Laboratory, Ill. In general these experiments supplement those conducted at Argonne.

TRANSPORT

Transport from areas of higher or lower radon concentration cannot be discounted, particularly during inversion, light-wind conditions; such stable periods may give rise to localized accumulations of radon leading to high concentrations in sheltered valleys, basins, and forested areas. While part of the observed short-period variations in concentration probably can be attributed to transport, any heterogeneity of horizontal distribution of radon should be masked by the larger variations in concentration due to vertical mixing rates and, to a lesser extent, by variations in exhalation rates (Junge [6]). However, this would not apply to areas adjacent to large bodies of water. Measurements taken at the two sites west of Washington, D.C., for several years have given no indication of preferred directions of prevailing high or low concentrations of natural radioactivity.

The relationship of wind speed to radon content in the air is not straightforward. Calculated ρ-values for 1- and 15-m. heights at the Sterling site are shown in figure 1 for various wind speeds, averaged to the nearest half knot (0.258 m./sec.) over each 20-min. filtration period. Inversion conditions defined by a temperature difference $(T_{15m.}-T_{1m.})>0.0°C.$ over the height interval, are designated in figure 1; the lower ρ-values associated with atmospheric stability may reflect the effects of limited vertical motion, causing an accumulation of fresh radon deficient in daughter products near the ground. This is discussed in more detail later.

An interesting feature of the data shown in figure 1 is the suggestion for lower ρ-values at wind speeds greater than about 16 kt.; this appears to be more evident at a height of 15 m. than at 1 m., but this is attributed to the selection of reported data. This may reflect an increase in the exhalation rate of radon that has accumulated in the top layers of the soil as a result of micro-oscillations

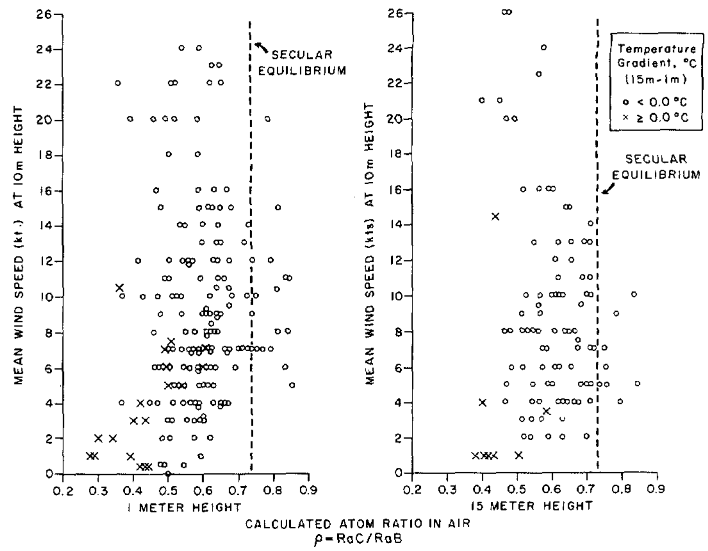

FIGURE 1.—Relationship of calculated RaC/RaB atom ratio in air to wind speed, at 1- and 15-m. heights at Sterling, Va.

in barometric pressure associated with high wind speeds near the ground. Kraner et al. [11] have reported on the mechanisms by which wind and atmospheric thermal stability deplete the ground surface layers of radon; they suggest that the micro-oscillations in pressure give rise to a "turbulent pumping" that dilutes and partially exchanges shallow layer soil gas with radon-free air from above the ground. Kovach [9, 10] and Schroeder et al. [20] also have reported on the reduction of the radon content in the topmost layers of soil during windy conditions.

Another explanation for the apparent departure from secular equilibrium between radon and its daughter products, in air near the ground during windy conditions, may be attributed to greater mechanical turbulence and thus increased vertical mixing near the surface. Such conditions could lead to a proportionally larger fraction of fresh radon at 15 m. than would exist under conditions of less vertical mixing at that height. This has been suggested by Jacobi and Andre [5] who report that with increasing turbulence the deviation from secular equilibrium between radon and its daughter products will be smaller near the ground, but the equilibrium departure will extend to greater heights.

Finally, it was interesting to note increased dust loads on the filters during windy days. Consideration was given to the fact that an excessively heavy dust content on the filter might be effective in discriminating against the weaker β emissions from the RaB. However, Lockhart and Patterson (to be published as a U.S. Naval Research Laboratory Report) have shown that a dust load of 1 mg./cm.^{2} has negligible effects on a computed ρ-value; this suggests that heavy dust collected on the filters does not jeopardize the counting technique.

Some of the derived ρ-values exceed the secular equilibrium value of 0.735; this is shown in figure 1 and in other data shown later. No explanation is offered. Natural atmospheric scavenging processes and/or discrimination of the radon daughter products by either the sampling or counting techniques may be responsible. Inspection of the data indicates that $\rho > 0.735$ cannot be attributed only to statistical counting error. The process of attachment of the decay atoms to particulate matter in the atmosphere [6], cannot be discounted as a mechanism for affecting filtration efficiency and the measuring technique.

REMOVAL PROCESSES

Another factor that may affect the vertical distribution of radon in the air has been suggested by Israel [4], who points out that adsorption of radon onto particles may be important. The possibility of depletion of radon daughter products in the air through fallout or scavenging effects has been mentioned by Hosler and Lockhart [3]; this could affect derived ρ-values. This can be shown from equation (1):

$$\rho = \frac{\text{RaC atoms in air}}{\text{RaB atoms in air}} = \frac{\lambda_b}{\lambda_c} \quad (1)$$

where λ_b and λ_c are the respective decay constants for RaB and RaC. If we assume preferential depletion of RaC, which may be more likely to occur in the atmosphere because of its longer residence in the air than RaA or RaB, and if for simplicity we let $\Lambda_b = 0$ and $\Lambda_c > 0$ where Λ is the removal coefficient for the respective isotopes, then equation (1) becomes

$$\rho = \frac{\lambda_b}{\lambda_c + \Lambda_c} \quad (2)$$

which yields the RaC depletion factor, Λ_c, for a given value of ρ for equation (3).

$$\Lambda_c = \frac{\lambda_b}{\rho} - \lambda_c \quad (3)$$

This depletion factor can be inverted to give a residence time, τ_c, which defines the mean time that a RaC atom remains in the atmosphere, correcting for decay. Figure 2 shows computed τ_c and ρ-values in the air that would result from a depletion of only RaC in the atmosphere, under steady-state conditions. Whether removal processes are more effective in scavenging RaC than RaA or RaB from the air near the ground during windy conditions, by impaction on vegetation or fallout on dust raised by the wind, cannot be determined from these data.

An additional complicating factor in relating atmospheric radon daughter product activity to meteorological conditions is the apparent response of atmospheric radioactivity content to variations in the electric field. An example of this "electrode effect" has been studied by Wilkening [23], who documented the anomalous decrease in radon daughter-ion activity accompanying electric field reversals in thunderstorm areas in New Mexico. Also, the discriminate collection of ions by sampling systems exposed to electric fields and the effect of air

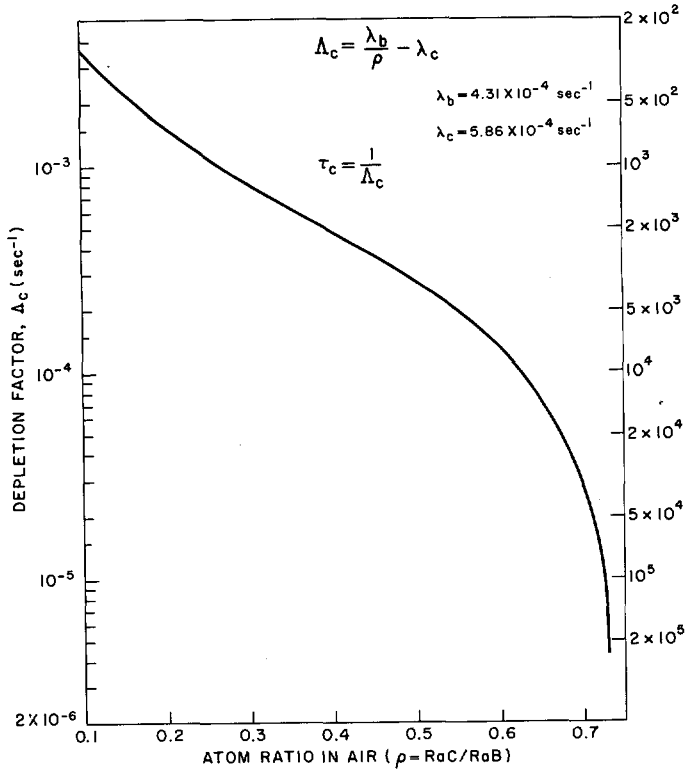

FIGURE 2.—Relationship between RaC/RaB atom ratio (ρ) and RaC depletion factor (Λ_c) and RaC residence time (τ_c) in air.

pollution on the conductivity and ion balance in the atmosphere have been pointed out by Cobb and Phillips [2] and Phillips et al. [19], respectively; this represents an additional potential source of error for filtration procedures to determine levels of atmospheric radioactivity.

Despite the numerous variables which enter into the relationship between radon and radon daughter product activity in the atmosphere, it has been demonstrated that during non-dusty conditions radon concentrations in air can be determined rather accurately ($\pm$ 20 percent) from measured radon daughter β activity (Hosler and Lockhart [3]). This suggests that potential errors in the derived radon concentration, resulting from the various complexities discussed heretofore, are small relative to the normal diurnal range of radon concentration observed near the ground. Measuring radon daughter product β activity over successive 20-min. intervals for periods of several hours provides data that are sufficiently reliable and descriptive of the radon concentration levels that exist for periods of several hours. Numerous examples are given later.

THERMAL STABILITY

With the establishment of surface-based radiation-type inversions around sunset, vertical motions are restricted. During the nocturnal stable period that follows, radon would be expected to accumulate near the ground from which it is exhaled; the accumulation of fresh radon deficient in daughter products would result initially in a departure from secular equilibrium, indicated by rela-

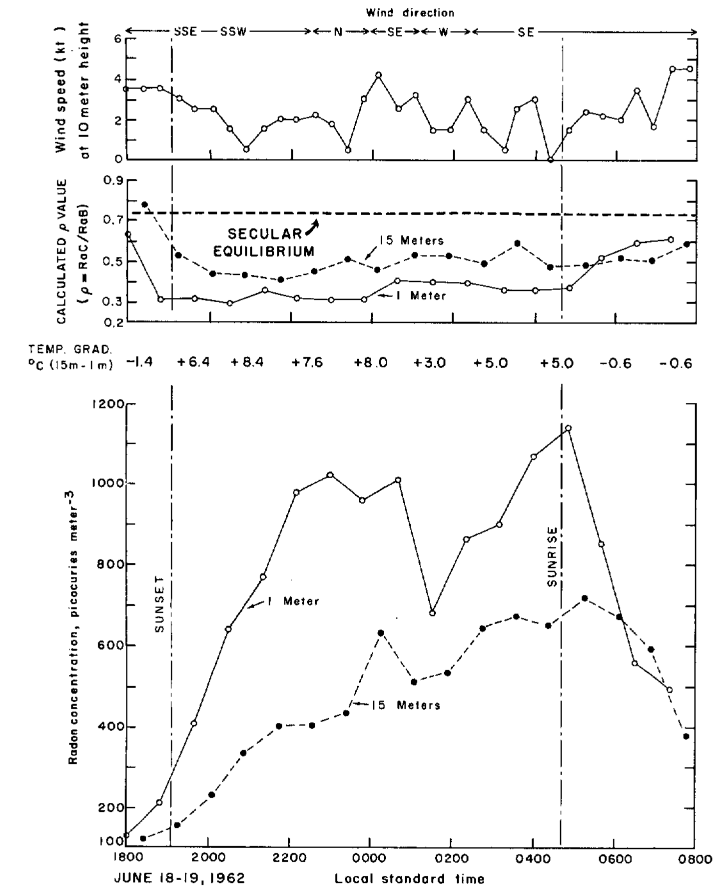

FIGURE 3.—Vertical distribution of radon and attendant meteorological data for nocturnal inversion case at Sterling, Va., June 18–19, 1962.

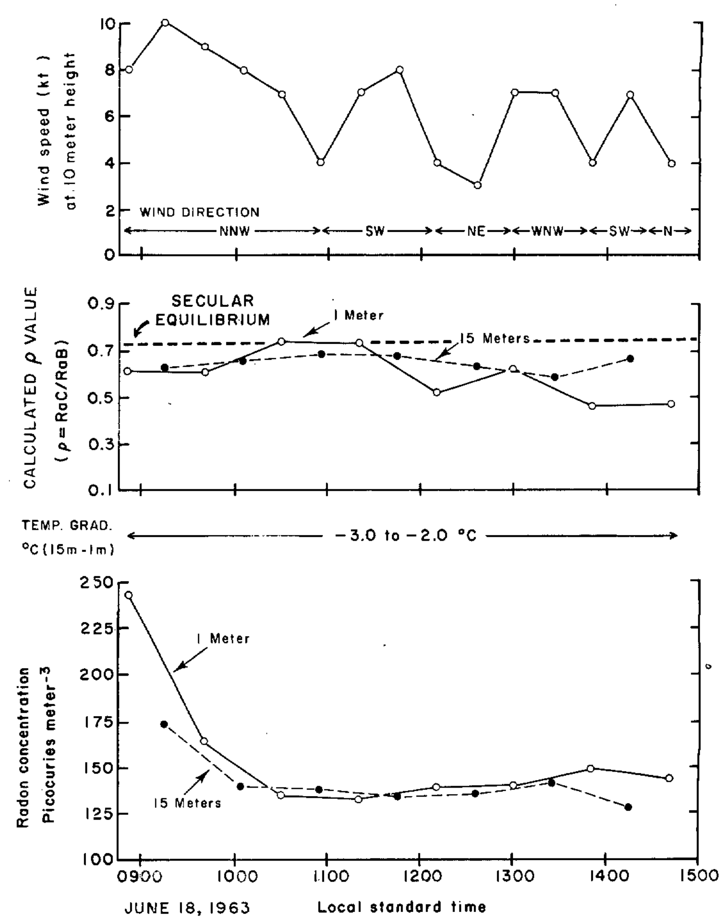

FIGURE 5.—Vertical distribution of radon and attendant meteorological data for daytime convective case at Sterling, Va., June 18, 1963.

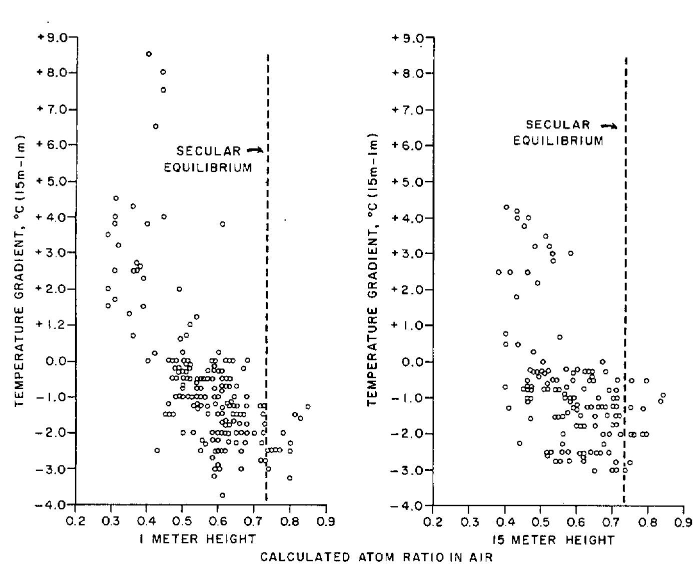

FIGURE 4.—Relationship of calculated RaC/RaB atom ratio in air to vertical temperature gradient, at 1- and 15-m. heights at Sterling, Va.

tively low values of ρ. Such stable regimes are numerous at the Sterling site and figure 3 graphically portrays such a case. The rapid accumulation of radon with the onset of the inversion is quite apparent; the lower ρ-values, particularly during the first few hours of inversion, apparently reflect the relatively large fraction of fresh radon making up the conglomerate. The lower ρ-values at the 1-m. elevation throughout the inversion period are compatible with the premise that the conglomerate nearer the radon source will possess a larger fraction of fresh radon deficient in daughter products.

From radioactivity measurements conducted at Sterling periodically in 1962 and 1963, calculated ρ-values for 1- and 15-m. heights are plotted in figure 4 for a wide range of stability conditions. At first glance it appears that a derived ρ-value might be correlated directly with vertical temperature gradient, at near-ground levels. However, any relationship between ρ and thermal stability must consider the time and duration of the stability regime, with respect to the observation time, since a ρ-value essentially represents the effective age or residence time

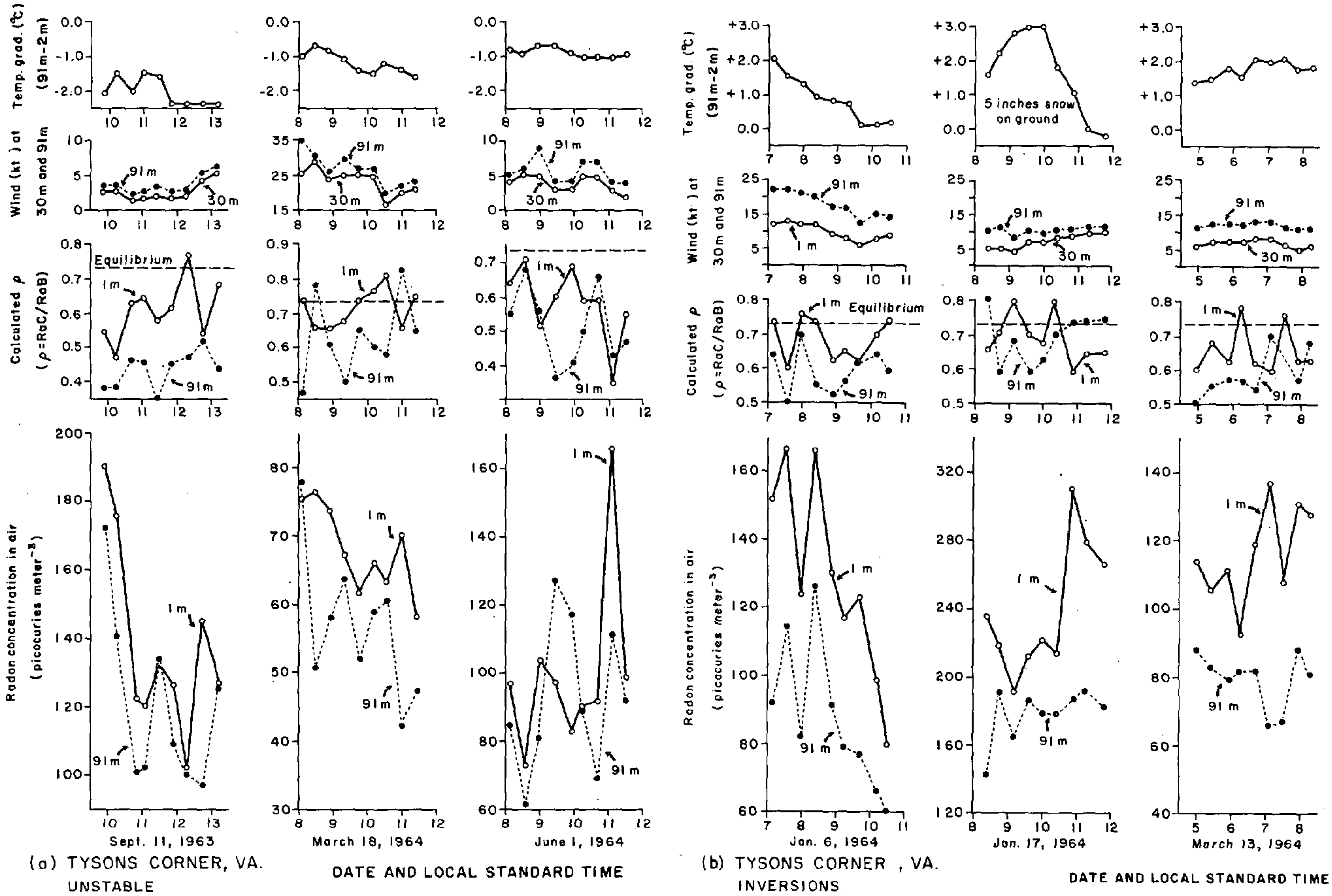

FIGURE 6.—Vertical distribution of radon and attendant meteorological data at Tysons Corner, Va. for (a) unstable and (b) inversion periods.

(discussed in section 3) of the radon conglomerate. Consequently, during steady-state inversion conditions for a constant exhalation rate, with increasing time the radon conglomerate will be composed of a proportionally larger amount of aging radon until equilibrium is attained; this is reflected by a gradual tendency for ρ to increase in value throughout the inversion period, as indicated in figure 3. Since most of the ρ-values plotted in figure 4 were obtained at the termination of the nocturnal inversion (immediately before sunrise), these values are probably biased to read 0.1 to 0.2 higher than values that would be obtained from observations taken immediately after sunset. Analysis of a series of experiments similar to that shown in figure 3 indicates that on the average at the Sterling site during nocturnal surface-based radiation inversions, ρ varies from about 0.3 at sunset to near 0.5 at sunrise at a height of 1 m.; from 0.4 at sunset to about 0.6 at sunrise at an elevation of 15 m.

During the convective daytime period there is considerable vertical motion near the earth's surface, and radon being exhaled from the soil is rapidly diffused into the atmosphere. An example of the vertical distribution of radon near the ground during a sunny daytime turbulent condition is shown in figure 5. The small vertical gradient of concentration and similar ρ-values, calculated for both 1- and 15-m. heights, reflect the homogeneous composition of the radon conglomerate near the ground under turbulent conditions.

Radon concentrations measured at an elevation of 91 m. are shown in figures 6a and 6b for unstable and inversion periods, respectively. These data reflect the fact that β-activity measurements conducted at the Tysons Corner tower site during all types of weather have shown a consistent departure from secular equilibrium ($\rho<0.735$) at an elevation of 91 m., based on computed ρ-values; this is incongruous to a state of equilibrium that would be expected to prevail at that elevation (Jacobi and Andre [5]), and inconsistent with the ρ-values obtained at 1 and 15 m. at the Sterling site. Any discriminate scavenging of RaC from the atmosphere would yield low values of ρ, which might be a manifestation of electric field tower effects on the radium ion balance in the air.

FUMIGATION

With the breakup of the surface-based inversion shortly after sunrise there follows an increase in vertical mixing near the ground. During this transition from stable to unstable conditions, radon-laden air near the ground moves upward while air with lower radon concentration is brought to the ground. For brief periods this can result in higher radon concentrations aloft than at the surface. Moses et al. [15] termed this phenomenon an "inverse fumigation condition". This phenomenon has been observed rather frequently at both sites, usually within an hour or two after sunrise. An example of this fumigation condition is indicated shortly after 0600 LST for the case shown in figure 3. For this transition period the lower ρ-value at 15 m. may be interpreted as indicating a larger fraction of fresh radon from below has appeared suddenly at the level; conversely, the higher ρ-value at 1 m. indicates a radon conglomerate of longer residence time, presumably from aloft.

ATMOSPHERIC STAGNATION

The existence and forecasting by the U.S. Weather Bureau of meteorological conditions favorable for the accumulation of air pollutants, over areas thousands of square miles for durations of several days, has been described by Niemeyer [17], Boettger [1], and Miller and Niemeyer [14]. In general, such stagnation episodes are associated with quasi-stationary warm anticyclones which usually affect the eastern half of the United States in summer and fall. The overall effect of this synoptic regime is to restrict the volume of air available for atmospheric dilution through (a) a persistent subsidence inversion, (b) strong nocturnal surface-based radiation inversions, and (c) low wind speeds. During stagnant weather conditions radioactivity measurements at both sites have yielded abnormally high radon concentrations (200–400 pc. m.$^{-3}$) during *midday* hours. Such concentrations are a factor of 2–4 higher than normally observed during non-stagnant conditions at midday in summer and fall at these sites.

The air pollution episode of November–December 1962 is a good example; this case has been described by Lynn et al. [13]. The initial stagnation potential forecast by the U.S. Weather Bureau, issued November 27, 1962, included an area from New England southwestward into West Virginia. Within a few days the stagnation area expanded to eventually affect 22 States from Maine to Arkansas. The Washington, D.C., metropolitan area was affected from November 29 through December 4; however, a buildup of radon activity was noted on the 28th, simultaneously with increasing SO_2 concentrations in downtown Washington, D.C. Air quality data for this period from the U.S. Public Health Service's Continuous Air Monitoring Program Station (CAMP) in downtown Washington, D.C., are shown in figure 7. Measurements of radon concentrations near the ground at midday

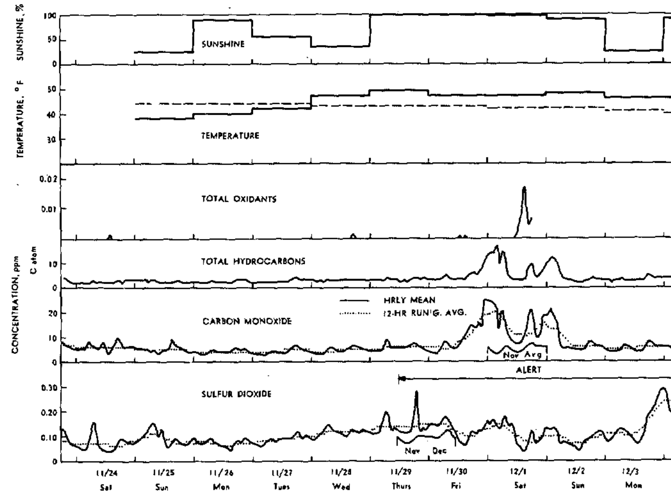

FIGURE 7.—Data from Public Health Service's air monitoring station at Washington, D.C. (after Lynn et al. [13]).

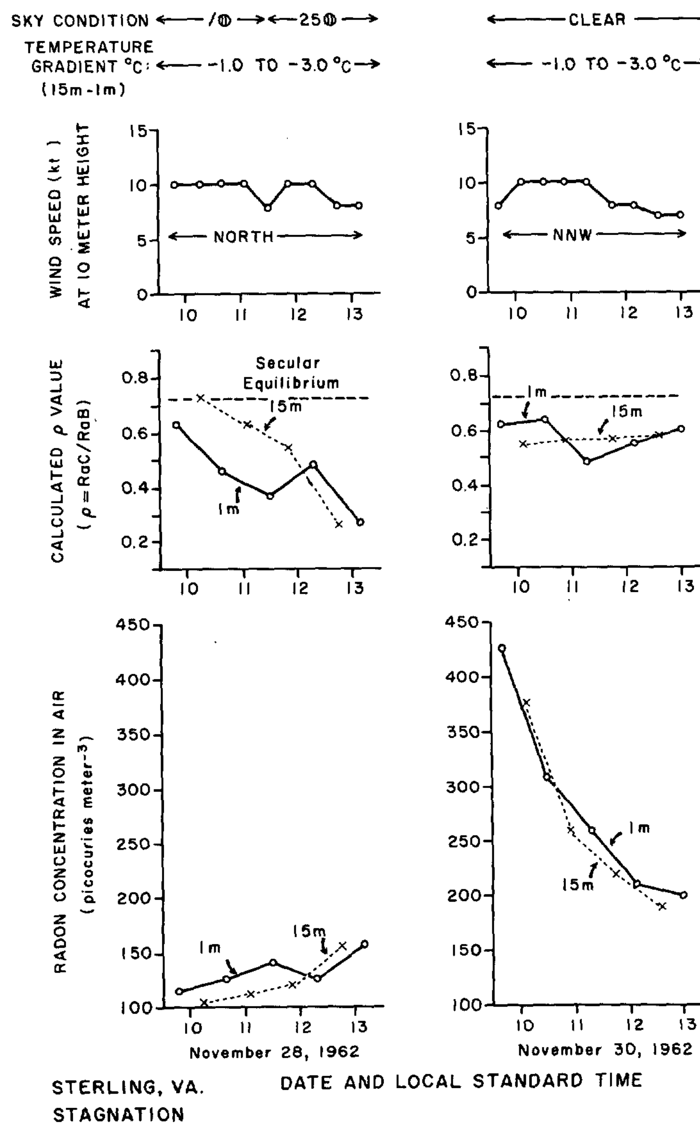

FIGURE 8.—Radon concentrations at Sterling, Va., during atmospheric stagnation days.

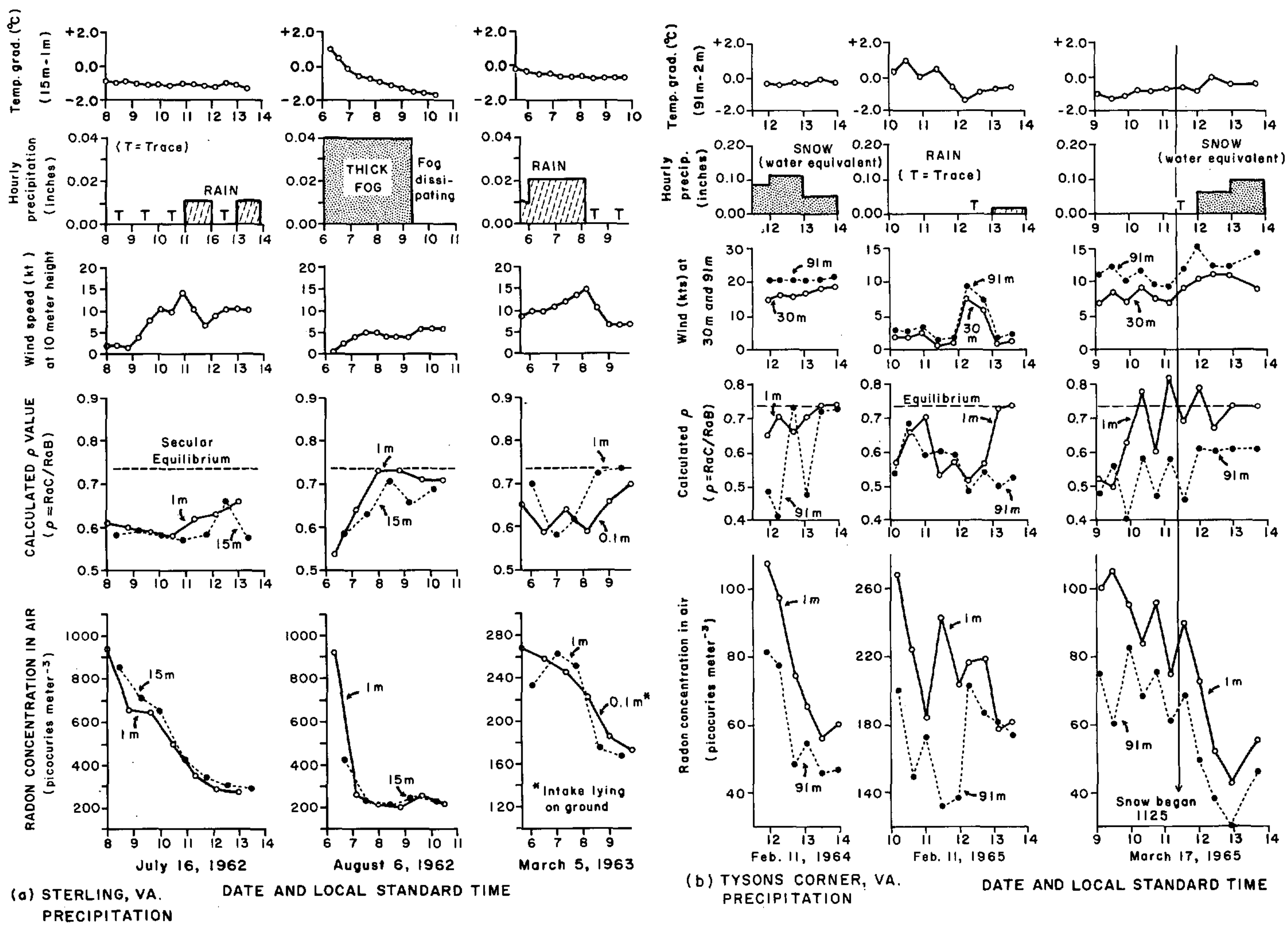

FIGURE 9.—Radon concentrations during precipitation periods at (a) Sterling, Va., and (b) Tysons Corner, Va.

were made at Sterling on the 28th and 30th; these concentrations and the attendant weather conditions are shown in figure 8. The buildup of activity on the 28th, following the dissipation of a surface-based radiation inversion, is unusual; this concentration increase was accompanied by relatively low ρ-values, suggesting a continuous buildup of fresh radon near the ground. Analysis of the radiosonde soundings for the 28th (taken at the Sterling site) indicated a persistent subsidence inversion based no higher than about 1000 m. above ground; this stability was reinforced by a northeasterly flow of cool stable air below the subsidence inversion. This limitation to height for vertical mixing probably contributed to the increase in radon content throughout the shallow mixed layer beneath the subsidence inversion throughout the daytime convective period.

The reduction in air volume available for dilution at low altitudes persisted for several days in the Washington, D.C. area. This was reflected on the 30th by radon concentrations of about 200 pc.m.$^{-3}$ observed near the ground at midday; such concentrations are several times higher than normally observed at this site during midday convective conditions. The persistence of high concentrations (factors of 2–4 higher than average) of natural radioactivity near the ground *throughout the daytime period* may be a reliable index of air pollution potential. This suggests that measurements of radon activity may provide useful data for assessing atmospheric dilution for air quality studies. Also, as an indicator of atmospheric diffusion rates, radon does not undergo a change in phase nor is it subject to photochemical reactions that affect many of the other constituents normally measured for an air quality assessment.

PRECIPITATION

Previous discussions deal with radon measurements taken during dry ground, non-precipitation conditions. Radon concentration measurements taken during periods of rain, snow, and heavy fog are plotted in figures 9a and 9b for the Sterling and Tysons Corner sites, respectively. From these and other data collected over the past several years, it is impossible to differentiate any effects of washout of radon daughter-carrying aerosols from other factors, such as changes in soil condition altering exhalation rates, or changes in evaporation processes, or turbulence during precipitation periods. No depletion of radon activity in air near the ground that can be attributed directly to precipitation is apparent from these data.

3. RESIDENCE TIME IN AIR

The relationship of a derived ρ-value to age or residence time in the air of the radon can be approached several ways. First, a heterogeneous mixture (components of different ages) of radon is considered; then, atmospheric radon is assumed to be in secular equilibrium with its primary daughter, RaA, and the RaA concentration in the air is assumed to remain constant throughout the filtration period (20 min.). The following expressions can then be used for determining the mixtures of two radon portions of different ages:

$$N_a = \text{constant} \tag{4}$$

$$N_b = XN_a\left(\frac{\lambda_a}{\lambda_b}\right)(1-e^{-\lambda_b t_1})+(1-X)N_a\left(\frac{\lambda_a}{\lambda_b}\right)(1-e^{-\lambda_b t_2}) \tag{5}$$

$$N_c = XN_b\left(\frac{\lambda_b}{\lambda_c}\right)(1-e^{-\lambda_c t_1})+(1-X)N_b\left(\frac{\lambda_b}{\lambda_c}\right)(1-e^{-\lambda_c t_2}) \tag{6}$$

where N_a, N_b, and N_c are the respective atom concentrations of RaA, RaB and RaC *in the air;* λ_a, λ_b, and λ_c are the respective decay constants; and X and $(1-X)$ are the fractions of the total RaA associated with radon parcels of ages t_1 and t_2, respectively. For simplicity and realism, t_1 can be assumed to be infinite (i.e., >2 hr.); t_2 is a variable parameter that represents the age of the second (fresh) radon component.

Figure 10 shows the relative air concentrations of RaB and RaC (ρ-values) for various amounts of fresh radon of age, t_2. With the assumption that a ρ-value of 0.5 represents the radon conglomerate at a height of 15 m. during windy conditions (fig. 1), this value can be interpreted to typify an infinite number of fractional compositions of fresh radon, varying from 100 percent of age 32 min. to about 68 percent of age <1 min. This approach illustrates the effect of fresh radon, deficient in daughter products, in producing a lower ρ-value, as well as the shortcomings of the parcel method for describing the age of radon conglomerates.

For meteorological considerations, determining an effective age or residence time for the radon conglomerate, rather than for its components, may be helpful in an assessment of various meteorological processes (Karol' and Malakhov [7]). In this approach, the residence time, τ, is defined as the average time since a radon atom entered the atmosphere unassociated with a RaB atom; this is not the same as the residence time (τ_c) referred to previously. By assuming either (a) an instantaneous source of fresh radon or (b) a continuous

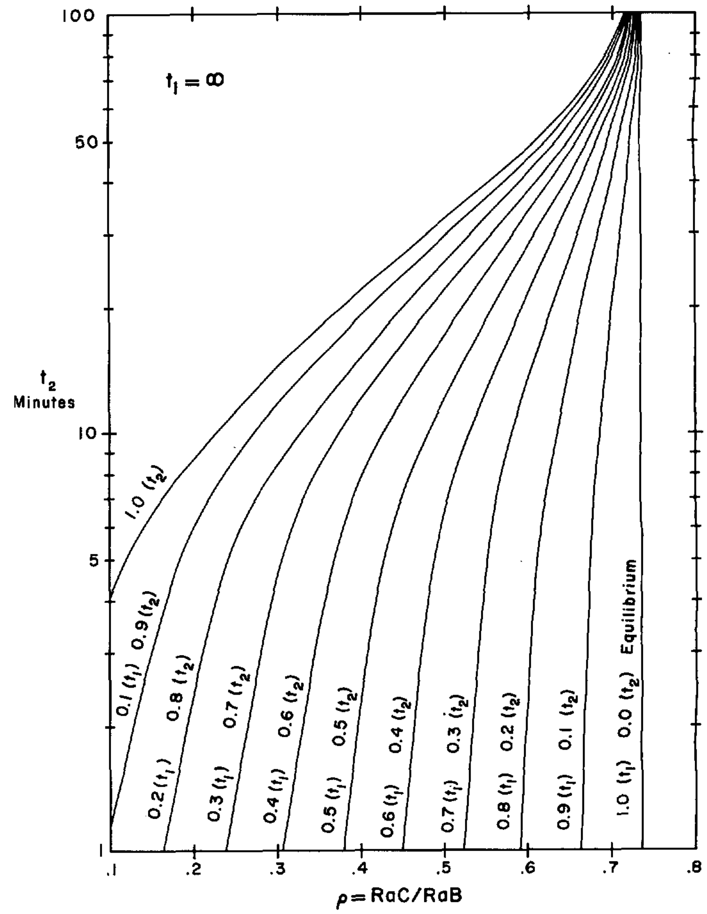

FIGURE 10.—Relationship of RaC/RaB atom ratio in air for various fractions of radon components of ages t_1 and t_2.

emission of fresh radon, the relationship between τ and ρ (fig. 11) and between τ and the radon/RaB activity ratio (fig. 12) in the air can be determined from a minute-by-minute calculation of the growth of the radon decay products, RaA, RaB, and RaC. This calculation provides a range of τ-values likely to be observed in the atmosphere for a given measurement of ρ. Intuitively, τ-values for the continuous emission are considered more representative of atmospheric conditions near the ground. The correction required to relate measured daughter activity (RaB) to radon concentration, for recent additions of fresh radon into the air, is determined from an analysis of the activity ratio versus τ in figure 12. This points out the fact that radon concentrations, calculated from measured daughter product activity, may be underestimated if secular equilibrium between radon and its daughter products is assumed to prevail near the ground.

On the basis of derived ρ-values obtained at near-ground elevations (figs. 1 and 4), the ages (τ) of the radon conglomerates determined from figure 11 for the Sterling site are listed in table 1 for different meteorological conditions. For comparison, even though the physics is entirely different, the effective residence time of RaC (τ_c) is listed in table 1. The tendency for a shorter effective residence

TABLE 1.—*Effective residence times in atmosphere of radon (τ) and RaC (τ_c) for various weather conditions at Sterling, Va.*

Height Above Ground (m.)	Weather		Average ρ-value	Effective residence time (min.)	
	Thermal Stability	Wind (kt.)		Radon (τ)	RaC (τ_c)
1	unstable	<16	0.62	100<τ<215	140
		>16	0.55	75<τ<145	85
	inversion	< 5	0.38	42<τ< 70	32
		> 5	0.50	64<τ<115	63
15	unstable	<16	0.64	105<τ<215	200
		>16	0.49	62<τ<110	58
	inversion	< 5	0.44	53<τ< 88	50

time of the radon conglomerate during inversion light wind conditions is interpreted to reflect the accumulation of fresh radon, deficient in daughter products, in the air near the ground during stable conditions.

4. METEOROLOGICAL APPLICATION

With a half-life of 3.825 days, radon may be considered a conservative constituent of the atmosphere for short-period (several hours) studies; it may serve as an atmospheric tracer to indicate the behavior of materials released to the air from a ground area source. Also, the variability of the radon daughter composition in the air may offer interesting clues to physical processes occurring in the boundary layer (Karol' and Malakhov [7]).

One of the potential sources of error involved in calculations of vertical diffusivity from radon profiles is the variation in radon exhalation rates. However, Wilkening and Hand [24] reported that the radon flux at the earth-air interface is relatively constant over a diurnal period, for a site in New Mexico; more recently, Sisigina [22], Kraner et al. [11], Schroeder et al. [20], and Pearson [18] have reported that the variations of radon exhalation rates from a given type of soil or bedrock surface are small, relative to variations in atmospheric concentrations of radon accompanying changes in meteorological conditions. This fact, together with the recent observations by Sisigina [21] and Kirichenko [8] of the vertical distribution of radon relative to changing meteorological conditions, plus the observations reported herein, suggests that the vertical profile of radon concentration near the ground may be a reliable indicator of atmospheric diffusion processes. In particular, the vertical concentration gradient may provide vertical diffusivity estimates under selected conditions. This approach will be used in future investigations, calling for a more rigorous measurement and analysis of turbulence statistics and vertical radon profiles over the 91-m. height interval at the Tysons Corner tower site.

5. SUMMARY AND CONCLUSIONS

Based on the premise that the RaC/RaB atom ratio (ρ) in the air is a measure of the extent of radioactive equilibrium that exists between radon and its decay products in the atmosphere, ρ can be calculated to deter-

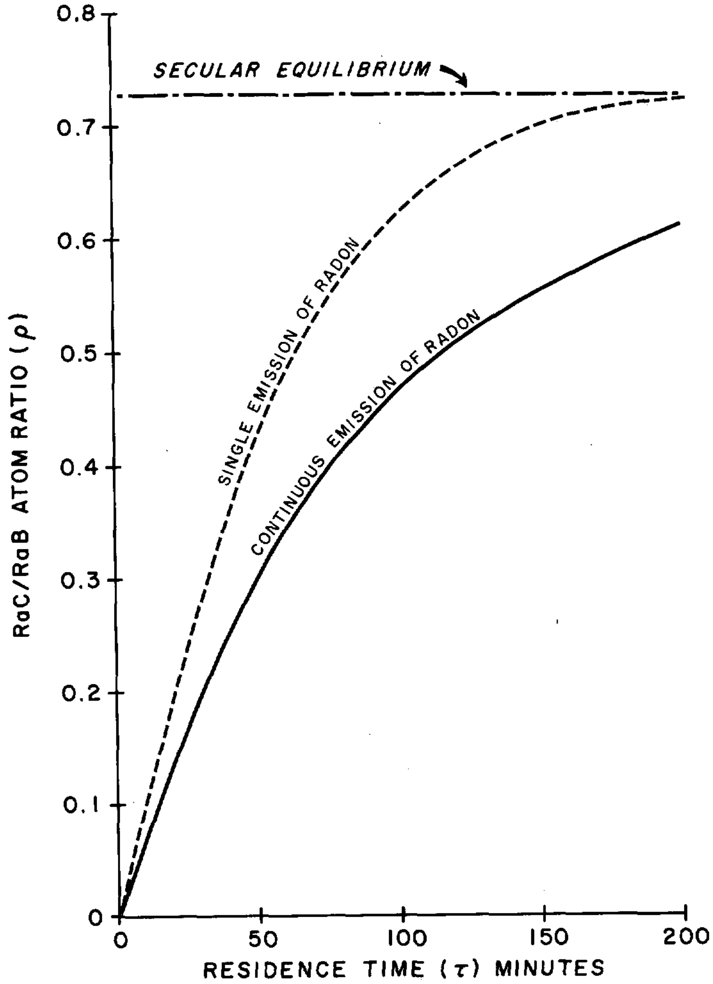

FIGURE 11.—Change in the RaC/RaB atom ratio (ρ) with residence time (τ) of the radon conglomerate after exhalation of radon gas from the soil.

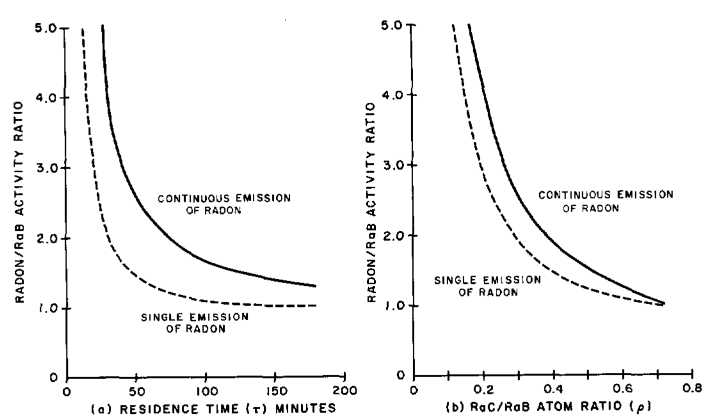

FIGURE 12.—Relationship of the radon/RaB activity ratio with (a) residence time (τ) and (b) RaC/RaB atom ratio (ρ) after exhalation of radon gas from soil.

mine an effective residence time in the air of the radon conglomerate filtered. In summary, these experiments show or suggest the following:

(a) Near the ground, surface winds in excess of about 16 kt. may suck out fresh radon from the topmost layers of soil. This may result from micro-oscillations in barometric pressure associated with mechanical turbulence during windy conditions.

(b) A departure from secular equilibrium between radon and its daughter products, RaB and RaC, prevails at heights of 1 and 15 m. above ground. This is attributed to the proximity of the radon source to these low altitudes; that is, a significant fraction of the radon conglomerate is fresh and thereby deficient in daughter products.

(c) A greater departure from secular equilibrium is observed during inversion conditions than during daytime convective periods at 1- and 15-m. heights.

(d) Preferential removal of RaC, relative to RaA and RaB, in the air would yield low ρ-values which, in turn, would indicate secular equilibrium departure; such preferential scavenging is difficult to prove or disprove from these investigations.

(e) Normally, near the ground, radon concentrations increase during the nocturnal stable period, reaching a maximum near sunrise; following the breakup of a nocturnal surface-based radiation-type inversion, radon concentrations decrease rapidly, reaching a minimum during the afternoon convective period.

(f) At two sites near Washington, D.C., radon concentrations at a height of 1 m. above ground ranged from 30–150 pc. m.$^{-3}$ during the daytime unstable period to 100–1000 pc. m.$^{-3}$ during the nocturnal inversion period. During stagnant weather conditions that persisted for several days, daytime concentrations were higher than normal by a factor of 2–4.

(g) Apparent "fumigation" has been observed at both sites, usually within 2 hr. following sunrise. This phenomenon is characterized by a higher radon concentration aloft than at the surface for brief periods following the breakup of a ground-based inversion.

(h) Any washout of radon or radon daughter-carrying aerosols near the ground by precipitation is not readily apparent.

(i) Variations in the earth's electric field may affect the radium ion balance in the atmosphere, particularly in the vicinity of towers.

APPENDIX: RADON DECAY SERIES

The decay products of the uranium series are introduced into the atmosphere through the rare gas, radon, which is exhaled from the soil into the air, where it sets up its own decay series as follows:

$$\text{(a) } Rn^{222} \xrightarrow[\text{3.8 days}]{\alpha} RaA\ (Po^{218}) \xrightarrow[\text{3 min.}]{\alpha} RaB\ (Pb^{214}) \xrightarrow[\text{26.8 min.}]{\beta^-}$$

$$RaC\ (Bi^{214}) \xrightarrow[\text{19.7 min.}]{\beta^-} RaC'\ (Po^{214}) \xrightarrow[10^{-4}\text{ sec.}]{\alpha} RaD$$

$$\text{(b) } RaD\ (Pb^{210}) \xrightarrow[\text{22 years}]{\beta^-} RaE\ (Bi^{210})$$

$$\xrightarrow[\text{5 days}]{\beta^-} RaF\ (Po^{210}) \xrightarrow[\text{140 days}]{\alpha} Pb^{206}\ \text{(stable)}$$

Concentration of RaD and its daughter products is negligible because of the long half-life of RaD and its continuous removal by precipitation; consequently, most of the natural radioactivity of the uranium series of elements in the atmosphere is due to radon and its daughter products, RaA, RaB, RaC, and RaC′. Airborne decay products of the thorium series generally account for only a few percent of the total natural radioactivity of the atmosphere.

The extent of radioactive equilibrium between radon and its daughter products in the air can be expressed by the atom concentration ratio (ρ) in the air, defined as ρ=RaC/RaB. On the basis of half-lives (minutes), ρ has a value of 0.735 when radon is in secular equilibrium with its daughter products.

$$\rho = RaC/RaB = 19.7/26.8 = 0.735$$

Any normal departure from this equilibrium results from a deficiency of RaC relative to RaB or a $\rho<0.735$, since any depletion of the precursors, RaA or RaB, also removes RaC and does not affect the value of ρ. A condition of $\rho>0.735$ can exist only if there is active removal of radon or RaA from the presence of its β-emitting daughter products, or depletion of RaB relative to RaC by some mechanical process.

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