A new method of continuous measurements of radon (Rn^{222}) and thoron (Rn^{220}) in the atmosphere

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ABSTRACT

To achieve continuous measurements, an aspiration process was developed, permitting a direct measurement of the radon (Rn^{222}) and thoron (Rn^{220}) contributions to the ionization. After passing through a filter that holds back all aerosols and radon and thoron daughter ions, the air streams through an approximately 300 liter ionization chamber. The current, measured with a vibrating reed electrometer, is composed of three parts: background, radon contribution and thoron contribution, with each component resolved by appropriate methods. The sensitivity limit amounts to some $0{,}8 \cdot 10^{-14}$ c/l for radon and $0{,}5 \cdot 10^{-14}$ c/l for thoron with a resolution time of about 220 sec. This report deals with the principle of measurement and its use for the determination of the radon and thoron content of the lower atmosphere.

1. Introduction

The natural radioactive gases radon (Rn^{222}) and thoron (Rn^{220}) are found concentrated in the lower atmosphere to an amount of about 10^{-16} curie per cm^3 (see, e.g., H. ISRAËL, 1962). To measure such small concentrations, generally methods of enrichment were used: The content of radon and its daughters of the atmosphere usually is measured after being concentrated by adsorption (radon) or filtering (daughters) from a fixed volume of air. In the thoron series only the long-lived daughters ThB (Pb^{212}) and ThC (Bi^{212}) can be measured on this way, whilst the thoron in view of its short mean life time (78 sec) cannot be collected in the same manner.

Due to this difficulty the thoron content of the atmosphere generally has been computed from the content of ThB found in the atmosphere, although the supposition of equilibrium between Tn and ThB is not allowed (J. FONTAN *et al.*, 1961; H. ISRAËL, 1962, 1964*b*; G. W. ISRAËL 1965*a*, and others). Direct measurements of the thoron content of the atmosphere were impossible until a short time ago. First J. FONTAN *et al.* (1961, 1962*a*, 1962*b*) developed a method to measure the thoron of the atmosphere directly, using an indirect method of enrichment, i.e. by collecting the ThB set free by the decay of thoron in a known volume of air.

Since the concentrations of radon and thoron in the atmosphere depend on the meteorological conditions, they can be used as tracers for atmospheric motions, e.g. for exchange studies. For this purpose methods of integration, as mentioned above, are unfit. Therefore we have to look for a measuring method allowing the recording of the radon content as well as the thoron content of the atmosphere continuously with a high time resolution. To realize this, an aspirating ionization chamber should be used (G. W. ISRAËL, 1964).

2. Principle

The principle of measurement we used is the following one: Atmospheric air, after being cleaned from aerosols and decay products, is aspirated through an ionization chamber. The current created in the chamber is due to three ionizing components, (1) the thoron decay, (2) the radon decay and (3) the background radiation, respectively. The three components have to be separated: Removing the thoron from the air before entering the chamber, the ionization is reduced to the radon decay and the

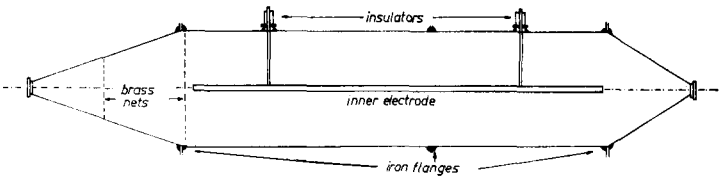

FIG. 1. The ionization chamber.

background effect. Keeping out the radon, too, the background radiation can be measured alone.

3. Sensitivity

The lowest concentration of radon or thoron in the air measurable in this way depends on the sensitivity of the equipment as well as on the statistical fluctuations of the ionizing processes.

First the ionization chamber has to have a minimum size. This size may be estimated, e.g. for thoron, by the following calculation.

Assuming a thoron concentration of c atoms per liter of air and a residence time t_r of the air in the chamber, there will occur z disintegrations of Tn atoms per sec given by the following equation:

$$z = \frac{c \cdot V}{t_r} \cdot (1 - e^{-\lambda \cdot t_r}). \qquad (1)$$

where V is the volume of the chamber in liters, and λ the decay constant of thoron. Equation (1) shows z growing nearly proportionally with V and $1/t_r$.

Letting q be the number of ions produced by one thoron decay and I the desired ionization current, V is given by equation (2):

$$V3 = \frac{I \cdot t_r}{q \cdot c} \cdot (1 - e^{-\lambda \cdot t_r})^{-1}. \qquad (2)$$

Now it may be required that a thoron concentration of 10^{-13} c/l (corresponding to 0.28 atoms of thoron per liter) should be measurable with an accuracy of ± 10 %; assuming that the current in the chamber created by 10^{-14} c/l should correspond to the sensitivity limit of a vibrating-reed-electrometer, i.e. about $3 \cdot 10^{-15}$ amp, we evaluate for a convenient residence time of 20 sec a chamber volume of *160 liters*.

This is the minimum size neglecting losses of the ionization by initial recombination and wall effects. Actually the size of the chamber was chosen about twice this value.

Besides this estimation, the real limit for the measurements will be settled by statistical conditions. First, an ionization chamber of this volume will show a considerable background noise. Indeed we found that these background disintegrations cause a current being manyfold of the effect to be expected from the decay of radon and thoron. But this background effect was found remarkably constant, so that it could be compensated nearly completely.

It is evident that the accuracy of the thoron measurements is limited by the statistical fluctuations of the background noise plus the radon decay, whilst the accuracy of the radon measurements depends only on the background fluctuations.

4. Equipment

Fig. 1 shows the ionization chamber. The chamber is a 324 liter volume copper cylinder condensor. The outer electrode is attached to a voltage of 240 volt, the inner electrode is earthed over a high-meg-resistor *Ra* of 10^{12} ohm (see Figs. 3 and 5) and connected to the input of a vibrating-reed-electrometer, *A*. For smoothing, a condensor *Ca* of about 170 pf (see also Figs. 3 and 5) effecting a time constant of 220 sec was chosen. The insulators (made of "Plexiglas") are doubled in the usual manner with an earthed inset (not to be seen in the figure).

The air stream (11.7 liter/sec) enters the chamber after having passed through an absolute filter (type "Dräger CH-630/2"; indicated by the serpentine line in the air entrance; see Figs. 3 and 5) to be cleaned from aerosols and decay products. Then, in the conical entrance, it passes two nets, to homogenize the stream

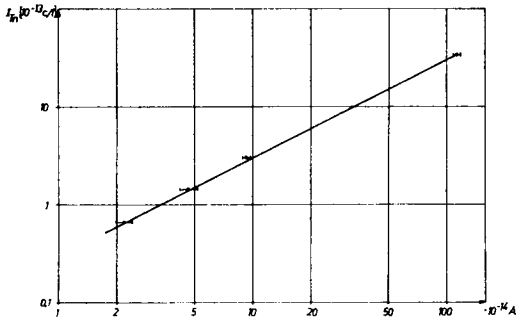

FIG. 2. Calibration.

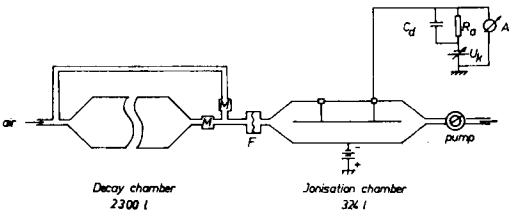

FIG. 3. Arrangement for separation of the radon and thoron caused currents.

velocity over the cross section of the chamber.

For calibration, the chamber was tested with air activated with thoron by a thorium-228 solution. Fig. 2 shows the result. The sensitivity was found at $3 \cdot 10^{-14}$ amp for a thoron content of 10^{-13} c/l. For radon measurements the sensitivity was found at $4.6 \cdot 10^{-14}$ amp for a radon content of 10^{-13} c/l.

Fig. 3 shows the arrangement to separate the radon from thoron caused ionization current. To remove the thoron, the air stream has to pass a 2300 liter decay chamber before entering the ionization chamber. Whilst flowing through this decay chamber, the thoron will disintegrate to about 90 %. Alternating ionization measurements with air streaming through this pre-chamber and with air entering the ionization chamber directly gives a course of the current as shown in Fig. 4.

The dashed horizontal line at the basis marks the background current. The differences between this line and the lower peaks of the curve show the radon effect, whilst the differences between the lower and the upper peaks are caused by the thoron ionization.

The thoron concentration was varying during the recorded time between 2.2 and $6.2 \cdot 10^{-13}$ c/l, the radon content between 0.39 and $0.96 \cdot 10^{-13}$ c/l.

For the sake of completeness, Fig. 5 gives an example of a record of the background ionization after having filled the chamber with air free from thoron and radon, showing that its variations are very small. They cause an uncertainty of about $\pm 10^{-15}$ amp. For the thoron measurements, this limit of accuracy is enlarged by the statistical fluctuations of the radon caused ionization.

Overall we found a limit of accuracy of about $\pm 8 \cdot 10^{-15}$ curie per liter for the radon measurements and a limit of about $\pm 5 \cdot 10^{-15}$ curie per liter for the thoron measurements. Thus both the radon and the thoron content of the atmosphere were measurable with a sufficient accuracy.

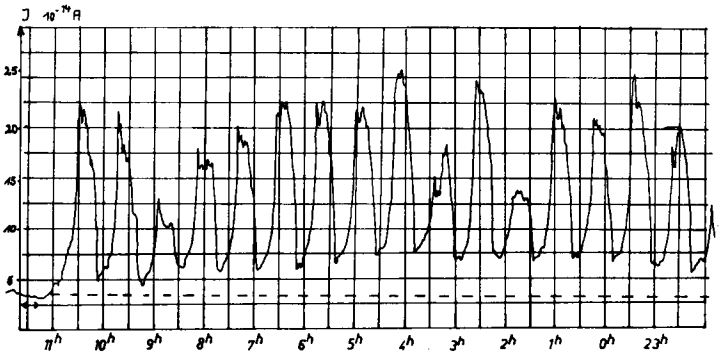

FIG. 4. Registration of Rn and Tn at Aix-la-Chapelle, 10th of June, 1964.

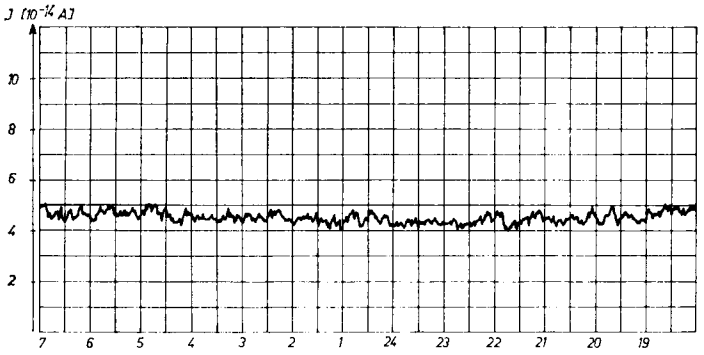

FIG. 5. Registration of the background ionization at Aix-la-Chapelle, 13th of March, 1964.

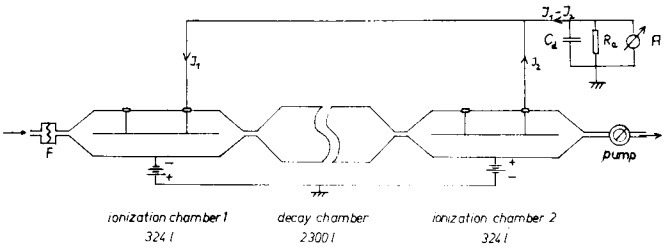

FIG. 6. Arrangement for thoron recordings.

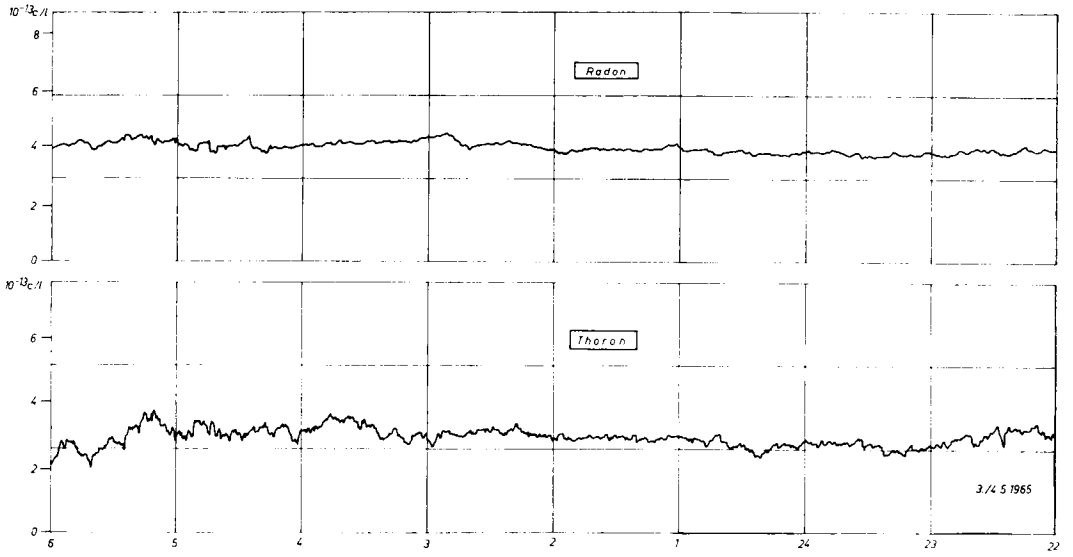

FIG. 7. Simultaneous recording of both the radon and the thoron content of atmospheric air.

The results we obtained during registrations of some months at different places will be discussed in the papers of G. W. Israël and of H. Israël, G. W. Israël and M. Horbert within this issue (see also G. W. ISRAËL, 1965*a*, 1965*b*, and H. ISRAËL & G. W. ISRAËL, 1965).

Fig. 6 shows an arrangement to record the thoron concentration continuously, using two

identical ionization chambers separated by the decay chamber. When flowing through this equipment, an air stream containing radon plus thoron will create in the first chamber an ionization current J_1 caused by radon plus thoron, whilst in the second chamber the current J_2 is caused by radon only. Thus attaching to the two chambers identical voltages of opposite sign the radon caused part J_1 can be compensated by J_2. The difference $J_1 - J_2$ gives a direct measure of the thoron concentration.

The equipment can be completed by inserting a third ionization chamber between chamber 2 and the pump to measure the radon caused current, alone. Doing so, the concentrations of both thoron and radon as well as of their variations with time can be recorded simultaneously in the same air stream.

Fig. 7 shows an example of such a simultaneous registration. The mean values of radon and thoron evaluated from this record were $4 \cdot 10^{-13}$ c/l radon and $3 \cdot 10^{-13}$ c/l thoron.

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НОВЫЙ МЕТОД НЕПРЕРЫВНЫХ ИЗМЕРЕНИЙ РАДОНА (Rn^{222} И Rn^{220}) В АТМОСФЕРЕ

Для получения непрерывных измерений был разработан процесс всасывания, позволяющий непосредственно измерять вклад в ионизацию радона (Rn^{220} и Rn^{222}). Сначала воздух проходит через фильтр задерживающий все аэрозоли и ионы продуктов распада радона, а затем течет через ионизационную камеру объемом в 300 л.

Измеряемый ток соответствующими методами разделяется на три части: основную, вклад Rn^{220} и Rn^{222}. Чувствительность равняется $0{,}8–10^{14}$ c/l для Rn^{222} и $0{,}5–10^{14}$ c/l для Rn^{220}. Этот доклад касается принципа измерения и его использования для определения содержания Rn^{220} и Rn^{222} в атмосфере.