CARBON-14 IN ATMOSPHERIC CARBON DIOXIDE

MARTIN CHUDÝ, ŠTEFAN ŠÁRÓ, MOJMÍR ŠELIGA, PAVEL POVINEC
Bratislava

INTRODUCTION

Before 1950 the natural radioactive carbon dioxide in the troposphere and lower stratosphere was in the steady-state equilibrium [1]. The generation of radiocarbon by cosmic radiation in the upper layer of the atmosphere as the consequence of the reaction N^{14} (n, p) C^{14} was balanced with the loss caused by radioactive decay. This state lasted more than thirty thousand years [2]. Since the explosion of nuclear weapons are accompanied by considerable neutron flux, after their realization the radiocarbon level in the atmosphere started to increase. In 1959 Hageman et al. [3] investigated the presence of radiocarbon in the stratosphere resulting from experiments with nuclear weapons.

The mechanism of radiocarbon penetration from the higher into the lower layers effects that the values of its concentration in the air near the earth surface are geographically not comparable [4]. Concentration of radiocarbon may be influenced by industrialization [5]. The values measured in places near industrial regions are permanently lower [4]. At the beginning of nuclear tests, in 1963—1964, the level of radiocarbon concentration in the atmosphere reached in comparison with its natural level an increase of about 60 % [4, 6, 7]. Nydal and Lövseth [8] give an essentially higher value. In the following years the concentration of carbon dioxide decreased [9, 10, 11].

The measurements of radiocarbon concentration in the atmosphere were carried out in the Radioactivity Data Processing Laboratory, Comenius University. An apparatus was constructed consisting of an equipment for obtaining carbon dioxide from the air near the earth surface, a second apparatus for purification of carbon dioxide, and a third consisting of counters with electronics. The activity of atmospheric carbon dioxide was determined by measurements in the form of carbon dioxide in the internal proportional counter.

PREPARATION OF GAS FILLING

To absorb carbon dioxide from the air a solution of barium hydroxide was used. Carbon dioxide was collected by bubbling air through this absorption medium into a sampler [12]. The carbon dioxide reacts in the following way:

$$CO_2 + Ba(OH)_2 \rightarrow BaCO_3 + H_2O.$$

The efficiency of such a separation is 100 %. Carbon dioxide was decomposed by hydrochloric acid on barium carbonate in the decomposition reaction. The carbon dioxide is formed according to the equation

$$BaCO_3 + 2\,HCl \rightarrow BaCl_2 + H_2O + CO_2.$$

It is purified in the apparatus the simplified scheme of which is shown in Fig. 1.

Carbon dioxide liberated in the decomposition reactor after passing through trap 1 condenses in trap 2. The purification is based on repeating the following cycle; carbon dioxide passes from trap 2 through the furnace containing activated copper (working temperature 200 °C), purification tubes containing anhydrone and dried potash, respectively, and trap 3 into trap 4. The apparatus is evacuated after every cycle. The purpose of this purification is to release the carbon dioxide from the admixture of oxygen, eventually from the minute quantities of water vapour and the sulphur compounds.

COUNTER AND COUNTER ELECTRONICS

The counter available for the measurement of atmospheric radiocarbon activity must have a high detection efficiency and a low background. There-

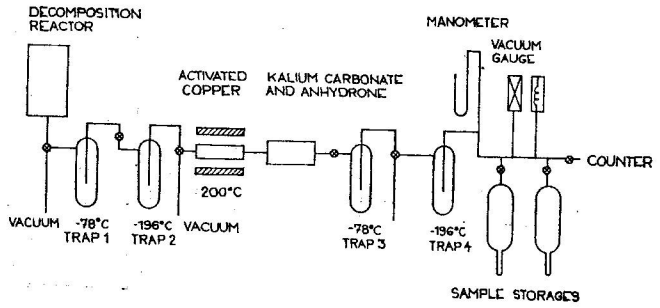

Fig. 1. Carbon dioxide purification system.

fore we use an internal proportional counter, a heavy shielding, and a cosmic radiation detector in anticoincidence.

The operating volume of the inner proportional counter is .4 l. The counter diameter is 4 cm. The cathode made of copper foil is 80 μm thick. The diameter of the molybdenum anode wire is 50 μm. The operating pressure of the carbon dioxide filling is 800 torr.

For the shielding counter connected with the inner proportional counter in anticoincidence we use a counter with 16 anode wires working in the proportional region. Its cathode is of stainless steel. The operating gas methane has the pressure of 780 torr. The counter section is shown in Fig. 2.

When measuring, the counter is placed in a shielding box of a total weight of 4000 kg, consisting of 10 cm lead, 10 cm mercury, and .5 cm iron.

The block diagram of the electronics is seen from Fig. 3. The pulses of

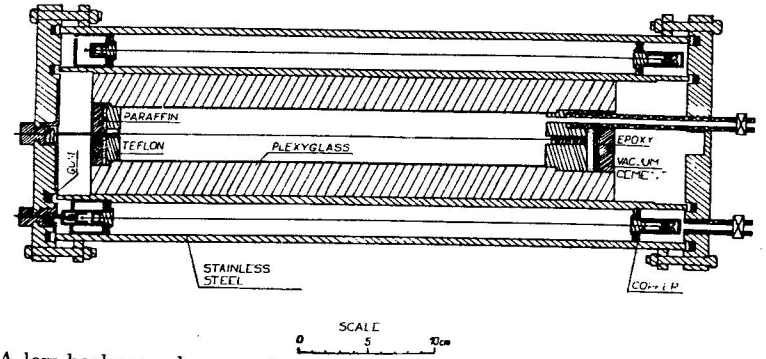

Fig. 2. A low background proportional counter with built-in anticoincidence detectors.

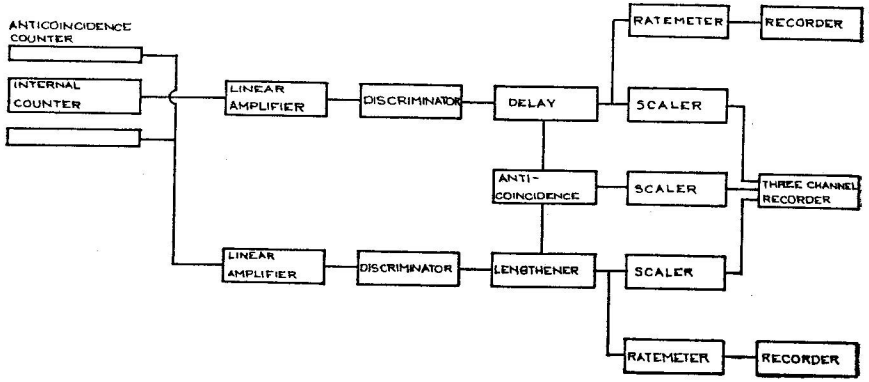

Fig. 3. Block diagram of the counting system.

the inner proportional counter pass through the cathode follower and the linear amplifier. After some delay the pulses are transformed into rectangular ones, and enter the anticoincidence circuit. Before being supplied to the anticoincidence circuit, the pulses from the anticoincidence counter are lengthened to 250 μs. The pulse counts registration is made automatically. In regular intervals the counts are registered in three channels; in the channel of the inner and the anticoincidence counter, and at the output of the anticoincidence circuit. For a permanent check we also recorded the counting rates in the channels of the inner and the anticoincidence circuit with pen recorders.

The inner proportional counter is fed through the stabilized high voltage power supply, refilled with batteries. The voltage fed to the anode is controlled by an electrostatic voltmeter.

With respect to the low channel threshold of the inner counter the spurious pulses may also contribute — through the condensator connecting the counter anode with the input of the cathode follower — to the counter background arising from the corona discharge and the punch. It seems better to use a ceramic capacitor with a higher nominal voltage than the one of the counter. In our design we use the condensator of 470 pF with the nominal voltage of 12 kV. A further source of spurious pulses may be the breakdowns captured by the counting system from some near electric apparatus. It is most efficient to eliminate the sources of the breakdowns, interference of electronic switches, relays and motors. An essential lowering of the spurious pulses has been reached by electromagnetic shielding of the input circuits by means of well-conducting materials, and by filtrating the main voltage. The 4 m long coaxial cable connecting the cathode follower with the input of the linear amplifier has a double shielding.

OPERATING CHARACTERISTICS

Admixtures of electronegative gases in carbon dioxide filling have an influence on the operating characteristics of the proportional counter. The operating characteristics of the inner counter obtained with purified carbon dioxide at a pressure of 800 torr are shown in Fig. 4. The plateau length is more than 550 V (Fig. 4) and the slope of the plateau is less than 2.5 per 100 V. Curve (a) corresponds to the counter without shielding, curve (b) to that in the shielding box, and curve (c) to the shield with anticoincidence.

The operating characteristics of the anticoincidence counter are in Fig. 5. The characteristic (d) is measured without shielding, the characteristic (e) is in the shielding box. The plateau length is 500 V at a slope of 2.5 per 100 V.

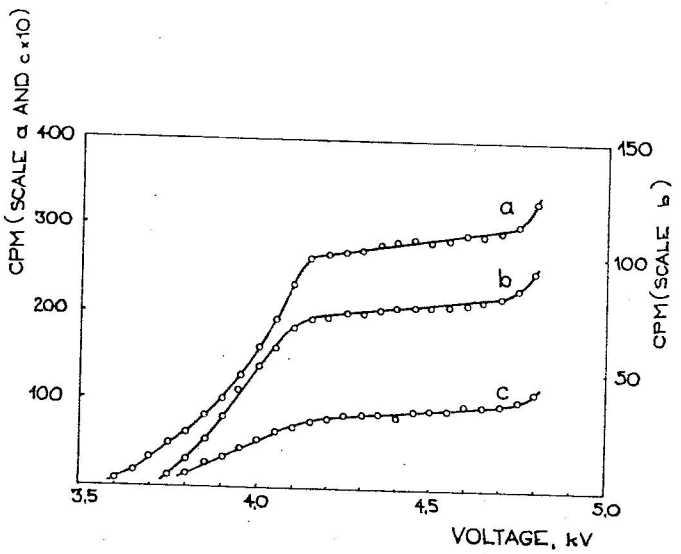

Fig. 4. Counting characteristics of the internal counter.

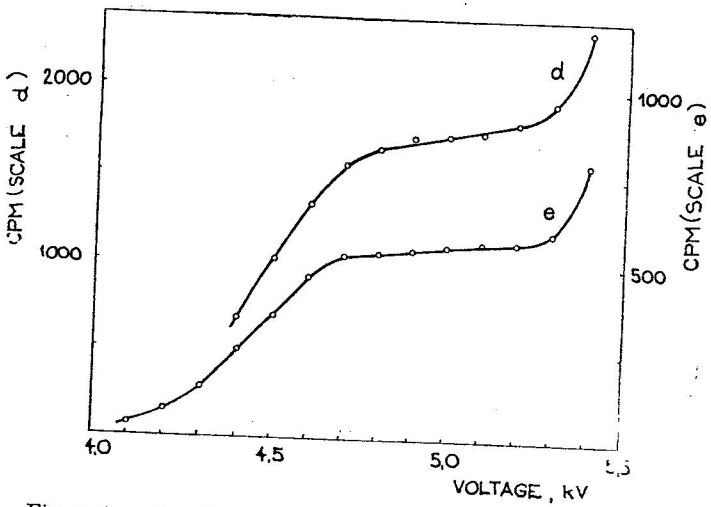

Fig. 5. Counting characteristics of the anticoincidence counter.

The input sensitivity of the electronic apparatus is in all cases 50 mV. At an operating voltage of 4.4 kV the total detection efficiency of the inner counter is 85 %. The background of the unshielded counter is about 300 pulses/min. With shielding and anticoincidence it dropped to 7—9 pulses/min. For the background determination we use carbon dioxide prepared from coal.

MEASUREMENT RESULTS

There were measured the carbon dioxide samples from the air at Trnava (48°22′ N Lat, 17°35′ E Long) 1 m over the earth surface. Tab. 1 contains the values of the activity of C^{14}, given in the disintegrations per minute of carbon-14 per gram of carbon in the atmospheric carbon dioxide. The percentual increase of the carbon-14 level above the level of the natural activity of radiocarbon is given as

$$\Delta C^{14} = \frac{N - N_0}{N_0} 100,$$

where $N_0 = 14 \pm 1$ is the carbon activity in disintegrations per min. of carbon-14 per g of carbon in the year 1950; N means the carbon activity in the sample measured (in decay per min. per g C). Counting errors shown in Tab. 1 are represented by the standard deviation.

Table 1

Dates	Carbon-14 content	
	dpm/g C	Δ C^{14} [%]
26. 6. 1967	14.9 ± .8	7
7. 9. 1967	19.3 ± .8	38
7. 10. 1967	18.2 ± .8	30
11. 10. 1967	18.6 ± .8	33
5. 12. 1967	20.3 ± .8	45

The fluctuations in the activity of C^{14}, which are higher than the calculated errors may be due to meteorological conditions. The lower value of the activity may be caused by the admixture of the inactive carbon dioxide at the sampling, the concentration of which in the air increased in consequence of Suesse's effect.*)

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Katedra jadrovej fyziky
Prírodovedeckej fakulty UK,
Bratislava