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ANTHROPOGENIC RADIOCARBON: PAST, PRESENT, AND FUTURE

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ABSTRACT. ^{14}C is one of the most important anthropogenic radionuclides released to the environment by human activities. Weapon testing raised the ^{14}C concentration in the atmosphere and biosphere to +100% above the natural level. This excess of atmospheric ^{14}C at present decreases with a half-life of ca 7 years. Recently, a new source of artificially produced ^{14}C in nuclear reactors has become important. Since 1967, the Bratislava ^{14}C laboratory has been measuring ^{14}C in atmospheric $^{14}CO_2$ and in a variety of biospheric samples in densely populated areas and in areas close to nuclear power plants. We have been able to identify a heavy-water reactor and the pressurized water reactors as sources of anthropogenic ^{14}C. ^{14}C concentrations show typical seasonal variations. These data are supported by measurements of ^{3}H and ^{85}Kr in the same locations. Results of calculations of future levels of anthropogenic ^{14}C in the environment due to increasing nuclear reactor installations are presented.

INTRODUCTION

Natural ^{14}C concentration in the environment is considerably modified by human activity when measurable quantities of ^{14}C are released to the atmosphere. These anthropogenic effects have two sources of origin. First, nuclear weapon testing raised the ^{14}C concentration in the north atmosphere and biosphere in 1963 to ca 100% above the natural level (Münnich, 1963; Nydal & Lövseth, 1965; Olsson, 1968) and in the south atmosphere and biosphere in 1964 to ca 65% above the natural level (Levin, Münnich & Weiss, 1980). After the nuclear moratorium in 1963, excess ^{14}C has decreased with small interruptions caused by subsequent nuclear testing (Levin, Münnich & Weiss, 1980).

Second, with the development of the nuclear industry for electricity generation, anthropogenic ^{14}C has been produced in nuclear reactors and released to the atmosphere in measurable quantities. Previous high estimates of the growth of the nuclear industry until the year 2000 would imply a considerable increase of ^{14}C concentration in the environment. The present more realistic estimates (lower by a factor of 5) indicate a much lower ^{14}C concentration increase. Nevertheless, ^{14}C is one of the most important anthropogenic radionuclides with a long half-life and a significant collective dose burden (McCartney *et al*, 1986). The subject requires systematic attention.

Since 1967, we have been measuring ^{14}C in atmospheric CO_2 and in various biospheric samples in densely populated areas (Bratislava), in non-industrialized areas (Modra), as well as at sites close to nuclear power plants (Bohunice and Žlkovce) (Povinec *et al*, 1968; Chudý *et al*, 1970; Povinec *et al*, 1973). The main purpose of this paper is to show the present anthropogenic sources of ^{14}C in the atmosphere and biosphere and to estimate ^{14}C levels for the near future.

METHODS

Monthly samples of atmospheric CO_2 have been collected at Bratislava using the method of static absorption of CO_2 in NaOH solution. Simultaneously, at Bohunice (the first Czechoslovakian nuclear power station) and at Žlkovce (the monitoring station close to Bohunice), short-term sampling took place using bubblers filled with NaOH solution. We have also developed a method of sampling CO_2 and H_2O using a molecular sieve CALSIT 5A, for simultaneous sampling as well as measurement of ^3H and ^{14}C concentration in the atmosphere (Povinec, 1975). To measure ^{14}C concentration in the stack of the nuclear station as well as in the air around the station, the air sample was pressurized into a container and analyzed in the laboratory. The sample was first combusted in a stream of O_2 in an electric furnace, where CO, CH_4 and higher hydrocarbons were converted to CO_2. CO_2 was then absorbed in bubblers filled with NaOH and finally liberated from prepared $BaCO_3$ by adding H_3PO_4 and purified from electronegative impurities (Povinec *et al,* 1968) or converted to CH_4 (Povinec, 1972). Several samples were measured using the liquid scintillation (C_6H_6) method (Povinec *et al,* 1980). Simultaneous ^3H and ^{14}C measurements were made using doubly labeled CH_4 (Povinec, 1975). Low-level proportional counters of Oeschger type and multi-element proportional counters have been used for ^{14}C measurements. Sampling of air well in advance of the reactor operation enabled us to find pre-operational concentrations of ^{14}C and ^3H in the atmosphere and to study the reactor influence on these concentrations.

Tree-ring samples were taken from a pine tree that grew in a non-industrialized area.

BOMB EFFECT

The time variation of ^{14}C in atmospheric CO_2, plant materials, and tree rings is shown in Figure 1. The results follow a typical pattern of ^{14}C con-

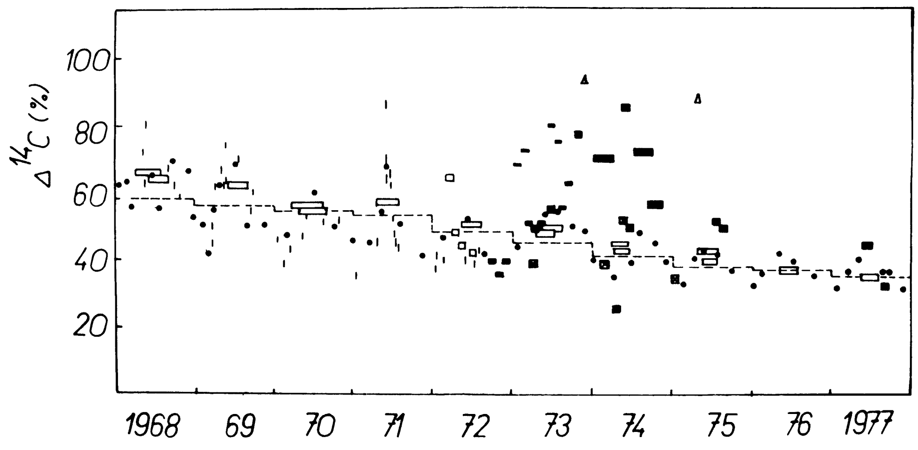

Fig 1. ^{14}C concentration in the troposphere. ● = Bratislava monthly samples; I = Bohunice short-term samples; ⊠ = Bohunice monthly samples; ■ = Žlkovce monthly samples; □ = Modra monthly samples; △ = the site of maximum ground concentration; ▭ = plant samples (tree-leaves, grass, etc) from Bohunice; --- = pine tree rings that grew in a non-industrialized area.

centration in the northern hemisphere as previously observed (Levin, Münnich & Weiss, 1980; Segl *et al,* 1983). Reasonable agreement between ^{14}C concentrations obtained by short-term sampling (3–4 hr) and the average monthly samples, as well as between atmospheric and biospheric (tree-leaves, grass, nuts, grains, sugar, tree rings) ^{14}C levels has been found.

The average annual ^{14}C concentration in atmospheric CO_2 collected at Bratislava from 1968 to the present is shown in Figure 2. These results clearly show a lower ^{14}C concentration (by 3–4%) in Bratislava air in comparison with the clean air ^{14}C background. This is caused by fossil-fuel combustion CO_2 sources in the environment of the sampling site. Bratislava samples (up to 1975) were taken in the center of the town and ^{14}C samples were later collected on the roof of a new department store on Mlynská dolina, which is on the outskirts.

The effects of nuclear testing in the atmosphere after 1963 can be identified in Figure 2, mostly in 1968–1970 and 1976, when the ^{14}C atmospheric inventory was disturbed by adding new quantities of freshly produced ^{14}C. Similar patterns are also observed for ^{3}H variations in atmospheric humidity measured for the same interval (Chudý *et al,* 1977).

REACTOR EFFECT

With increasing nuclear-produced electric power ^{14}C production has also become important as a new source of anthropogenic radioactivity. ^{14}C is produced in nuclear reactors mostly in the following reactions:

$$^{14}N\,(n, p)\,^{14}C \qquad \sigma = 175\ \mathrm{fm}^2$$
$$^{17}O\,(n, \alpha)\,^{14}C \qquad \sigma = 40\ \mathrm{fm}^2$$
$$^{13}C\,(n, \gamma)\,^{14}C \qquad \sigma = 0.1\ \mathrm{fm}^2$$

We calculated ^{14}C production in various types of nuclear reactors (a heavy water reactor with CO_2 coolant, a pressurized water reactor (PWR), a boiling water reactor (BWR), and a graphite reactor). To study local effects of nuclear reactors on ^{14}C concentration in the environment, we concentrated on the first two types of reactors (Chudý & Povinec, 1982).

The ^{14}C contribution to local contamination of the environment is measurable, in the case of nuclear reactors, using CO_2 under high pressure

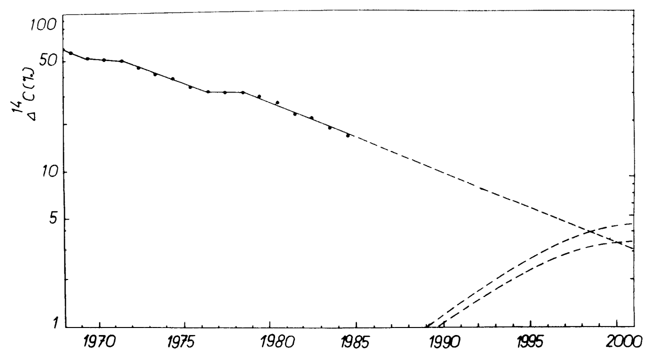

Fig 2. Annual ^{14}C concentration in the Bratislava air and predicted levels of bomb and reactor effects (lower curves show the boundary ^{14}C levels for predicted growth of nuclear industry).

as a coolant. As a consequence of the neutron irradiation of the coolant, ^{14}C is produced in the primary circuit of the reactor and is released to the environment through the ventilation system.

For the 150 MWe heavy water reactor cooled with CO_2, the saturation level in the ^{14}C activity of the coolant is reached after ca 10 weeks of reactor operation. The ^{14}C specific activity of the coolant will be ca 10 $GBq \cdot m^{-3}$ of CO_2 (Chudý & Povinec, 1982).

In light water reactors, ^{14}C production is predominantly from the reaction ^{17}O (n, α) ^{14}C on oxygen present in the molecule of water. The mean production of ^{14}C in the water of PWR is ca 0.3 $TBq \cdot GWe^{-1} \cdot yr^{-1}$. This is at least 20 times lower compared to a heavy water reactor. ^{14}C production in a BWR is almost the same as a PWR.

Except for the coolant, ^{14}C is also produced in the nuclear fuel, predominantly on ^{14}N and ^{17}O left in the fuel. ^{14}C production in this source is ca 4 times higher than in the light water coolant. However, ^{14}C from this source is released to the atmosphere in reprocessing plants. Therefore, similar to ^{85}Kr, a large reprocessing plant may be a more important source of ^{14}C than a nuclear reactor.

Figure 1 shows ^{14}C concentration in the atmosphere around the nuclear power plant at Bohunice (the 150 MWe heavy water reactor was put into operation at the end of 1972). A comparison with non-reactor sites (Bratislava air and tree-ring samples) shows a mean excess in Δ ^{14}C activity at Bohunice of ca 25%. Even higher ^{14}C levels (>100% above normal) were measured at the site of maximum ground concentration determined by given meteorologic conditions. On the other hand, ^{14}C in tree leaves (nut) and in nuts when compared with the clean air tree-ring samples shows a mean excess in Δ ^{14}C activity of ca 4% and 6%, respectively.

More recently (1982–1985), we measured ^{14}C concentration in the atmosphere and in the stack of the Bohunice nuclear power plant (2 reactors, each of 420 MWe). Measurements of ^{14}C concentration in the stack showed that these reactors release ^{14}C not only in the form of CO_2 (30%), but also in the form of CO, CH_4 and higher hydrocarbons (70%). A typical ^{14}C concentration in the stack is ca 20 $Bq \cdot m^{-3}$ (0.54 $mCi \cdot m^{-3}$) of air. This is very low in comparison with other radioactive gases, eg, ^{85}Kr (~70 $Bq \cdot m^{-3}$), ^{135}Xe (1000–3000 $Bq \cdot m^{-3}$) and ^{133}Xe (~1500 $Bq \cdot m^{-3}$ of air) (Cimbák, 1984).

We measured ^{14}C in the atmosphere around this plant at the site of maximum ground concentration. This short-term sample collection (2–3 hr) is compared with similar samples obtained in Bratislava and with the averaged monthly samples. Bohunice samples show an excess of ca 5% compared to the Bratislava short-term samples. However, they are within errors when compared with Bratislava monthly samples. Therefore, more measurements of ^{14}C in atmospheric and biospheric samples are necessary to better understand the influence of reactor operations on ^{14}C levels in the local environment.

FUTURE ^{14}C LEVELS

Figure 2 shows estimated future levels of ^{14}C in the atmosphere. The anthropogenic effect caused by nuclear bomb tests in the atmosphere has a decreasing tendency. The ^{14}C concentration in atmospheric CO_2 at present decreases with a half-life of ca 7 years. If there are no more significant nuclear bomb tests in the atmosphere, the bomb effect will disappear in the next 10–15 years and will be completely compensated by the fossil fuel (Suess) effect.

However the reactor effect will have an increasing tendency, although not so fast as was supposed earlier. According to the International Atomic Energy Agency (Bull IAEA, 1984, 1985), world nuclear power will reach ca 600 GWe by the year 2000. Mostly light water reactors are planned for construction. We calculated production rates of radioactive gases which are important for global contamination of the environment. The highest levels will be due to ^{85}Kr, ^{3}H and ^{14}C. Figure 2 shows the calculated increase of ^{14}C concentration in the atmosphere due to the reactor effect. The reactor contribution in the year 2000 will be about the same as the natural production rate. Therefore, the reactor effect will cause not only the local contamination of the environment around nuclear power plants, but beyond the year 2000 it will have an influence on global contamination, if new technical developments do not help to reduce ^{14}C discharges from reprocessing plants and reactors.

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