Radon-daughter ions and their relation to some electrical properties of the atmosphere

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ABSTRACT

The mobilities and concentrations of the short-lived decay products of radon existing as positive small ions in the lower atmosphere are measured and compared with corresponding values for ordinary small ions in the atmosphere. Seventyfive per cent of the radon-daughter ions with mobilities greater than 0.06 $cm^2\ sec^{-1}\ volt^{-1}$ are found in the range of 0.25 to 1.50 $cm^2\ sec^{-1}\ volt^{-1}$. Ion characteristic curves give no evidence for distinct mobility groups within the resolution available. Radon-daughter ions on the average are present in the ratio of 3 parts per 100,000 of ordinary small ions in the atmosphere. A good correlation between radon-daughter ion concentration and total small-ion content is found over a range of 100 to 1200 small ions cm^{-3}. Radon-daughter ions are found to disappear almost completely at ground level under an active thunderstorm due to upward migration of the ions under the influence of strong electric fields. The importance of radon-daughter ions in the study of the electric environment of thunderstorms is indicated.

1. Introduction

The ions formed on the decay of radon-222 and its short-lived daughters have a finite probability of existing as positive ions for a short period following the disintegration process. They may then recombine to form neutral atoms either before or after attachment to aerosols as shown in Fig. 1.

Radon-222, RaA (Po^{218}), and RaC′ (Po^{214}) all decay with the emission of an alpha particle. The recoil velocity of the residual nucleus and perturbations in the wave functions of the electrons in the outer orbits due to a sudden change in nuclear charge are sufficient to produce ionization with reasonable probability. (KNIPP and TELLER, 1941). The beta emitters RaB (Pb^{214}) and RaC (Bi^{214}) can be expected to have at least a single charge since the process of beta emission (negatrons in both cases) is equivalent to the addition of a single positive charge to the nucleus. Some characteristics of radon-daughter ions are investigated in what follows.

2. Mobility distribution of radon-daughter ions

The mobilities of the radon-daughter ions have been measured in order to see how they compare with values obtained for ordinary small ions in the atmosphere. Fig. 2 shows the apparatus used. The outer tube is 15.6 cm in radius by 295 cm long. The central electrode is bare copper wire, 0.25 mm in radius. The negative potential applied to the central wire can be varied continuously from zero; however, a limit of the order of −4000 volts must be observed in order to avoid corona and the resultant recombination that takes place when corona currents exist.

The airflow is monitored by means of a hot-wire anemometer probe. The mean stream velocity is variable over a range of 0.41 to 3.1 m/sec. Turbulent flow obtains at these speeds with a Reynolds number of 20,000 occurring at 1.0 m/sec. This condition places a limit on the ability to resolve closely spaced mobility groups. Large volumes of air (260 m^3 at 1.0 m/sec in a 60 minute collection) must be processed in order to reduce statistical errors in counting.

Experiments to determine mobilities were performed in the early morning hours shortly before and after sunrise in order to make use of the higher concentrations of these ions found at this time of day and in order to take advantage of a closer approach to equilibrium

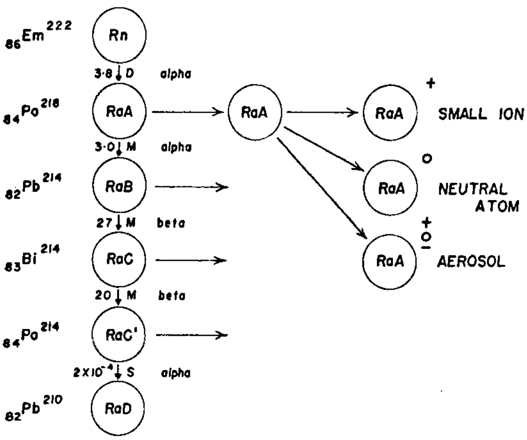

FIG. 1. Radon-222 and its short-lived daughter products.

of the radon daughters with the parent substance usually occurring at this time.

The method used in measuring the mobiltiy spectrum is shown in Fig. 3. For a mean air velocity U and a negative wire potential V, the limiting mobilities K_i for each of the equal sections of the wire are given by the relation

$$K_i = \frac{U(R^2 - r^2)}{2\,VL} \ln \frac{R}{r} \quad \text{where} \quad i = 1, 2, 3, 4, 5$$

R and r are the radii of the cylinder and wire respectively, L is the distance measured from the entrance of the cylinder, and l is the length of each section of wire so that $L = il$.

In a given run a ratio of U/V is selected and ions are collected for a time t, usually 60 min. At the end of a collection the total length of wire is cut into 20 equal lengths for counting under an alpha scintillation detector. The activity of each subgroup of four wires is combined to give the activity distribution along the

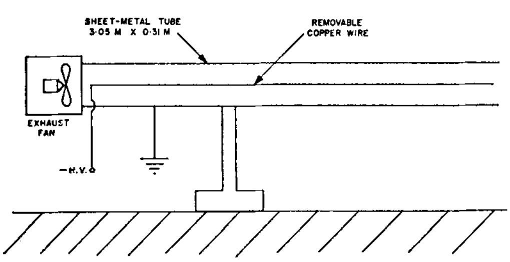

FIG. 2. Radon-daughter ion collector used in mobility measurements.

central wire as in Fig. 3 where the results for U/V values of 0.5, 0.1, and 0.05 cm sec^{-1} volt^{-1} are shown. The three runs are normalized to 1000 counts per minute for the first 60 cm length of wire. For a U/V value of 0.05 cm sec^{-1} volt^{-1}, essentially all of the small ions entering the tube are collected in the first 60 cm length. This length corresponds to a $k > 0.67$ cm^2 sec^{-1} volt^{-1}. As U/V increases by a factor of 10 to 0.5 cm sec^{-1} volt^{-1}, only all ions with $k > 1.3$ cm^2 sec^{-1} volt^{-1} are collected and over $\frac{3}{4}$ of the activity carried by lower mobility ions escapes from the cylinder. These data indicate clearly that the apparatus functions in a reliable manner and that most of the activity is attached to ions with mobilities greater than 0.67 cm^2 sec^{-1} volt^{-1}.

A distribution of mobilities can be obtained from the data represented in Fig. 3. Let N^{I} N^{II}, N^{III}, N^{IV}, and N^{V} be the activities at the end of the collection time on each of the five equal wire segments having limit mobilities of k_1, k_2, k_3, k_4, and k_5 respectively. In practice all 20 wire subsegments were counted for 3 min each at three separate time intervals so their activities at $t = 0$ could be determined by extrapolation. If it is assumed that the ions are distributed at the mobilities, k_1, k_2, k_3, k_4, k_5

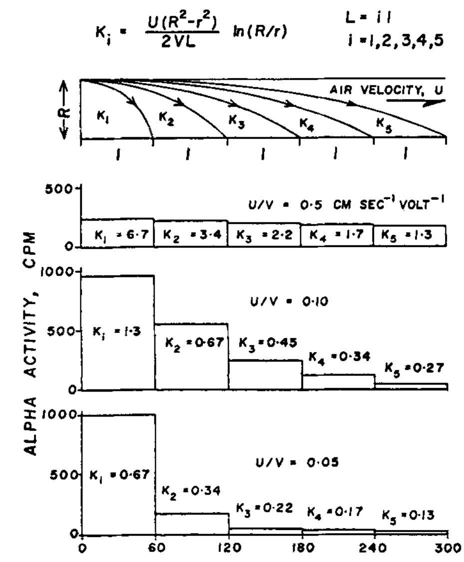

FIG. 3. Method used in measuring the mobility spectrum. Distribution of activity along the wire due to radon small ions is shown for three values of U/V.

with activities N_1, N_2, N_3, N_4, N_5, the activity N_5 will be N^{V} plus a like amount on the other four segments.
Hence,

$$N_5 = 5N^{V}$$

and similarly

$$N_4 = 4(N^{IV} - N^{V})$$
$$N_3 = 3(N^{III} - N^{IV})$$
$$N_2 = 2(N^{II} - N^{III})$$
$$N_1 = N^{I} - N^{II}$$

The net activities N_1, N_2, N_3, N_4, and N_5 appropriate to a given set of mobilities are then determined dependent on a certain combination of U and V values.

Results are given in Fig. 4 which represents the composite of 14 separate runs. On the assumption that the activities in the different mobility ranges are proportional to the number of ions present, one concludes that 46 % of the ions studied have mobilities from 0.25 to 0.75 cm^2 sec^{-1} volt^{-1}, 75 % of the ions have mobilities from 0.25 to 1.50 cm^2 sec^{-1} volt^{-1}, 20 % of the ions have mobilities greater than 1.50 cm^2 sec^{-1} volt^{-1}. Clearly radon-daughter ions have mobilities similar to the small ions of the atmosphere which are in the range of 1 to 2 cm^2 sec^{-1} volt^{-1} (CHALMERS 1957).

To investigate the identification of discrete ion mobilities, ion characteristic curves for several typical runs are shown in Fig. 5. Curves I, II, and III are from the present study and have U/V values corresponding to lower mobility limits of 0.13, 0.27, and 0.54 cm^2 sec^{-1} volt^{-1}. In no case can a specific mobility be identified with certainty. Curve I shows saturation at an L value of 150 cm corresponding to a mobility of 0.26 cm^2 sec^{-1} volt^{-1}.

It is interesting to note that discrete mobilities have been found by other investigators. The data of FONTAN (1964) using filtered air is plotted in Curve IV. Breaks in the curve can be identified corresponding to reported values of k of 0.82 cm^{2}sec^{-1} volt^{-1} and 1.65 cm^2 sec^{-1} volt^{-1}. VOHRA (1964) reports 100 % efficiency in his lowvolume radon-thoron ion collector when ions with mobilities greater than 3.5 cm^2 sec^{-1} volt^{-1} are collected.

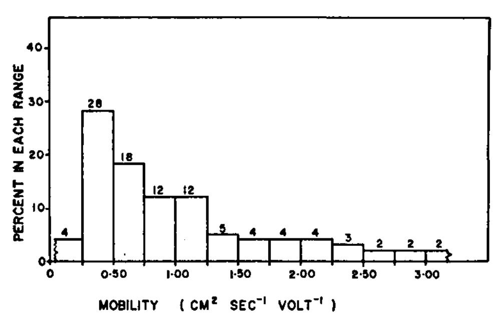

FIG. 4. Mobility distribution of radon-daughter small ions in the atmosphere.

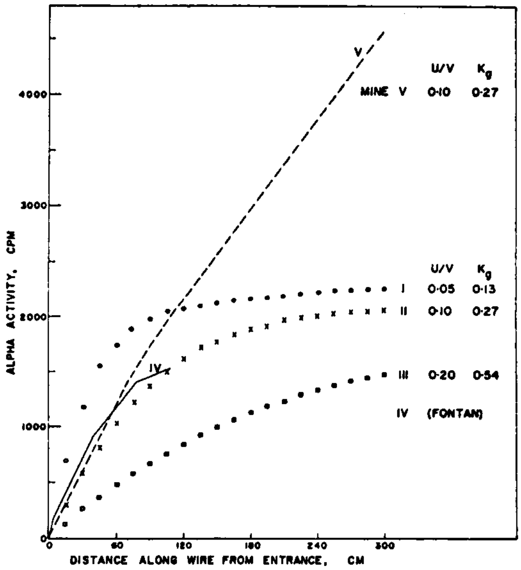

FIG. 5. Characteristic curves for radon-daughter ions.

The environment of the air in which the mobilities of the radon-daughter ions are measured can greatly alter the mobility pattern. Curve V of Fig. 5 is from data taken in an abandoned mine driven into the side of a mountain on the campus of the New Mexico Institute of Mining and Technology. Analysis of rock samples taken from the tunnel show no anomalous amounts or uranium, yet radon concentrations are of the order of 100 times that found in the free air. Radon-daughter ion mobility data taken in the mine reflect the higher concentrations, but again there is only weak evidence for any discrete mobility groups. A large majority of these ions in the mine tunnel had mobilities less than 0.2 cm^2 sec^{-1} volt^{-1} which would place them in the so-called "intermediate ion" group (STANLEY, 1964).

3. Concentration of radon-daughter ions

The total concentration of radon-daughter ions can be obtained by use of the ion characteristic curves of Fig. 5. Extrapolation of

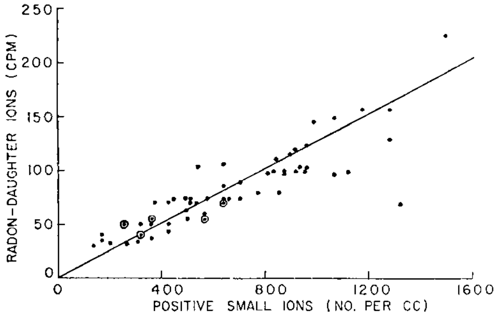

FIG. 6. Correlation between the concentration of radon-daughter ions and the total small-ion content of the atmosphere.

Curve I gives a value of 2300 counts per minute for that particular run. A mean of such runs is 1500 alpha counts per minute which are those due to RaC′ (Po^{214}) only, since in the counting of the wire samples the 3.08 min RaA has effectively disappeared. The counting efficiency ε of the scintillation detector used was determined to be 0.10 ± 0.05 using calibrated sources. The concentration of RaC atoms q_c can then be determined from

$$q_C = \frac{N}{\varepsilon \lambda \pi R^2 U t}$$

where N is 1500 counts per minute, λ the effective decay constant, $\pi R^2 U$ is the volume flow rate (4.7 m^3/min for $U = 1$ m/sec) and t is the collection time.

If all four short-lived radon daughters are assumed to contribute equally to the positive small-ion population, one gets an average concentration of 2×10^{-2} ion cm^{-3}. RaC′ is excluded because of its short life compared with the mean life of positive small ions in the atmosphere.

Measured values of total positive small-ion concentration under fair-weather conditions in the same vicinity give 550 ions cm^{-3} (JONASSEN and WILKENING 1965). Based on this value, radon-daughter small ions in the atmosphere account for

$$\frac{2 \times 10^{-2} \text{ ion cm}^{-3}}{550 \text{ ion cm}^{-3}} = 3 \times 10^{-5}$$

of the total positive small-ion content. This fraction is subject to large fluctuation of a factor of 10 or more over extended periods of time and should be considered as an order of magnitude only.

Based on measurements of the mean radon concentration (0.26 picocurie per liter) and the activity due to the radon ions, it can be shown that the radon daughter small ions account for approximately 8 per cent of the total activity from radon and its daughter products.

4. Correlation between radon-daughter ions and the total small-ion content of the atmosphere

Since the radon-daughter ions are known to carry positive charges, and they have been shown to have mobilities falling in the ranges of ordinary atmospheric small ions, their concentrations in the atmosphere under normal fair-weather conditions should be subject to the same fluctuation that is observed for the total positive, small-ion content of the atmosphere. The total small-ion content is calculated from data obtained with a Gerdien-type apparatus operated as a conductivity meter assuming proportionality between conductivity and small-ion content. When the results of simultaneous determinations of the radon-daughter ion content and the total small-ion content are plotted under fair-weather conditions as shown in Fig. 6, the correlation coefficient is +0.8. Hence, the hypothesis that the radon-daughter ions in the atmosphere behave as ordinary small ions under conditions of these experiments in the clear air of the American Southwest is confirmed.

JONASSEN and WILKENING (1965) have shown that for fair-weather conditions both in the Rio Grande valley of Central New Mexico and on a nearby mountaintop, the atmospheric conductivity is directly proportional to the concentration of small ions. Hence, the importance of radon-daughter ions as "tracers" in experiments under these conditions is apparent.

5. Effect of thunderstorms on the radon-ion content of the atmosphere

A striking example of the participation of radon ions in the gross electrical structure of the atmosphere is exhibited by the effect of the electrical field of a thunderstorm on radon-ion concentrations measured at ground level beneath the storm (WILKENING and KAWANO 1961, WILKENING 1964). Fig. 7 shows the electric field, radon-daughter ion activity, and radon concentration as a thunderstorm moved over a

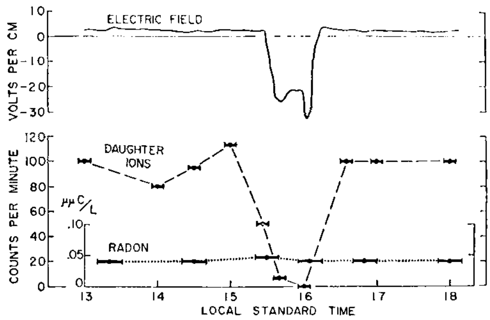

FIG. 7. Decrease in the radon-daughter ion content of the atmosphere due to the electric field of a thunderstorm.

mountain station. The disappearance of the positive daughter-ions is attributed to the presence of strong "negative" electric fields associated with thunderstorms (CHALMERS, 1957). The radon-daughter ions have a sufficiently high mobility (about 1 $cm^2 sec^{-1} volt^{-1}$) so that under the influence of strong electric fields associated with thunderstorms (>20 volt/cm) the drift velocity of the ions is sufficient and their mean life in the charged state is ample (many seconds to minutes) to remove the radon-daughter ions from near ground level. Work is continuing in our laboratory on the use of radon-daughter ions in the "thunderstorm effect" and other phenomena related to the fair-weather and thunderstorm electrical environments.

6. Summary

Radon and its short-lived daughters in the atmosphere form positive ions at the instant of decay. Measurements on the mobilities of these ions indicate that they fall in the "small ion" category. Seventy-five per cent of the ions are found to have mobilities in the range of 0.25 to 1.50 $cm^2 sec^{-1} volt^{-1}$. No evidence is found for distinct mobility groups.

Although wide variations are noted, there are on the average only 3 ions per 100,000 ordinary small ions in the atmosphere. A correlation coefficient of +0.8 is obtained between the radon-daughter ion concentration and total small-ion content over a small ion concentration range of 100 to over 1200 ions cm^{-3}.

An interesting application of the use of radon-daughter ions in the atmosphere is found in the study of the electrical environment of a thunderstorm. Radon ions disappear almost completely under an active storm due to an upward migration of ions under the influence of strong electric fields.

Acknowledgement

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ИОНИЗИРОВАННЫЕ ПРОДУКТЫ РАСПАДА РАДОНА И ИХ СВЯЗЬ С НЕКОТОРЫМИ ЭЛЕКТРИЧЕСКИМИ СВОЙСТВАМИ АТМОСФЕРЫ

Измерены подвижности и концентрации короткоживущих продуктов распада радона, существующих в нижней атмосфере в виде положительных ионов. Эти величины сравниваются с соответствующими величинами для обычных ионов в атмосфере. 75% рассматриваемых ионов с подвижностями, большими 0,06 $см^2/сек^{-1}/вольт^{-1}$ найдены в интервале от 0,25 до 1,50 $см^2/сек^{-1}/вольт^{-1}$. Характеристические кривые для ионов в пределах имевшегося разрешения не дают возможности различить группы ионов по их подвижности. Ионы радонного происхождения присутствуют в атмосфере в среднем в количестве 3 на 100 000 обычных легких ионов. Найдена хорошая корреляция между концентрацией радонного происхождения и общим количеством легких ионов в интервале их концентраций от 100 до 1200 на $см^3$. Найдено, что дочерние ионы радона почти полностью отсутствуют на уровне земли, вследствие сильных гроз благодаря направленной вверх миграции ионов и сильных электрических полях. Указано на важность ионов радонного происхождения при изучении электрического поля вблизи гроз.