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A new method to measure Radon and Thoron in flowing gases and its use to determine the Thoron-content of atmospheric air

by

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Introduction

The measurement of Rn(Rn^{222}) and Tn(Rn^{220}) in air is of interest mainly for three purposes :

a) Distribution studies in the atmosphere
b) Control of the inhalation hazard of workers in the uranium- und thorium-mining and milling industry
c) Determination of the Ra^{226}- or Th^{232}-body burden

The conventional methods to measure low concentrations of Radon and Thoron in gases, which use compensated ionization chambers, α-cavity-scintillators or the adsorption on activated charcoal, are rather difficult in application and need much time. Except the method developed by HURSH and LOVAAS [1,2] they allow no continuous measurement of low Rn-concentrations and only a rather qualitative determination of low Tn-concentrations in presence of Rn, as it is the case in atmospheric air. The conventional indirect single-filter-method is only applicable, if the ratio of Rn, Tn respectively, to its decay products in the air is known; but under most conditions this is not the case.

Therefore a simple method will be useful, which allows a continuous control of low Rn-concentrations in flowing gases and quantitative determination of low Tn-concentrations in presence of Rn. Such a method was developed by BLANC et al. [3] and JACOBI [4]. In the following the principle and some of our applications of the 'double-filter-method' are described.

Principle of the method

The method is based on a simple double-filter principle, which is schematically shown in figure 1. The Rn- or Tn-containing gas streams at first through a primary fibrous filter of high efficiency, which retains the Rn- und Tn-decay products primarily existing in the inlet gas, whereas Rn und Tn pass the filter undisturbed. Then this gas enters a chamber where new RaA(Po^{218})- or ThA(Po^{216})- and ThB(Pb^{212})-atoms are formed from Rn- or Tn-atoms which decay in the chamber. Their concentration increases with increasing residence time of the gas in the chamber. These fresh (secondary) decay products are filtered out by a secondary filter at the air outlet of the chamber.

The enrichment of the secondary decay products on the outlet filter follows the same laws as in the case of the one-filter-method, which is used to measure Rn- und Tn-decay products in air. The activity of Rn- or Tn-decay products on this filter is direct proportional to the Rn- or Tn-concentration in the gas entering the chamber, because it is produced from the Rn- or Tn-atoms decaying in the definite colume of the chamber between the inlet and the outlet filters. The determination of the Rn- or Tn-concentration in the gas flow is therefore reduced to a simple measurement of their decay products on the outlet filter which can be done by conventional counting methods and analysis of the filter decay-curve. By measurment of the outlet-filter-activity during sampling, the method allows a continuous control of the Rn-concentration in the gas-flow.

The sensitivity of the method depends on the residence time $\vartheta = V/v$ of the gas in the decay chamber (V = chamber volume, v = gas-flow rate) and the chamber geometry, because some of the secondary decay products are deposited on the walls of the chamber before reaching the outlet filter.

To study the effect of wall deposition, in preliminary investigations narrow tubes of different length and diameter were used as decay chambers. Fig. 2 shows the measured increase of the RaA/Rn-ratio in the flowing air with ~~pipe~~ tube length L

at different flow-rates. As expected, this ratio increases according to an exponential law of the form :

$$(a_{RaA}/a_{Rn})_L = (a_{RaA}/a_{Rn})_\infty \cdot [1 - \exp(-\text{const} \cdot L)]$$

The saturation value is limited by the deposition to the walls of the tube. This value increases with increasing diameter and decreasing flow-rate. From these experiments follows, that rather large chambers are necessary to approach radioactive equilibrium between Rn and RaA in the flowing air in the chamber. A device of this type which was used to determine the Tn-content of atmospheric air, is described later.

Instead of a fibrous secondary filter an electrostatic precipitator in the decay chamber behind the inlet filter can be used. The principle of this method is shown in figure 3. This principle makes use of the fact that after the α-decay of Rn or Tn most of the formed RaA- or ThB-atoms are positive charged ions at the beginning of their life-time. By the electric field in the chamber these ions are precipitated on the negative electrode. Therefore, the activity of this collector is proportional to the Rn- or Tn-concentration in the chamber and the effective volume of the precipitation chamber.

In contrary to the first described method, the sensitivity of this device is independent of the gas-flow-rate as long as the residence time of the air in the chamber is small compared to the half-life-time of Rn or Tn and the air velocity is small compared to the velocity of the ions in the electric field. A device of this type which was used for the continuous control of low Rn-concentrations in air, is described in the following.

Device for the continuous control of Rn in air

The cross section of the device is shown in figure 4. Having passed two fibrous filters, the air streams into a cylindrical precipitation chamber of 140 l volume, made of polyvinylchloride. At the outlet of the chamber, the air is suck off by a pump through holes and a manifold.

The inside of the front wall and the cylindrical surface of the chamber are covered with a conducting film and form the positive electrode. The negative collector electrode on which the positively charged ions of the decay products are deposited, is placed at the center of the conical back wall of the chamber. This collector consists of a Al-covered circular polyester foil of 40 mm diameter and 0,9 - 1 mg/cm^2 thickness. Behind this foil a ZnS-scintillator of 45 mm diameter is attached for measuring the α-activity of RaA and RaC' on the collector foil. Collector foil, scintillator, multiplier and cathode-follower are mounted in a tube which can be shifted on the chamber axis. The output pulses feed an integrating ratemeter which allows to record either the total counts or the counting rate on the connected recorder.

The Rn-sensitivity of the device, which is defined as the ratio of the α-counting rate at equilibrium to the specific Rn-activity in the inlet air, was checked by emanating $RaCl_2$-solutions of known Ra^{226}-activity and by a calibrated device of compensated ionization chambers. The sensitivity is independent of the air-flow-rate, as was proved in the range between 0,15 - 1,5 m^3/h, which corresponds to an air exchange rate in the chamber of about 1 - 10 h^{-1}. The sensitivity varies typically with the precipitation voltage U, as shown in figure 5 for different positions of the collector electrode in the chamber. The insertion length L is the distance of the collector from the manifold at the end of the chamber. Figure 5 shows that with increasing voltage the sensitivity rises rapidly until a maximum is reached in the range of 1,0 - 1,5 kV and then decreases slowly. Above about U = 5 kV, the sensitivity is rather constant. The maximum Rn-sensitivity with U = 1,25 kV and L = 7,5 cm is 2,2 cpm/10^{-10} cm^{-3}. The background counting rate is 0,2 - 0,3 cpm.

The mean specific Rn-activity in atmospheric air near ground is about 10^{-10} c/m^3. For this concentration the device gives 130 counts in a period of 1 h with a mean statistical error of $\pm$ 10 %. As the device is insensitive to variations of humidity or the aerosol content in the inlet air, it is therefore well suited for the continuous control of the Rn-concentration in atmospheric air. A simultaneous measurement of the Rn-decay products on the primary filter of the device allows a continuous determination of the RaB/Rn-ratio, RaC/Rn-ratio, respectively, in air.

The time resolution of the device is mainly determined by the buildup of RaC' on the negative collector, similar to the conditions when using the conventional filter method with a fixed filter. The equilibrium is reached after about 3 hours. To improve the time resolution, it is necessary to discriminate the RaC'-α-particles and to count only the α-particles of RaA. For this purpose the collector foil and the scintillation detector were replaced by a Si-semiconductor-detector with an effective diameter of 15,2 mm (ca. 180 mm^2 area). In this case the radioactive ions were directly precipitated on the Al-covered surface of the Si-semiconductor.

The resulting α-spectrum after different sampling intervals is presented in figure 6, to show the buildup of the RaA- and RaC'-peaks during sampling. The equilibrium for RaA is reached after 20 min, for RaC' after about 3 hours. Although both peaks are smeared to lower energies by absorption effects, a discrimination between RaA and RaC' is possible. In equilibrium the area of the RaC'-peak is about a factor 4 greater than the area of the RaA-peak. On account of this fact, one must presume that not only some of the RaA-atoms formed in the air of the chamber are deposited on the collector but as well a percentage of positive RaB-ions which results from the α-decay of the rest-RaA in the chamber air. It follows that a cutoff of the RaC'-peak would improve the time resolution of the device by a factor of about 10, but would reduce the Rn-sensitivity by a factor of about 0,2. Yet it must be emphasized that the

described precipitation chamber and detector devices do not have the optimum geometry; certainly, it will be possible to improve the Rn-sensitivity.

Measurement of Thoron in atmospheric air

The principle of the device for measurement of low Tn-concentrations in air was already explained in fig. 1. The device uses a cylindrical chamber between the primary and secondary filter with a diameter of 25 cm and a volume of 50 l. The air flow-rate was normally 2,5 m^3/h ($\pm$ 10 %), which corresponds to a mean residence time of the air in the chamber of about 1,2 minutes; during this time about 60 % of the Tn-atoms which pass through the first filter will decay in the chamber. According to the diffusion loss to the walls only a definite percentage of the freshly built-up ThA and ThB-atoms reach the secondary filter, where they are enriched.

After a definite sampling time the primary and secondary filter are removed and their α-activity measured in a proportional counter with approximate 2π-geometry. From the decay-curves both components, the Rn-decay products and the Tn-decay-products, can be analysed on each of the filter. The activity of the primary filter is proportional to the activity of the decay products in the inlet air, whereas the activity on the secondary filter is proportional to the concentration of Rn, Tn respectively, in the inlet air.

The Rn- and Tn-sensitivity of the device were determined by emanating $RaCl_2$- and $Th(NO_3)_4$-solutions of known concentration into the chamber through the inlet filter. It resulted a Rn-sensitivity of

$$\frac{Z_{RaA + RaC'}}{a_{Rn}} = 1,96 \pm 0,08 \quad \frac{cpm}{10^{-10}\ c/m^3}$$

where $Z_{RaA + RaC'}$ is the α-counting rate from Rn-decay products on the secondary filter 5 minutes after the end of a 3 h-sampling interval, when equilibrium on the filter is reached.

The Tn-sensitivity was

$$\frac{Z_{ThC + ThC'}}{a_{Th}} = 0,70 \pm 0,05 \ \frac{cpm}{10^{-10} \ c/m^3} ,$$

where $Z_{ThC + ThC'}$ is the α-counting rate from Tn-decay products on the secondary filter at the end of a 48 h-sampling interval; after this time 96 % of the equilibrium-activity on the filter is reached. From these values the sensitivity for smaller sampling intervals can be determined. As the background of the counter is 0,3 cpm, a mean Tn-concentration of 10^{-10} c/m^3 = 100 pc/m^3 in air during a 24 h-sampling interval can be measured with a mean statistical error of ± 10 %.

This double filter method has compared with the conventional methods the advantage, that it allows in a simple way a quantitative discrimination between Rn and Tn and therefore for the first time a quantitative measurement of the Tn-content in atmospheric air.

With this device altogether 51 Tn measurements were made over a plain, grass-grown surface. In a first group of measurements the air was suck off in 0,2 m altitude, in the second group in 2 m altitude. Simultaneously, the ThB-content was determined from the ThC + ThC' -activity on the inlet filter of the device. Furthermore, the RaB-concentration in 2 m altitude, and - as indicator for the atmospheric stability - the wind velocity in 9 m and the temperature in 0,05 m, 0,5 m, 2 m and 9 m altitude were continously measured during the sampling intervals, which ranged on each day from 8.00 - 17.00 h and from 17.00 - 8.00 over night. The resulting mean values are compiled in table1.

The result is a mean-Tn-content of nearly 500 pc/m^3 in 0,2 m altitude and of 160 pc/m^3 in 2 m altitude. The specific Tn-activity exceeds the

Table 1 : Measured mean specific activity of Tn, ThB and RaB in atmospheric air and meteorological parameters during the sampling periods

Sampling interval *)	Spec. activity (pc/m3) Tn	ThB	RaB	Activity ratios ThB/Tn	Tn/RaB	ThB/RaB	Meteorol. parameters **) u (m/s)	grad ϑ (°C/m)	I (mm)
a) Tn, ThB in 0,2 m, RaB in 2 m altitude (3.-17. Sept. 1963)									
8.00-17.00 (10)	350	0,8	110	0,003	(3,2)	(0,007)	3,2	- 0,6	0,6
17.00-8.00 (8)	500	4,5	210	0,010	(2,4)	(0,021)	0,6	+ 1,0	0
daily mean value	450	3,1	170	0,007	(2,7)	(0,017)	1,6	+ 0,4	0,6 (total)
b) Tn, ThB and RaB in 2 m altitude (12. Aug. - 2. Sept., 18.-24. Sept. 1963)									
8.00-17.00 (14)	130	1,0	70	0,008	1,9	0,014	5,1	- 0,3	15,4
17.00-8.00 (23)	180	1,9	130	0,010	1,4	0,014	3,2	+ 0,4	25,6
daily mean value	160	1,5	105	0,010	1,8	0,014	3,9	+ 0,1	41 (total)

*) in ( ) number of measurements

**) u = mean wind velocity in 9 m altitude; grad ϑ = mean temperature gradient (0-2 m); I = rainfall during the total sampling time

corresponding specific activity of RaB, Rn respectively, by a factor of 2 and of ThB by a factor of about 100. Obviously near ground level ThB is far away from radioactive equilibrium with Tn.

These results are not astonishing if one takes in account that all exhaled Tn-atoms decay in the air layer near ground because of their short half-life-time of 54,5 sec, whereas Rn, ThB and their decay products are distributed by vertical mixing over a great part of the troposphere. Recently, we published the results of theoretical computations of the vertical distribution of Rn, Tn and their decay products in the atmosphere on the basis of the mixing theory [5,6]. For these calculations a mean Tn-exhalation rate from ground of 100 Tn-atoms/m^2 sec was assumed which corresponds to the measured mean Rn-exhalation rate taking into regard the different half-life-time of both emanations. Yet it must be emphasized that the ThB/Tn-ratio is independent of the Tn-exhalation rate. In table 2 the calculated mean values of the specific Tn- and ThB-activity and the ThB/Tn-ratio in 0,2 m and 2 m altitude under normal turbulence conditions are compared with the measured daily mean values of these quantities. The agreement is quite well and confirms the results.

Table 2 : Comparation of the measured mean values of the specific Tn- and ThB-activity and the ThB/Tn-ratio in atmospheric air with mean values predicted from the mixing theory

Quantity	0,2 m altitude		2 m altitude	
	exper.	calcul.	exper.	calcul.
Tn-content (pc/m^3)	450	600	160	200
ThB-content (pc/m^3)	3,1	3,0	1,5	2,5
ThB/Tn-ratio	0,007	0,005	0,010	0,012

Conclusions

The described applications of the 'double-filter method' show that this method allows in a simple way the measurement of low Rn- or Tn-concentrations in flowing gases. Compared with the conventional methods it especially has the following advantages :

(a) Rn can be controlled continously down to concentrations below 100 pc/m^3.

(b) It allows the objection of Tn in gases in presence of Rn down to concentrations of about 10 pc/m^3 and therefore for the first time a direct quantitative measurement of the Tn-content in atmospheric air.

(c) Simultaneously with Rn or Tn their decay products can be determined by measurement of the primary filter-activity.

(d) The method joins high Rn- or Tn-sensitivity with simplicity and stability; the technical effort is low and only conventional filter counting techniques are required.

The method therefore seems to be well suited for the measurement of Rn and Tn in atmospheric air and in breath as well as for the control of the inhalation hazard in uranium- or thorium-processing industries.

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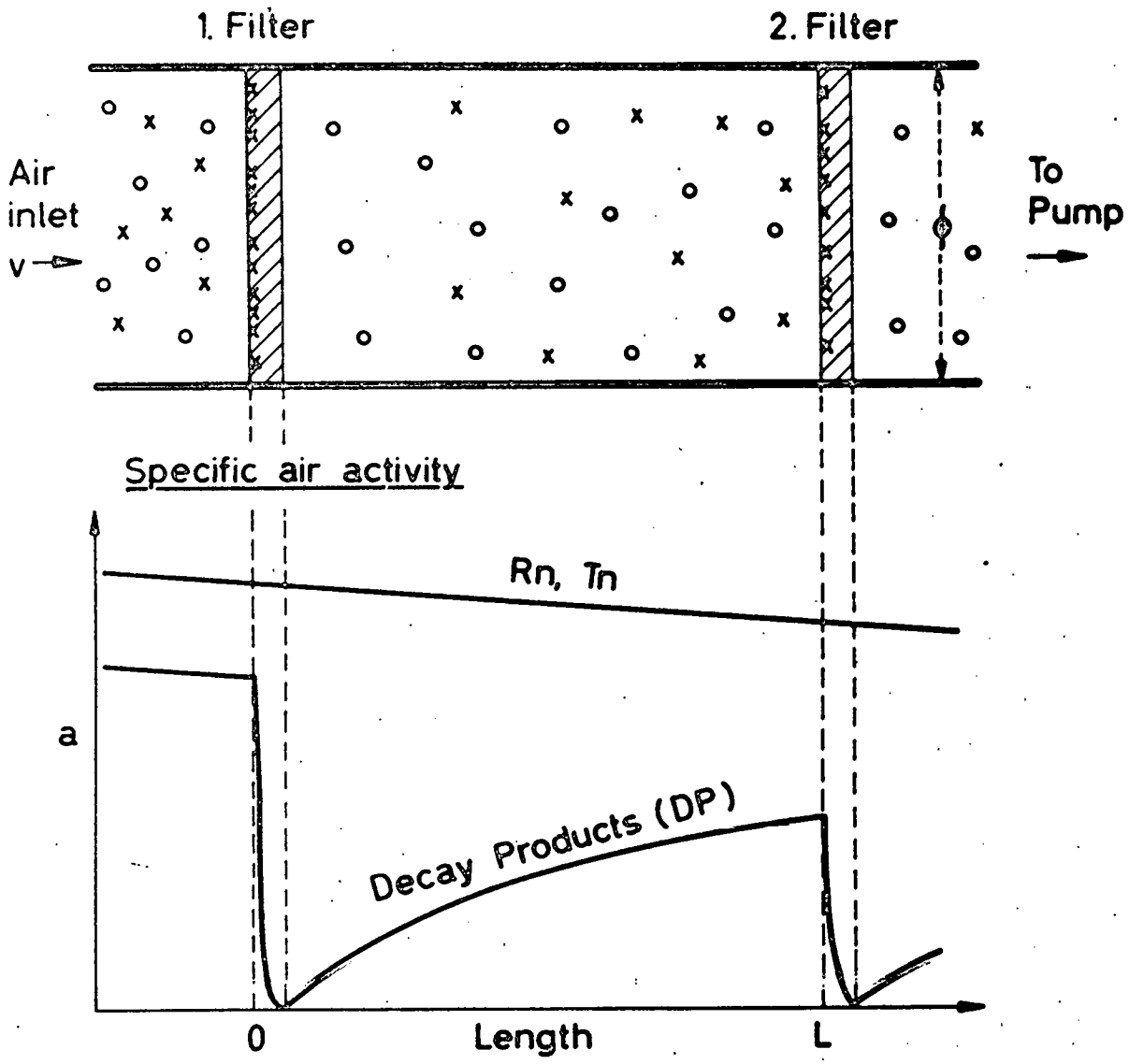


Filter activity:

1. Filter $A_1 \sim a_{DP}(0) \cdot v$

2. Filter $A_2 \sim a_{DP}(L) \cdot v \sim a_{Rn,\,Tn}(0) \cdot f(\phi, L, v)$

Fig. 1: Principle of the double filter method with a fibrous second filter

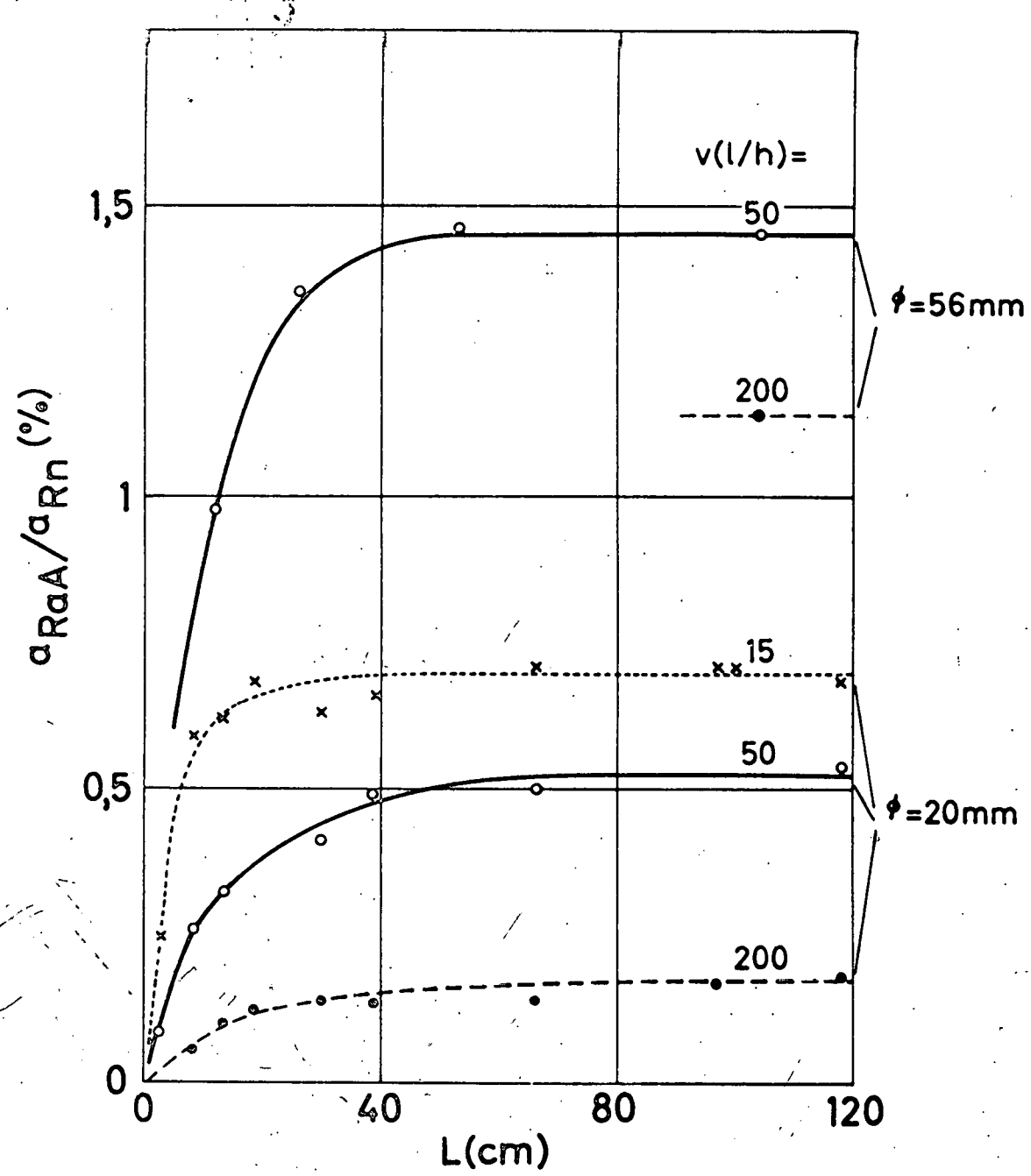
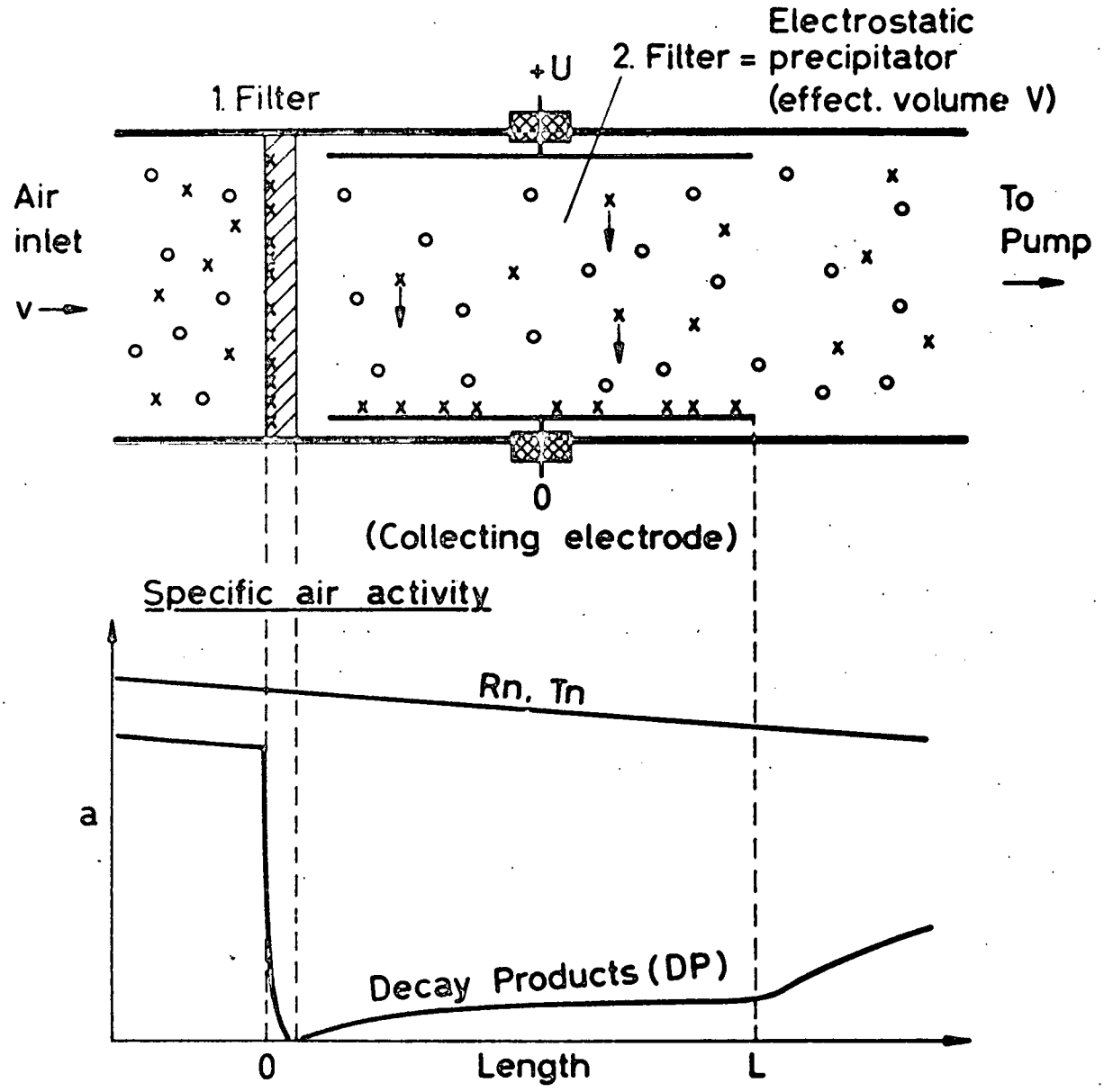


Fig. 2: Buildup of RaA in air streaming through a pipe after prefiltration at L = 0



Filter activity:

1. Filter $A_1 \sim a_{DP}(0) \cdot v$

Collecting electrode $A_2 \sim a_{Rn,Tn}(0) \cdot V \cdot \eta(U)$

Fig. 3: **Principle of the double filter method with an electrostatic precipitator as second filter**

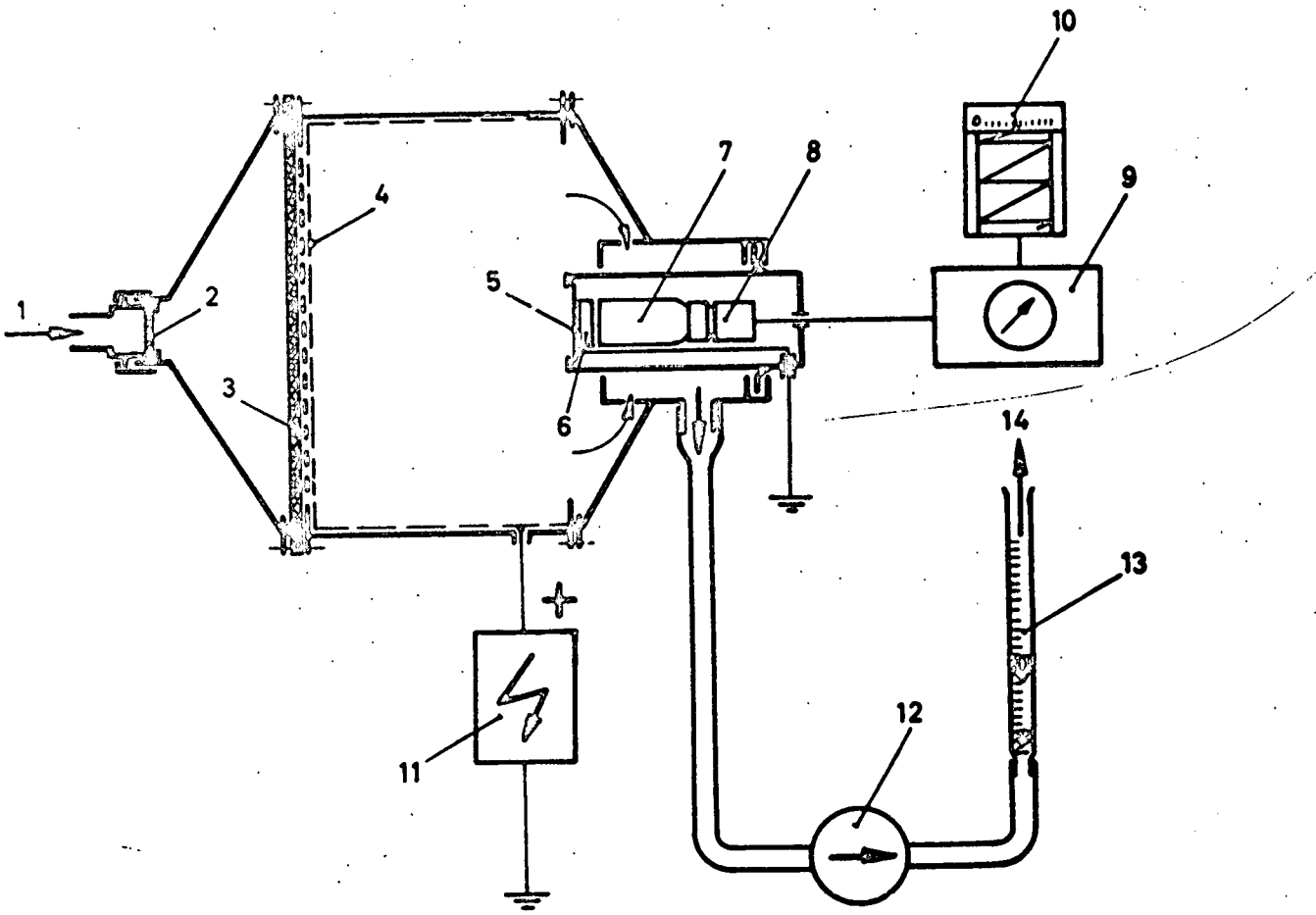


Fig. 4: Device for the continuous measurement of Rn^{222} in atmospheric air

(1 = air inlet; 2, 3 = 2 primary fibrous filters; 4 = positive electrode; 5 = negative collector electrode; 6 = ZnS-scintillator; 7 = multiplier; 8 = cathode follower; 9 = integrating ratemeter; 10 = recorder; 11 = precipitation voltage power supply; 12 = pump; 13 = air-flow-meter; 14 = air outlet)

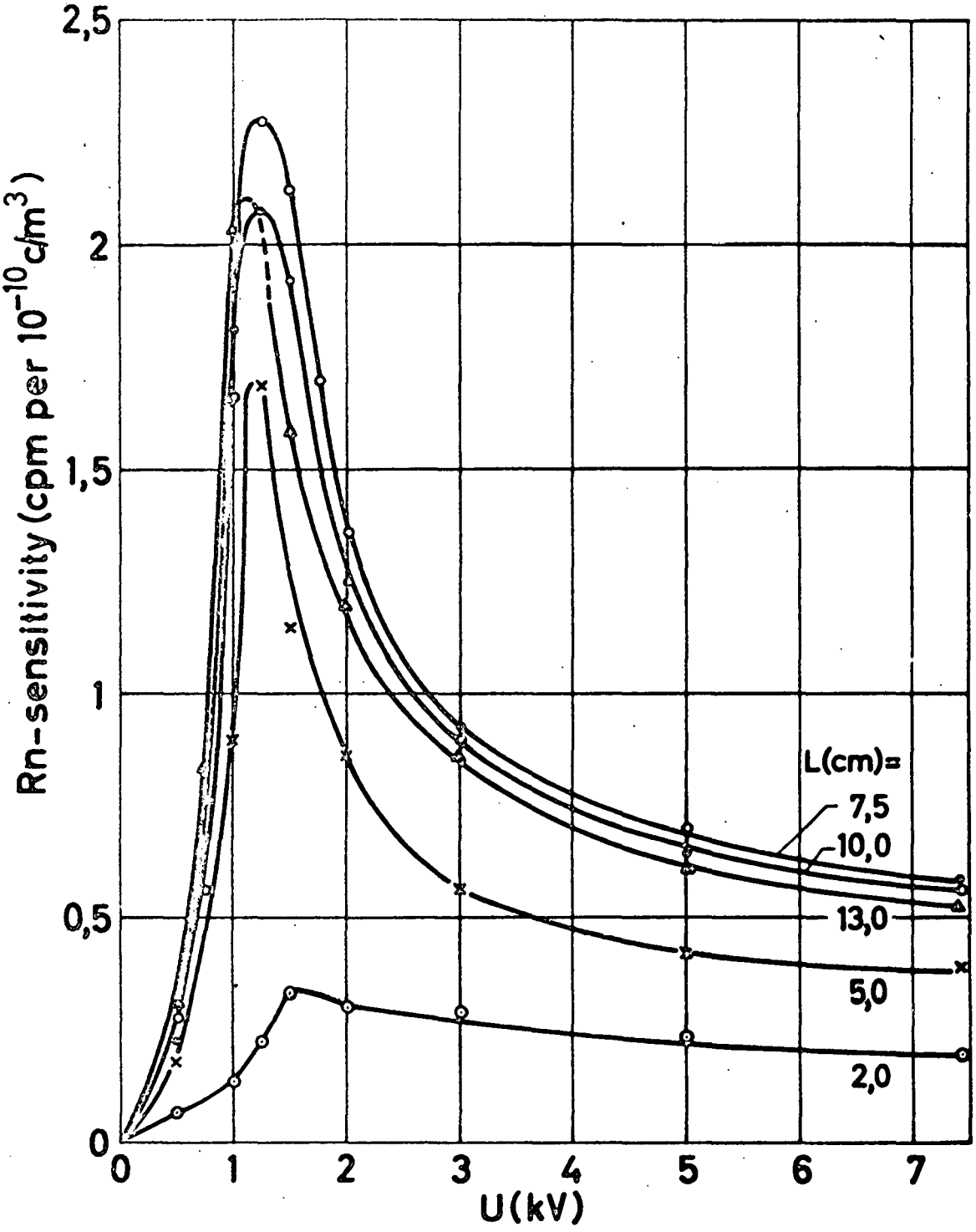


Fig. 5: Rn^{222}-sensitivity versus precipitation voltage U (Parameter: Insertion length L of the collector in the chamber)

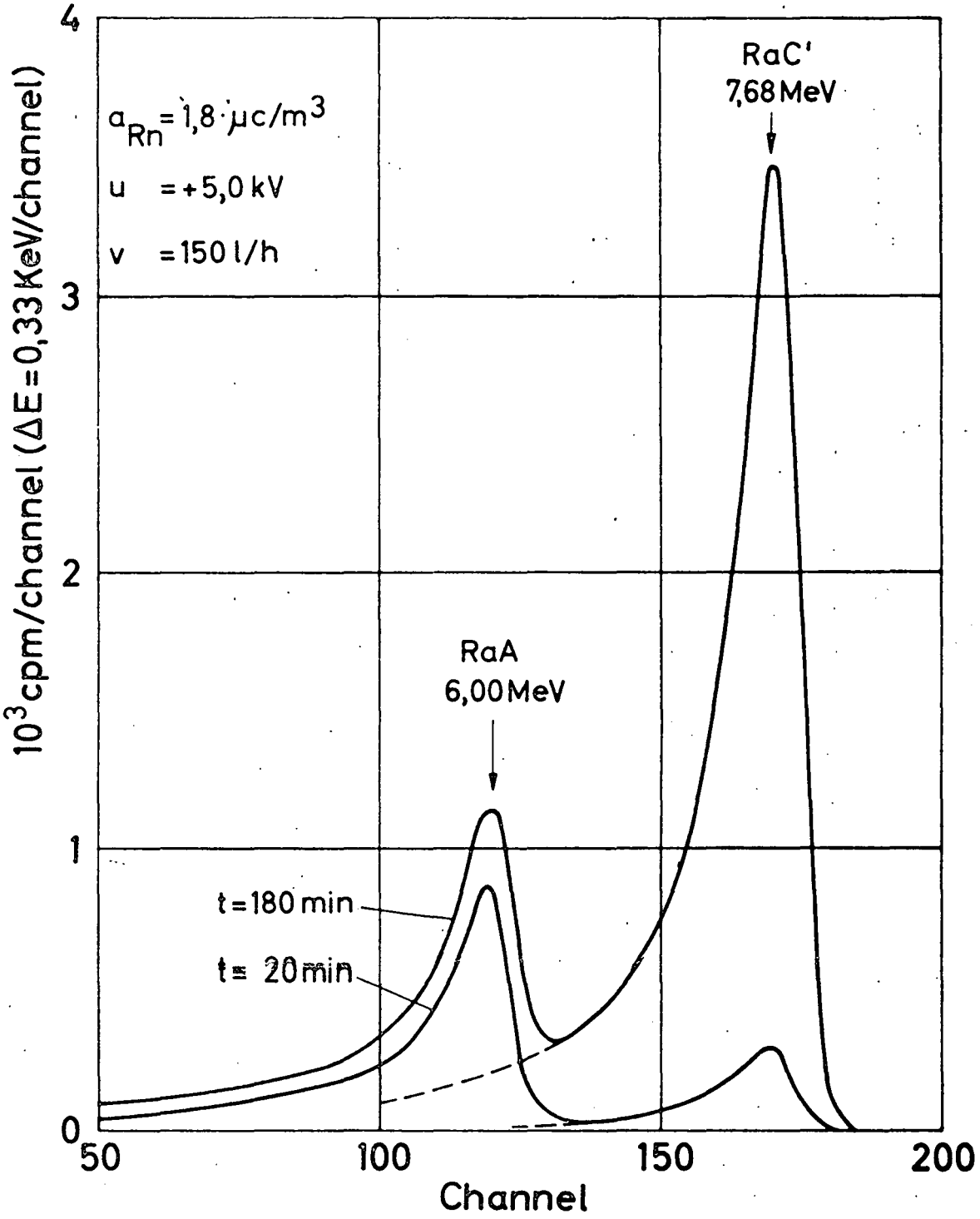


Fig. 6: α-spectrum of Rn^{222}-decay products collected on the surface of a Si-semiconductor detector