FOURTH INTERNATIONAL SYMPOSIUM ON
THE NATURAL RADIATION ENVIRONMENT
LISBOA, PORTUGAL, DEC. 7-11, 1987

CONF-871208--4

DOE/ER/60662--7

DE89 009375

RADON-222 AND ITS SHORT-LIVED DECAY PRODUCTS IN ATTACHED AND FREE-ION FORMS IN THE ATMOSPHERE

MARVIN WILKENING

AND

EDWARD McNAMEE

DEPARTMENT OF PHYSICS

NEW MEXICO INSTITUTE OF MINING AND TECHNOLOGY

SOCORRO, NEW MEXICO 87801 USA

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

SHORT TITLE: DECAY PRODUCTS OF ^{222}RN IN FREE-ION FORM

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

RADON-222 AND ITS DECAY PRODUCTS IN ATTACHED AND FREE ION FORMS IN THE ATMOSPHERE

MARVIN H. WILKENING
AND
EDWARD McNAMEE

ABSTRACT

The short-lived radionuclides in the radon decay series account for a major part of the internal dose under normal conditions. While most of this comes from atoms attached to aerosols, a significant component is due to decay products in the form of positive ions. In this study the average indoor ^{222}Rn level of 32 Bq m^{-3} yielded an activity of about 2 Bq m^{-3} from the positively charged ions. Out of a total potential alpha-energy concentration of 7.1×10^{-8} J m^{-3}, the positive decay-product ions accounted for two percent. The nuclides of the radon decay series are the chief ionizing agents in the lower atmosphere. The mean indoor ^{222}Rn level in this work produces about 20×10^{6} ion pairs m^{-3} s^{-1}. This is sufficient to yield positive and negative ion densities in the air of 130×10^{6} m^{-3} and conductivity values of about 4×10^{-14} $(ohm - m)^{-1}$. The ^{222}Rn positive ions make up only about 7 ppm of the total positive ions present. Correlations among the measured quantities are given which aid in the understanding of ^{222}Rn daughters in indoor air.

INTRODUCTION

The contributions of the decay products of ^{222}Rn to the internal dose depend upon a number of factors including the degree of equilibrium between radon and the decay products and the fraction of daughters that exist in free ion or unattached form.

Details regarding the formation and properties of these ions have been treated previously[1-3]. The processes which allow the decay product ions to remain for a short time in their positive charge states, combine with other atoms in associative or chemi-ionization processes, and provide "clustering" with polar molecules such as water, have been studied by a number of investigators[4-6]. Regardless of the mechanisms involved, the fact that radon decay products begin their existence in the positive ion state makes them an interesting and important component of the atmosphere.

For normal negative ion densities and aerosol concentrations attachment processes exceed recombination in typical situations by a factor of 100 or more. This characteristic of the decay product positive ions accounts for the fact that most of the dose from radon and thoron is from atoms attached to aerosol particles. The concentration of these ions and their interaction with electrical parameters in the indoor atmosphere is treated in what follows.

ELECTRICAL PARAMETERS OF THE ATMOSPHERE

Ionization. Ion pair production rates for the indoor environment are enhanced due to elevated levels of radon and thoron. The mean indoor radiation levels in this study produce about 20×10^6 ion pairs m^{-3} s^{-1}. This is sufficient to yield positive and negative ion densities of the order of 130×10^6 m^{-3}.

Conductivity and Ion Densities. With ion densities of the levels characteristic of indoor air, it can be expected that the indoor atmosphere is a conducting medium. The conductivity and ion density of the atmosphere are related as shown in the relation $\lambda = N e k$ where λ is the total electrical conductivity, N is the small ion density, k is the mobility, and e is the electronic charge. The ion densities for positive and negative carriers (N^+ and N^-)

and the conductivities, λ^+ and λ^-, are approximately equal provided the mobilities are nearly the same as is generally the case. The mobility of the daughter ions which is a measure of their drift velocity in electric fields is in the range of 0.25 to 1.50 x 10^{-4} m^2 V^{-1} s^{-1} (7). This puts them in the same class as ordinary atmospheric small ions(8). It is these "small" ions that are responsible for electric currents and charge transfer phenomena in the atmosphere. The radon decay product positive ions make up a very minor component of the total positive ion density on a purely numerical basis.

INSTRUMENTATION

Positive and negative atmospheric small-ion concentrations and conductivity were measured with a Gerdien-type instrument(9). Radon and thoron concentrations as well as their decay products and the resulting potential alpha energy concentration (PAEC) were determined using an electrostatic precipitator system(10). The radon daughter positive ions were collected in a Gerdien-type apparatus similar to the conductivity apparatus except that the inner collector electrode is replaced by a lucite cylinder covered by a removable aluminum foil. Positive small ions with preset lower mobility limits are collected on the foil for a 10-minute period after which the daughter ion concentrations are determined by the Thomas method.

Aerosol concentrations were measured with a condensation nucleus (CN) counter and for the most recent work, an electrical aerosol analyzer was used. The CN counter is sensitive to aerosols in the 0.001 to 0.1 μm range, while the aerosol analyzer has a range of from 0.0032 to 1.0 μm. Data taken simultaneously indoors with both instruments yielded a high correlation coefficient (0.93). A meteorological station was operated outdoors near the building being used.

The sites chosen for this study were intended to provide a wide spread in values for the parameters being studied rather than for analysis of radon exposures. Measurements were made at three indoor locations. One was an unoccupied four-room, single story

house having concrete block walls and concrete slab floors. Due to cracks and poor sealing, the mean ventilation rate was 1.8 hr^{-1}[11]. The other two rooms were in a campus building; one was a first floor research laboratory and the other was an instrumentation and utilities room.

This study extended over a period of two years, and included a total of 83 days when data were recorded making both diurnal and seasonal programs available.

RESULTS

Radon and its Decay Products. Concentrations of radon, the members of its decay series, and aerosols are highly variable when examined on a time scale of the order of a day. An example is shown in Figure 1 which illustrates a midday peak aerosol concentration and large changes in PAEC and aerosol size as well. For purposes of this study values of the parameters during the early morning (AM), 0500-0700 Local Standard Time (LST) and (PM), 1400-1600 LST were used both to obtain typical high and low values for the room and to avoid large variation. No data were included from days when abnormal weather or unusual indoor events occurred. Figure 1 also shows that the radon decay product PAEC is a minimum during midday when aerosol concentrations are highest. Although the exposure is generally higher when the aerosol concentration increases, in this case the PAEC is simply following the ^{222}Rn which is, as expected, at a minimum during midday.

Radon and decay product levels are shown in Table 1 along with aerosol concentration and electric parameters. Instrumental errors in the values shown are of the order of 15%. The standard deviation representing the variability in a given parameter is much larger ranging from 25 to 50% or more. Mean ^{222}Rn concentration drops by a factor of 5 in going from the single story house (Room 1) to Room 3 on campus. The decreases in the PAEC for the "attached" daughters is not quite as dramatic for the three rooms and the exposure levels from the decay product positive ions are quite low at only 2.4% of that due to the attached daughters and with only a factor of two decrease between Room 1 and Room 3. Radon

and its daughter radionuclides showed a consistent ratio of 1.4 between AM and PM sampling periods. An average ratio of 8 was found between indoor and outdoor ^{222}Rn levels for the three rooms. This ratio decreased somewhat both for the attached decay products and the free ions. The data of Table 1 show the broad levels of ^{222}Rn and its decay products available for the analysis of the effects upon the atmospheric parameters.

Since ion-pair production depends primarily upon ^{222}Rn and its decay products, it can be expected that similar trends in positive and negative conductivities (λ^+ and λ^-), and the overall ion densities (N^+ and N^-) would occur as seen in the table also. Again AM to PM ratios are almost all greater than one averaging 1.4. Similarly the decay product positive free ions decrease in going from Room 1 to 3 in much the same manner in spite of their very low densities. The aerosols, on the other hand, have their primary source in room interiors and as a result PM values almost always exceed the AM levels. It can be seen in Figure 1 that aerosol diameters tend to be larger at night than during daytime hours with a range of 0.03 to 0.08 μm similar to previous measurements in the area[12].

Correlation Studies. Coefficients were calculated for different pairs of parameters. The ^{222}Rn decay product free ions show a high correlation with the total positive ion density even though the free ions are present in quantities amounting to only a few parts per million. Positive conductivity, λ^+, shows a good correlation in the AM hours with the total positive ion density, N^+, but large increases in aerosols in the daytime can reduce the relation to a negative (inverse) value. Aerosol concentrations show strong negative correlation with the AM ion density (-0.4), conductivity (-0.7) and the total PAEC due to the ^{222}Rn decay products (-0.9). As the aerosols become more abundant, ions attach and are lost for conductance due to a large decrease in mobility. The free ion concentration shows a high correlation with normal ion densities of either sign. On the other hand there is a negative (inverse) correlation between aerosol concentration and conductivity.

Morning (0500-0700) values of ^{222}Rn and the decay products in

both attached and free ion forms were all higher than afternoon (1400-1600) levels. The same was true for the total ion densities (N^+ and N^-) and the positive (λ_t^+) electrical conductivity. Aerosol concentrations though quite variable were usually higher in the afternoon. Of all parameters measured, only aerosols, positive and negative conductivity, and total ion densities showed higher values outdoors than indoors on the average. ^{222}Rn and the attached daughter product levels indoors exceeded corresponding outdoor levels by a factor of 8. The free ions from radon decay play important roles in the atmosphere both from their contribution to internal dose as well as for use as tracers in atmospheric processes.

CONCLUSIONS

The short-lived decay products of ^{222}Rn include an important component that exists as positively charged ions in the atmosphere. These ions account for only about 2% of the total PAEC associated with decay product attached to aerosols. Further, the positive free ions were found to be present in a typical indoor environment to the extent of only 7 parts per million of the total atmospheric small ions. The ^{222}Rn decay product free ions show a high correlation with the total positive ion density. Aerosol concentrations show negative (inverse) correlation with the AM values of ion density, conductivity and the total PAEC. As the aerosols become more abundant, ions attach and result in decreased mobility and less conductivity. Indoor values of ^{222}Rn and the decay products exceeded outdoor levels by a factor of 8. On the other hand aerosols, positive and negative conductivity, and the total ion densities were at lower levels indoors than outdoors.

ACKNOWLEDGMENTS

The authors are indebted to their colleague, Dr. S.D. Schery and to assistants Ed Johnson and Paul and Cindy Sisson.

The work was supported by contracts DE AS04 84ER60216 and DE FG04 86ER60432 with the Office of Health and Environmental Research of the U.S. Department of Energy and by the Geophysical Research Center of the New Mexico Institute of Mining and Technology.

REFERENCES

1. Busigin, A., VanderVooren, A.W., Babcock, J.C., and Phillips, C.R., The nature of unattached RaA (^{218}Po) particles, Health Phys. 40, 335-343 (1981).

2. Shimo, M., Asano, Y., Hayashi, K. and Ikebe, Y., On some properties of ^{222}Rn short-lived decay products in air, Health Phys. 48(1), 75-86 (1985).

3. Wilkening, M., Effect of radon on the electrical properties of indoor air, Radon and Its Decay Products, American Chemical Society, New York, NY, (1986).

4. Mohnen, V.A., Formation, nature, and mobility of ions of atmospheric importance, Electrical Processes in Atmospheres. Steinkopff, Darmstadt (1977).

5. Rudnick, S.N., Hinds, W.C., Maher, E.F. and First, M.W., Effect of plateout, air motion and dust removal on radon decay product concentration in a simulated residence, Health Phys. 45(2), 463-470 (1983).

6. Raes, F., A model for size distribution of radon decay products in realistic environments, Radon and Its Decay Products, American Chemical Society, Washington, DC (1987).

7. Bricard, J., and Pradel, J., Electric charge and radioactivity of naturally occurring aerosols, Aeros. Sci.(1), 91-104 (1966).

8. Chalmers, J.A., Atmospheric Electricity, Second Edition, Pergamon Press, New York, NY, 85-87 (1969).

9. Jonassen, N. and Wilkening, M.H., Conductivity and concentration of small ions in the lower atmosphere, J. Geophys. Res. 70(4), 779-784 (1965).

10. Andrews, L.L., Schery, S.D. and Wilkening, M.H., An electrostatic precipitator for the study of airborne radioactivity, Health Phys. 46(4), 801-808 (1984).

11. Zarcone, J.J., Schery, S.D., Wilkening, M.H. and McNamee, E.A., A Comparison of Measurements of Thoron, Radon and Their Daughters in a Test House with Model Predictions., Atmos. Environ. 20(6) 1273-1279 (1986).

12. George, A.C., Wilkening, M.H., Knutson, E.O., Sinclair, D. and Andrews, L., Measurements of radon and radon daughter aerosols in Socorro, New Mexico, Aerosol Sci. and Tech. 3, 277-281 (1984).

Table 1. The decay products of ^{222}Rn compared with atmospheric electrical parameters. (1mWL = 2.08×10^{-8} J m^{-3})

	Room 1	Room 2	Room 3	Indoor Mean	Outdoor Mean	Ratio Indr/Outdr
^{222}Rn (Bq m^{-3})	60	23	12	32	4.0	8.0
PAEC-attached ($\times 10^{-8}$ J m^{-3})	11	5.6	4.9	7.1	1.0	7.1
PAEC-free ions ($\times 10^{-8}$ J m^{-3})	0.25	0.12	0.12	0.16		
Aerosols ($\times 10^{9}$ m^{-3})	4.6	14	11	10	17	0.6
Conductivity, λ^+ $\times 10^{-14}$ (ohm-m)$^{-1}$	6.6	3.3	2.4	4.1	2.8	1.5
Conductivity, λ^- $\times 10^{-14}$ (ohm-m)$^{-1}$	8.4	2.4	2.6	4.5	3.1	1.5
Ion density, N^+ ($\times 10^{9}$ m^{-3})	0.17	0.10	0.13	0.13	0.17	0.8
Ion density, N^- ($\times 10^{9}$ m^{-3})	0.15	0.14	0.12	0.13	0.15	0.9
^{222}Rn free ions n^+ (no. m^{-3})	1420	780	660	950	760	1.2
Ratio: $\frac{n^+}{N^+}$ ($\times 10^{-6}$)	8.4	7.8	5.0	7.3	4.5	1.6

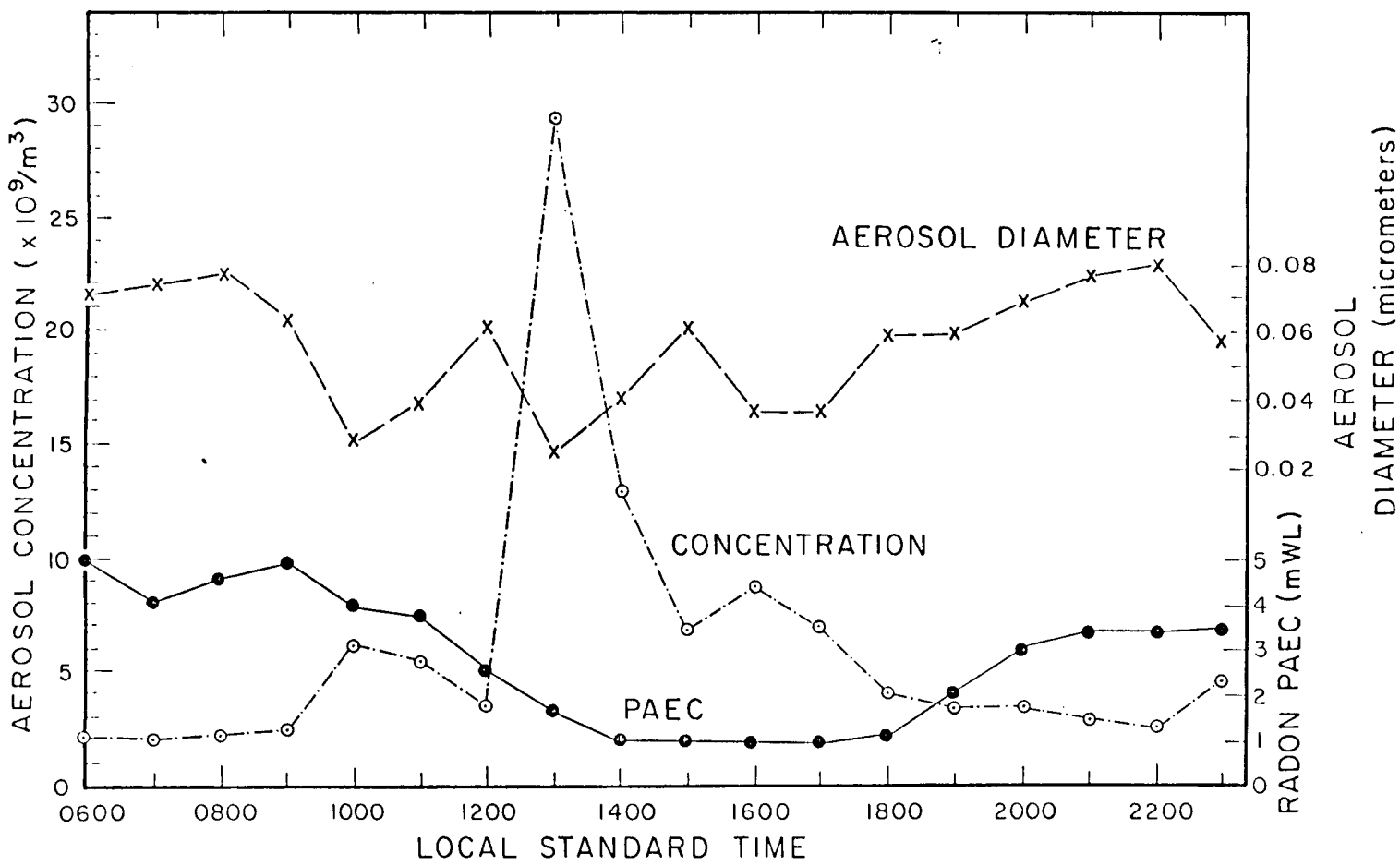

Figure 1. Typical day-night variation in exposure to ^{222}Rn decay products (PAEC) in comparison with aerosol concentration and size in an indoor environment. (1mWL = 2.08×10^{-8} J m^{-3}).