Results of continuous measurements of radon and its decay products in the lower atmosphere

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ABSTRACT

With a new continuous measurement method, the radon content of the atmosphere at different levels above ground between 0.5 and 4 m was recorded at Aachen, Germany. Evaluation of simultaneous measurements of the radon daughters with a stepwise progressive filter (Schrittfilter) showed a close relation between radon and its daughters and the meteorological factors at the measuring place. Questions discussed in the paper include the daily variations of the radon concentration in relation to the exchange (Austausch), the radioactive equilibrium of radon and its daughters at different heights, and influences of the wind direction. It was found that during temperature inversions the concentration of radon and its daughters was enlarged whilst on overcast days the mean daily variation of both the radon and the radon daughter concentration was smaller. Moreover, the radon and thoron daughters were correlated with the dust content of the air.

1. Introduction

At the "Institut für Geophysik und Meteorologie der Technischen Hochschule Aachen" a new method for the measurement and recording of the radon and thoron concentration of the atmosphere has been developed: The atmospheric air to be investigated is sucked through an ionization chamber of about 300 liter content, after having been cleaned from aerosols and radioactive inductions by an "absolute" filter (H. Israël and G. W. Israël, see this issue; see also G. W. Israël, 1965). Using this method, recordings of the radon content of the lower atmosphere were carried out at different places and up to several months at each place.

In addition to this method for investigations of the short-lived decay products, a filter method was used. The equipment was a so-called "Schrittfilter-Gerät" (W. Schröck-Vietor, 1962) working as follows:

Dust is collected on the filter by aspiration for a given time (one to several hours, respectively) without being moved during this time. After the aspiration, the filter band is removed to bring another, yet undusted part of it to the aspiration point. The sample is analyzed in the usual manner to separate the decay products of radon and thoron and the fission products, as well. This means that the sample has to pass a series of three detectors with adequate time intervals.

In this paper, some of the results we have obtained up to now concerning (1) the effects of meteorological influences on the radon content and (2) the equilibrium problem of radon and its decay products are reported.

2. Results

(a) *Variations caused by meteorological influences*

Hitherto, by using normal emanometric measuring methods, (see, e.g. I. H. Blifford *et al.*, 1956; H. Moses *et al.*, 1960; W. Jacobi, 1962; J. Servant, 1964; J. E. Pearson, 1965) there was researched the daily variation of radon and also, with the aid of several different filter methods, of the short-lived radon daughters (see, e.g. H. J. Gale & L. H. J. Peaple, 1958; M. H. Wilkening, 1959).

Fig. 1 shows the mean daily variation of the radon concentration (curves) measured with the new equipment and the concentration of the decay products (circles) given as percentage of the mean.

The full curve shows, in connection with the filled circles, the course during clear days. The behavior during overcast days is given by the

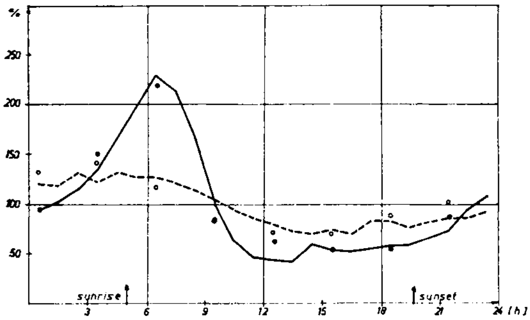

FIG. 1. Mean daily variations of radon (lines) and its decay products (dots) for clear and overcast days, given as percentage of the mean; clear days: full line and full dots; overcast days: broken line and little circles.

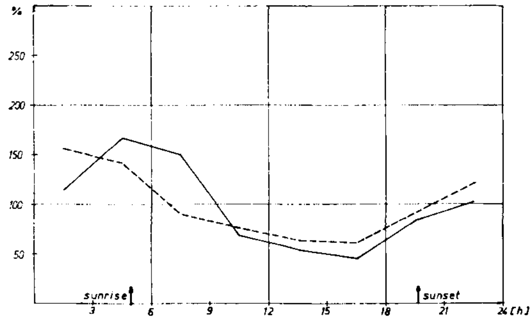

FIG. 2. Mean daily variations of ThB for clear and overcast days given as percentage of the mean.

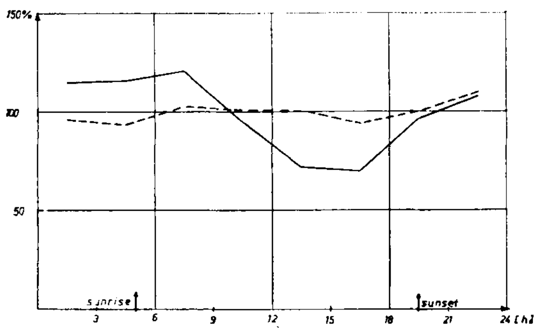

FIG. 3. Mean daily variations of air pollution at the test station for clear and overcast days given as percentage of the mean.

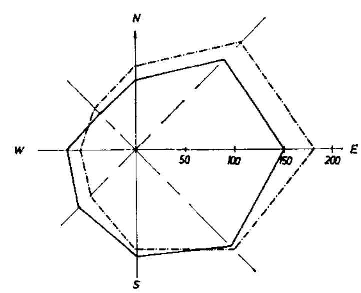

FIG. 4. The content of radon (full line) and RaB (dashed line) of the air versus the wind direction.

dashed curve and the open circles. The mean value of both the radon and its decay product concentration was found to be nearly the same during clear and overcast days ($0.7 \cdot 10^{-13}$ curie per liter) whilst, as expected, the course is quite different according to the different exchange conditions. The morning peak during clear days may be caused by an inversion built up by the heat radiation ("Ausstrahlung") of the ground during cloudless nights and the subsequent cooling of the air near the ground surface. This causes an enrichment of radon below the inversion layer, disappearing when the inversion vanishes after sunrise. It might be mentioned that a similar behavior has been found for the ThB content and the aerosol content of the air, as well.

Fig. 2 shows the daily variation of the atmospherical ThB density during clear days (full curve) and overcast days (dashed curve) in per cent deviation from the mean value found at $0.03 \cdot 10^{-13}$ c/l.

Both curves have a large fluctuation amplitude. In spite of this fact, the difference between the fluctuations at both weather conditions amounts to about 40 % maximally. In the average the difference is less than 20 %. During overcast days at sunrise, the concentration slope does not reach the correspondent value of the radon daughters. This result was expected, as a consequence of the long half-life of ThB (10 h); the decay rate lies essentially lower than for the short-lived radon daughters.

Fig. 3 visualizes very plainly the daily variation of the air pollution at the test place. A measure unit for the aerosol density was taken from the blackening rate of the filter spots of the "stepwise progressing filter arrangement" described above.

Fig. 4 shows the dependence of both the radon

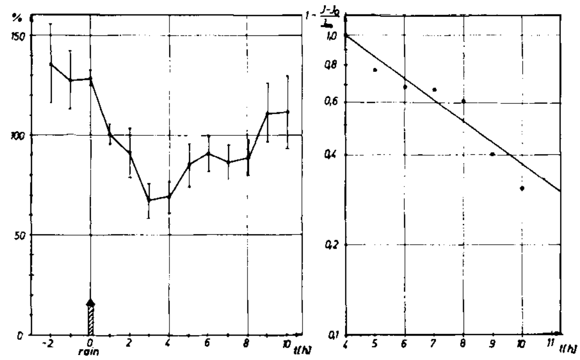

FIG. 5. The variation of the radon content of the lowest atmosphere in connection with rainfall.

content (full curve) and the RaB content (dashed curve) on the wind direction.

Although the two curves were found at different times, the course is quite the same. As explained already in an earlier publication (H. ISRAËL & S. STILLER, 1963), this dependence might be caused by the geographical situation of Aix-la-Chapelle, showing the well-known difference between the radon load of the sea-breeze and the air having flown over land for a longer time (Aix-la-Chapelle is situated at the south-east of the channel coast of Belgium and the Netherlands, distance about 200 km).

Fig. 5 shows a surprising reaction of the radon concentration within the lowest atmosphere in connection with rainfall. The left side shows the radon content from two hours before the start of rain until 10 hours after the end of rain. The duration time of the rainfall is condensed to the point 0 given by the arrow. Only rain duration times of less than 3 hours were considered and rainfall rates of more than 1 mm/hour, as well. During rain the radon content does not change remarkably, whilst after the end of it, the concentration drops to about half the value, within three hours. Then the radon content increases nearly exponentially (see the right side of the fig. 5) with a half-value-time of about 8 hours. Concerning the decay products, the material is not sufficient to give a corresponding picture. But it seems that the tendency of variation is about the same. Until now, we have not succeeded in finding a hypothesis explaining this effect. As published by G. W. ISRAËL (see this issue), the behavior of the thoron concentration has been found to be similar.

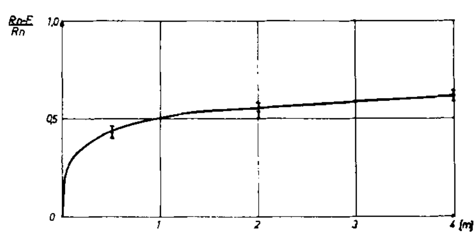

FIG. 6. The ratio of the concentration of the short-lived radon decay products ("Rn–F") to the concentration of radon ("Rn")—given in curie—in the first meters of the atmosphere.

(b) The equilibrium problem

Another question we were busy with is the problem of equilibrium between radon and its decay products (see, e.g., W. SCHMIDT, 1925; O. G. SUTTON, 1947, 1953; G. J. TAYLOR, 1935/36; H. LETTAU, 1951; W. JACOBI, 1962, 1963).

Integrating the exchange equation

$$\frac{\partial}{\partial z}\left(K \cdot \frac{\partial}{\partial z} c\right) - \lambda c = 0$$

$K = K(z)$, "Austausch"-coefficient
c = radon concentration
z = height coordinate (positive upward)
λ = decay constant of radon

with the usual assumptions we expect that the ratio between radon and its decay products will show a dependence upon the "Austausch"-coefficient.

Concerning its increase with height, the ratio between the radon and the decay products should approach the equilibrium ratio with increasing altitude.

Assuming the "Austausch"-coefficient to be proportional to the height, we may expect a course of the concerned ratio as shown in the curve of fig. 6. The curve is calculated by a numerical integration of the equation above, assuming a slope of the eddy diffusion coefficient according to

$$K = \alpha z + \beta, \quad \alpha, \beta = \text{const} > 0$$

and a radon exhalation rate of 1 atom/sec cm^2. The decay product rate in the air is taken to be zero at ground level.

Looking at the ratios measured at altitudes of 0.5, 2 and 4 m, they are found to agree very well with the calculated behavior. That means that the presuppositions made for the integration of the "Austausch"-equation were correct.

3. Conclusions

Summarizing the results presented above, we should conclude:

(1) The recordings of the radon content of the atmosphere and its variations show to be familiarly governed by meteorological influences.

(2) The investigation of the equilibrium status between the radioactive gases and their decay products is very important for studies of the exchange conditions, especially in the lowest region of the atmosphere.

(3) The use of such registrations as tracers for atmospheric changes promises interesting results concerning the "Austausch"-problem.

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РЕЗУЛЬТАТЫ НЕПРЕРЫВНЫХ ИЗМЕРЕНИЙ РАДОНА И ПРОДУКТОВ ЕГО РАСПАДА В НИЖНЕЙ АТМОСФЕРЕ

С помощью нового метода производились непрерывные измерения содержания в различных слоях атмосферы между 0,5 и 4 м от поверхности земли (Aachen, Германия). Оценка одновременных измерений продуктов распада радона с помощью ступенчатого фильтра (Schrittfilter) показала тесную связь между радоном, его дочерними продуктами и метереологическими факторами в месте измерения.

В этой статье обсуждаются в частности вопросы ежедневных изменений концентрации радона в связи с обменом (Austausch), радиоактивное равновесие радона и продуктов его распада на различных высотах и влияние направления ветра.

Было найдено, что во время инверсии температуры концентрация радона и продуктов его распада увеличивалась, в то время как в облочные дни среднедневная вариация этой концентрации была меньше. Кроме того, концентрация радона и дочерних продуктов радона была связана с количеством пыли содержащейся в воздухе.