

# DETERMINATION OF RADON DAUGHTER PRODUCTS NEAR LAND-SEA DISCONTINUITIES BY GAMMA SPECTROMETRY

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**Abstract**—The low number of experimental measurements of the levels of radon daughter products in the air near exhalation source discontinuities has led us to carry out a long series of measurements of these levels. The correlations between these levels and different meteorological parameters have been studied.

## 1. INTRODUCTION

Determination of the concentrations in the air of short-life radon (Rn-222) daughter products is important in as much as it has a bearing on a wide variety of problems relating to the environment. Problems such as the irradiation of mineworkers' lungs, the origin and trajectory of air masses or the washing of atmospheric aerosols by rainfall (Wilkenning *et al.*, 1976; Stranden, 1979) may be studied by determining the concentrations in the environment of short-life radon daughter products (Bouville *et al.*, 1971; Subba *et al.*, 1969). Therefore, in view of the low number of experimental works to be found in the bibliography with respect to the measurements of these elements close to land-sea discontinuities, we have carried out a series of measurements of the concentrations in the air close to the ground of short-life gamma emitting radon daughter products.

## 2. EXPERIMENTAL METHOD

The measurements were carried out close to the Science Faculty of the University of Santander (Spain), located some 500 m from the sea in a semi-built-up area, with an undulating layer of lawn cover. Radon daughter products were captured by filtration, an airflow being directed through a Whatman 41 filter with a surface area of 440 cm<sup>2</sup>. A suction pump produces a flow through the filter of 100 m<sup>3</sup>/hr which, given the area of the filter, generates a through flow speed sufficient to ensure a 100% retention in the filter of those aerosols of the size in which radon daughter products become fixed (Dueñas, 1976). The filtration time was 30 min. Samples were taken in the early morning, the air intake being placed at a height of 1.5 m above ground level.

To measure the activity of the radon daughter products trapped in the filter, we used gamma spectrometry. For this purpose, we used a 3 × 3 in. NaI (Tl) scintillation detector connected to an amplifier, this in turn being coupled up to an Intertechnique Histomat multichannel analyzer which analyzes the activity found in the samples, the results being shown either directly on an oscilloscope screen or through a teleprinter. The detector was placed within a lead sheath, the internal dimensions of which were 60 × 60 × 60 cm, the walls being 10 cm thick and internally lined with sheets of zinc and cadmium to reduce background noise and the X-ray contribution of the lead. The filters were cut to a fixed geometry and placed on the upper face of the detector. The spectrometry chain was calibrated by means of an Ra-226 sample in equilibrium with its daughter products, with a known activity and possessing the same geometry as the filters.

To calculate the activity in the air of radon daughter products, we worked on the hypothesis that the radioactive equilibrium between the first, Po-218 or Ra A, and the second, Pb-214 or Ra B, daughter products is checked in the place of measurement. This hypothesis, a reasonable one by virtue of the different half-lives of the two elements, adds a condition to the

three well-known equations that express the accumulation in the filter of the first three radon daughter products, Ra A, Ra B and Bi-214 or Ra C, during the filtration period. Bateman's equations describe the evolution of the activity of the three daughter products in the filter once the filtration period had ended. The integration of these three equations between the times at which counting commences and ends allows three relations to be established between the activities in the air being sought and the number of counts corresponding to the gamma emitters detected. Through the radioactivity equilibrium mentioned above, the three relationships referred to may be reduced to two which makes it possible to calculate the activity in the air of Ra B and Ra C on the basis of the two data obtained from the gamma spectrum of the filters. The two data used were the number of counts below the triplet of energy levels 0.24, 0.29 and 0.35 MeV corresponding to Ra B and the number of counts below the peak of 0.61 MeV of Ra C, once the corresponding background noise has been allowed for. These photo-peaks appear well defined in the gamma spectra of the samples given the conditions of the filtration time and count time (15 min after filtration period used). Furthermore, we then carried out new counts of 15 min each in order to corroborate the radioactive decay of the Ra C in the filter. Using this method of several counts, it is also possible to determine the concentrations of this element in the air. The results obtained through these two methods are similar, although the second produces a greater degree of error.

The gamma spectrometry chain gives a detection efficiency for the geometry used of 9% in the region of the energy corresponding to the Ra B triplet and an efficiency of 6.5% in the region of the 0.61 MeV peak of Ra C. The sensitivity of the apparatus makes it possible to measure  $1.5 \cdot 10^{-14}$  curies/l of Ra C in the air under the experimental conditions described. The error calculated for the activities measured in no case exceeds 8% of the corresponding value.

### 3. RESULTS

Using this experimental method, we carried out a total of 140 determinations of the activity in the air of Ra B and Ra C over a period of two and a half years, between January 1976 and August 1978. The average activity values obtained were  $10.9 \times 10^{-14}$  curies/l for both Ra B and Ra C. In most of the measurements carried out, the values for the activities of both elements in the air were very similar, thus indicating a high degree of approximation in the radioactive equilibrium between them. For this reason, we will refer only to Ra C.

Bearing in mind that the stability of the atmosphere is necessarily a principal factor in determining the concentration of Ra C in the air, we classified the activity values obtained according to the different types of stability prevailing during the sample-taking period. The stability criteria were specified in terms of the wind speed, based on the oscillations refiltered on the band of an anemocinograph, the sunshine and the amount of cloud present. Applying these criteria, we tabled our measurements under six different types of stability, from type A (very unstable) to type F (moderately stable) following the criteria established by Pasquill (1968).

Table 1 shows the average values, in curies/l  $\times 10^{-14}$ , obtained for the different types of stability for which we possess statistics of 20 activity values or more, as well as the number of values for each type. Each average value is also accompanied by an indication of the characteristic deviation found for the total number of values measured for each type of stability. Examination of the table first reveals that the average values obtained for the activity of Ra C in the air, in all cases, lower than those found by other authors for the activity of radon in the air in continental regions (Biro *et al.*, 1970; Jacobi *et al.*, 1963). Recognising a

Table 1.

Stability	A	C	E	F
Ra C	$3.53 \pm 1.50$	$7.55 \pm 4.83$	$10.80 \pm 6.41$	$15.66 \pm 5.02$

radioactive equilibrium between these two elements for this comparison, the importance is clearly shown of the interchange of land air and sea air at the point where the measurements were carried out. In fact, since radon exhalation over the sea is much lower than on land, air masses travelling over the sea possess lower radon concentration than air masses with an overland trajectory. Therefore, the interchange of air occurring at a land-sea discontinuity should produce a lower value for radon concentrations in the air than that to be found in clearly inland areas, as the measurements carried out would appear to indicate.

Furthermore, it will be seen that as the module and oscillation of the wind speed increases, thus modifying the prevailing type of stability, the concentration of Ra C in the air decreases. This fact is no doubt related to the increase in the vertical diffusion of matter in the lower layers of the atmosphere due to the friction of the wind with the land. Parallel to this effect, we may assume the additional effect of the reduction of Ra C concentrations in the air during the more unstable periods due to the fact that the rainfall that usually accompanies such periods will reduce the exhalation of radon by obstructing soil pores (Malakhov *et al.*, 1962).

Finally, we can deduce from Table 1 that of the different types of stability it is types C and E that present the greatest variability with respect to the Ra C concentrations measured. The concentrations corresponding to the conditions of greatest and least stability are, on the other hand, more constant. Within each of the four types of stability studied, we have attempted to establish relationships between Ra C concentrations and different meteorological variables: atmospheric pressure, relative air humidity, air temperature, sunshine and the amount of rainfall. Nevertheless, we were only able to obtain reasonable relationships within the range of type F stability. The most clearly defined of these was that found between Ra C concentrations and the total sunshine recorded on the day preceding the day on which measurements were made.

Figure 1 shows the relationship between Ra C levels and the number of hours of sunshine recorded during the days previous to those on which measurements were taken. In this diagram the error assigned to each level has been calculated as the square root of the average value found within the period under consideration. As might be expected, it can be seen from Fig. 1 that there is a direct relationship between the two magnitudes in conformity with the fact that, due to subsequent heat emission through radiation, strong heating of the soil is associated with important temperature inversions and, therefore, with high radon values.

As a final point, it is worth pointing out that, for this period, it is possible to apply the condition of a radioactive equilibrium between radon and its daughter products with only a slight margin of error. The values are therefore representative of the concentrations of radon and Ra B present in the atmosphere, thus indicating that in these conditions of stability it is possible to make continuous measurements of the Ra C peak throughout the day in order to

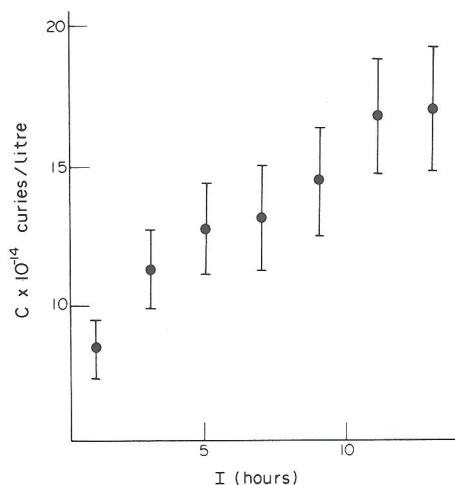


Fig. 1. Variation in the concentration of Ra C with the sunshine for stability type F.

establish its daily variation and, on the basis of this, carry out a study of the daily variations in the vertical stability (Soto, 1978; Staley, 1966).

### CONCLUSIONS

Over a sufficiently ample period of time, a series of measurements were carried out of the concentrations of the elements Ra B and Ra C short-life daughter products or radon, present in the air near a land-sea discontinuity using a gamma spectrometry chain. A definite relationship was found between the values of Ra C concentrations in the air and the stability existing at the time of taking samples, as a result of which we classified the measurements results according to the corresponding types of stability. This method of analysis possesses an advantage over the traditional studies of seasonal variations in the concentrations of radon and its daughter products inasmuch as there is a guarantee that the values classified present similar meteorological characteristics and thus the possible influence of the different meteorological variables may be more clearly shown, the values found being additionally significant for each of the normally occurring atmospheric situations.

### REFERENCES

- Biro, A., Adroguer, B. and Fontan, J. (1970) *J. Geophys. Res.* **75**, 12.  
Bouville, A., Leroy, B., Guedalia, D., Delos, Y. and Fontan, J. (1971) *J. Aerosol Sci.* **2**, 381-87.  
Dueñas, C. (1976) Tesis Doctoral. Universidad de Valladolid.  
Jacobi, W. and Andre, K. (1961). *J. Geophys. Res.* **18**, 3799-3814.  
Malakhov, S. G. and Solodikhina, A. (1962) Edited by I. L. Karol. Malakhov, Moscow, AEC 6128-TID 4500.  
Pasquill, F. (1968) Van Nostrand, London.  
Soto, J. (1978) Tesis Doctoral. Universidad de Santander.  
Staley, D. (1966) *J. Geophys. Res.* **71**, 14.  
Stranden E. (1979) *Phys. Med. Biol.* **24**, 913-20.  
Subba Ramu, M. C. and Vohra, K. G. (1969) *Tellus* **XXI**, 3.  
Wilkenning, M. (1976) *Hlth Phys.* **31**, 139-45.