LONG-TIME ATMOSPHERIC ^3H AND ^{14}C RECORD IN CROATIA

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INTRODUCTION

The natural production of radioisotopes ^{14}C and ^3H in the atmosphere is in an equilibrium with their radioactive decay and their world-wide distribution has attained a steady state with subtle variations depending on the latitude, season and solar modulation. This steady state, however, was disturbed significantly during sixties, when the natural activities of ^{14}C and ^3H increased due to extensive nuclear bomb tests. ^{14}C and ^3H distribution in the atmosphere started to be monitored about at the same time, therefore, the peaks due to the anthropogenically produced ^{14}C and ^3H were recorded. Since then, atmospheric ^{14}C and ^3H activities are regularly measured as a part of a network (1, 2, 3) or local monitoring (4, 5).

In this paper we present the complete record of ^{14}C and ^3H data measured in Zagreb. We present an attempt of analytical representation of our data, and we compare our data with recently published ^{14}C records in Europe (1, 4) and in Canada (5), as well as with the tritium data in precipitation published by IAEA (3).

SAMPLING AND MEASUREMENTS

Measurements of atmospheric ^3H and ^{14}C activity have been performed in our laboratory since 1976 and 1978, respectively. Since then, tritium in monthly precipitation samples at Zagreb has been regularly measured, as well as precipitation at Ljubljana (Republic of Slovenia), and these results have been published in IAEA-WMO Technical Reports series (5). For shorter periods of time, tritium in monthly precipitation was measured at Plitvice National Park, SE from Zagreb, and in Rijeka, at the North Adriatic coast. Recently, we started to collect precipitation samples also on Mt. Medvednica, station Puntijarka, at ~1000 m a.s.l., nearby the city of Zagreb. We have been measuring also tritium activity in atmospheric water vapor since 1988 at the Institute and at several sampling sites in down-town Zagreb or in its surroundings, with the aim of studying possible local tritium contamination.

^{14}C in biosphere has been monitored through atmospheric CO_2, annual plants and leaves. Samples were collected at the sparsely populated area of Plitvice National Park and Mt. Medvednica, at the density populated area of Zagreb and in wider area of nuclear power plant Krško, which is located in Slovenia, ~20 km from the Croatian border. Measurements of ^{14}C activity of tree rings grown in the Plitvice National Park, at Mt. Matra (Hungary) and near Krško were used for reconstruction the past radiocarbon in the atmosphere.

Atmospheric water vapour were collected by absorption on silicagel. Samples of CO_2 were collected monthly by absorption on saturated NaOH solution. For ^{14}C measurement in tree rings cellulose samples of individual tree-rings were separated. All the samples for ^3H and ^{14}C measurements were converted to gas methane which is used as a counting gas in proportional counters.

RESULTS AND DISCUSSION

^3H in the atmosphere

^3H concentration in precipitation at three continental stations, Zagreb (1976-1995), Ljubljana (1981-1995) and Plitvice (1980-1982) closely follows each other showing seasonal changes typical for the temperature zone of the northern hemisphere with a decreasing yearly mean values. The yearly mean values in Zagreb and Ljubljana are closely correlated with those at Vienna station (3), as the closest continental station within the IAEA-WMO network. By using this

correlation we were able to reconstruct the past tritium activity in Zagreb (6). The ^3H measurements at Puntijarka station started in 1995. This station is assumed to be less affected by local contamination effects, such as fossil-fuel combustion or tritium release, than the nearby city of Zagreb. The first results show no significant difference in ^3H concentration between Zagreb and Puntijarka station. However, ^3H concentration at the coastal station Rijeka is, on the average, half of that at Zagreb, and is closely correlated to the nearest Mediterranean station Genoa.

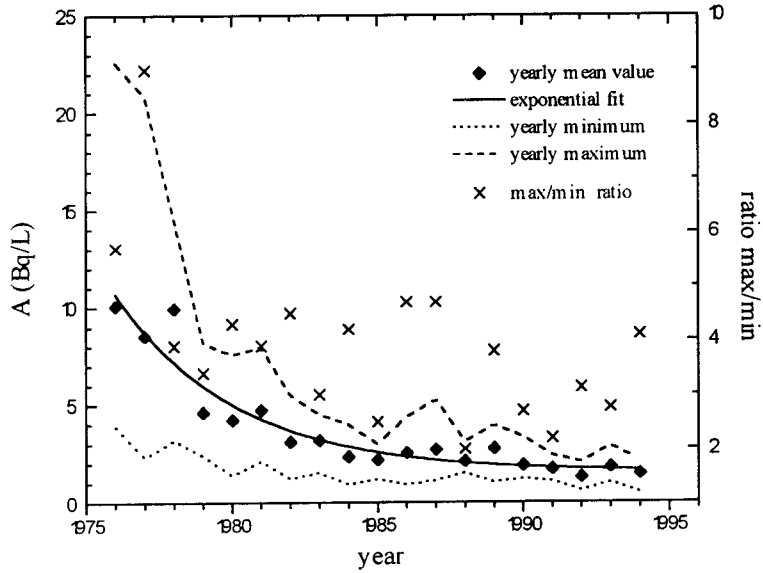

Figure 1. Statistically elaborated ^3H data in Zagreb precipitation.

The twenty year record of ^3H data in Zagreb precipitation is statistically elaborated in Fig. 1. Yearly mean values of Zagreb precipitation decrease from 10.1 Bq/L in 1976 to 0.5 Bq/L in 1994 and the decrease can be well described by the exponential decay curve (full line, Fig. 1). We show the maximal and the minimal activities in monthly precipitation during the year. Both values decrease following again approximately exponential decay. The lowest value, measured mainly in the winter time, in the last three years is 0.5 Bq/L, very close to the assumed natural pre-bomb tritium activities. The maximal value, measured mainly in summer period, are between 2 and 5 Bq/L. Similar activities were measured by Meijer *et al.* (4). We also present the behavior of the ratio of the maximal to the minimal yearly values. The values of max/min ratio fluctuate between 2 and 5 with an exemption in 1977 with a value of 9.

^3H concentration in atmospheric water vapour at Ruđer Bošković Institute was several times higher than that in precipitation from the same period. At all other investigated places the ^3H concentration in water vapour was very close to that in precipitation, so no local contamination similar to that at the Institute was observed. ^3H concentration in water vapour is much more locally affected than that in precipitation and it is a good indicator for local contamination.

^{14}C in the atmosphere

The summary of all ^{14}C environmental and atmospheric results is shown in Fig. 2. Values of atmospheric CO_2 are mean yearly values, those of plants are either single or average values of several plants, and "tree" stands for tree-rings. Curves fitted to the data of Plitvice tree,

period 1964-1986 (full line), as well as Zagreb atmospheric CO_2, period 1983-1995 (dotted line), are shown in Fig. 2. Data in Fig. 2 can be discussed in several time periods: 1) up to 1964, increase of $^{14}CO_2$ activity, 2) 1964-1969, period of fast decay of $^{14}CO_2$, app. 50-65‰ per year, 3) 1970-1979, lower decay rate, app. 20-30‰ per year, and 4) 1980-present, decay rate app. 10‰ per year. Assuming that no new sources will introduce new ^{14}C into the atmosphere and that the rate of fossil fuel combustion will remain constant, the extrapolation of the ^{14}C decay curve to the year 2000 gives $\Delta^{14}C$ value of 116‰ (fitted line, Plitvice tree) and 62‰ (fitted line, Zagreb CO_2), respectively. It means that atmospheric ^{14}C activity will approach 11% and 6%, respectively, above prebomb levels by the turn of the century. For comparison we used atmospheric $^{14}CO_2$ data from Germany (1), from the Netherlands (3) and from Canada (4). All data show the exponential decay of mean yearly ^{14}C activities, with half life of app. 8 years. Data from Canada (Fig. 2) agree very well with ^{14}C activities measured in Plitvice tree, resulting in similar extrapolated ^{14}C activity for 2000. Lower $^{14}CO_2$ activities in Zagreb indicate greater influence of fossil fuel CO_2.

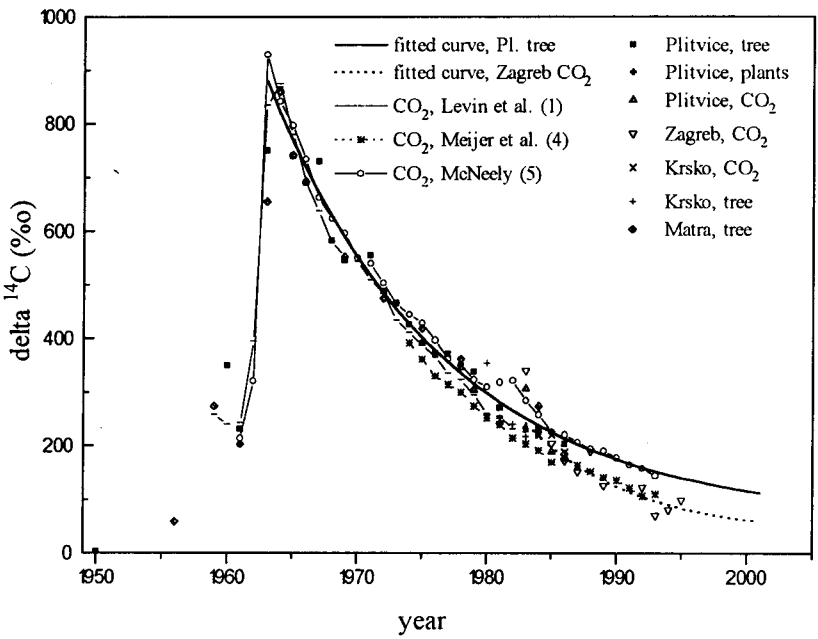

Figure 2. The ^{14}C activity of atmospheric CO_2 and tree rings. Fitted curve for Zagreb CO_2 and Plitvice tree extrapolated to 2000 and compared with the ^{14}C measurements in Germany (1), the Netherlands (4) and Canada (5).

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