# ENVIRONMENTAL <sup>14</sup>C LEVELS AROUND THE 632 MWe NUCLEAR POWER PLANT KRŠKO IN YUGOSLAVIA

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ABSTRACT. Measurements of <sup>14</sup>C activity of atmospheric CO<sub>2</sub>, vegetables, and tree rings in the area of the 632 MWe power plant Krško in Slovenia, NW Yugoslavia, have been in progress since January, 1984. Sampling stations are located in the area ca 400km<sup>2</sup> and are distributed in the direction of the prevailing westerly winds. The closest sampling point is 1.5km NE of the plant exhaust stack, the farthest sampling point is 30km E of the plant. A sampling site at National Park Plitvice in central Croatia was chosen as the <sup>14</sup>C reference point not affected by the power plant.

An average excess of 2.2% above the reference point activity during normal periods of reactor operation was observed 1.5km from the plant smokestack. Calculations of the <sup>14</sup>C release from the power plant was estimated at 0.1 TBq/a. The tree ring activity near the plant followed the <sup>14</sup>C activity of the Northern Hemisphere in the past decade. No influence of the power plant release was observed on the tree ring activity.

#### INTRODUCTION

The global <sup>14</sup>C activity ("clean-air") level is governed in densely populated and industrial areas by opposing anthropogenic activities that result in 1) depletion of local <sup>14</sup>C concentration due to fossil fuel combustion in industrial areas, 2) increase of local <sup>14</sup>C concentration due to emission of <sup>14</sup>C from nuclear power plants and reprocessing plants.

The production of <sup>114</sup>C in nuclear power plants is recently of great interest, since almost all <sup>14</sup>C originating during operation of the reactor is released to the atmosphere. <sup>14</sup>C is produced in reactor systems mainly by activation processes in the moderator, the coolant, and the core. The <sup>14</sup>C release is of particular biologic importance since a significant fraction of organic matter consists of carbon. <sup>14</sup>C participates in the form of CO<sub>2</sub> in the biocycle and causes an additional contribution to the radiologic body burden. The local and collective doses due to <sup>14</sup>C release from reactors represent only a small fraction of the total dose. The man-made <sup>14</sup>C enters the carbon cycle resulting in global dispersion. Due to its long half-life (5730 yr) <sup>14</sup>C contributes to the long-term irradiation.

The Nuclear Power Plant Krško, with net electrical output of 632 MW, is the first nuclear power plant in Yugoslavia and began operation in 1981. It is located in southeast Slovenia, northwest Yugoslavia, on the left bank of Sava River in the industrial zone of Krško (45° 45′ 15″ N, 15° 31′ 14″ E), 37km northwest of Zagreb. The immediate and surrounding area of the plant belongs partly to the wooded secondary mountain ranges of the Alps to the north and to the wooded secondary mountain ranges of the Dinaric Mts to the west. The major part belongs to the wine-growing Panonian Boundary Hills and the Panonian Plain. The climate is moderate and partly influenced by the nearby Alps. The humidity is relatively high, ca 85%, with frequent fog, especially in the Sava River basin. The region is known for moderate winds mainly along the Sava River flow (NW-SE).

## MEASUREMENT PROCEDURE AT THE RUDJER BOŠKOVIĆ RADIOCARBON LABORATORY

Standard procedures for <sup>14</sup>C dating have been used to determine the increase of the natural <sup>14</sup>C level in the area of the nuclear facility. By applying this method, we can reach the lowest limit equal to ca 0.37 mBq/m<sup>3</sup> (0.01 pCi/m<sup>3</sup>). It is possible to determine an increase of <sup>14</sup>C level of only several percent above the present atmospheric level. This is the expected <sup>14</sup>C excess to be observed if the release of <sup>14</sup>C to the atmosphere is 0.2–0.4 TBq/a (5–10 Ci/a).

Measurements of <sup>14</sup>C activity in the area of Nuclear Power Plant Krško

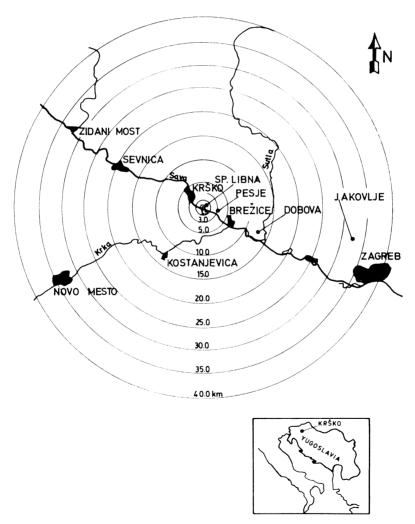


Fig 1. Sampling sites near Nuclear Power Plant Krško, NW Yugoslavia

were concentrated on  $CO_2$  since it is the main carrier of <sup>14</sup>C that enters plants growing in the area. Three measuring points were chosen, taking into account geography and weather: Libna, ca 1.5km from the plant smokestack, Dobova, 12km E, and Jakovlje near Bistra, 34km E (Fig 1).

A convenient reference sampling site was chosen for "clean-air" measurements in the scarcely populated area of Plitvice National Park, 150km south of the power plant (44° 50′ N, 15° 35′ E). Measurements of the <sup>14</sup>C concentration in the atmosphere at this site were performed independently from the monitoring program since 1979.

Samples of atmospheric  $CO_2$  collected continuously for a month were taken at three points since January 1984. Ca 125ml of saturated carbonatefree sodium hydroxide were exposed in a tray to the open atmosphere, out of reach of unauthorized persons. The absorbed  $CO_2$  in the form of sodium carbonate was processed in the same way as calcium or barium carbonates.  $CO_2$  is released by dissolving the sodium carbonate containing trapped atmospheric  $CO_2$  in diluted hydrochloric acid. After purification and removal of water vapor, the evolved  $CO_2$  was stored for subsequent processing (Srdoč *et al*, 1979).

Concentration of <sup>14</sup>C in cereals, vegetables, and grass were also measured. Samples of wheat, corn, carrot, beet, lettuce, onion, parsley, hay, and aftermath were taken during the growing period in 1984. Organic samples were collected and combusted by standard methods used in most <sup>14</sup>C laboratories.

To reconstruct past concentrations of  ${}^{14}$ C in the area, a linden tree (*Tilia* sp) was felled at Libna, ca 1.5km from the plant smokestack, and the activity of tree rings was measured. Individual rings were identified by B Petrić, Faculty of Forestry, University of Zagreb.

Results were processed on a HP-1000 computer. As the modern standard, 95% of the NBS oxalic acid activity was used. Several corrections of the data were made, such as barometric and temperature corrections (Obelić & Planinić, 1977; Obelić, 1980).

#### RESULTS

The results of our measurements are presented in graphs, showing the <sup>14</sup>C activity in percentage of the modern standard (0.95 of NBS oxalic acid). <sup>14</sup>C activity measurements in the environment of the Nuclear Power Plant Krško are shown in Figure 2. A mean difference of CO<sub>2</sub> activity of (2.2  $\pm$  0.9)% between the sampling point closest to the power plant (Libna) and sampling point Jakovlje was found. The maximal increase of atmospheric activity of (4.5  $\pm$  2.1)% coincides with the yearly change of 33% of fuel elements in the reactor core. By comparing the data measured at Dobova and that at Jakovlje it seems that the data at Dobova are 1.5% lower than at Jakovlje, although Dobova is closer to Krško. This could be explained by the Suess effect and the direction of local winds, because Dobova is more exposed to easterly winds blowing from the industrial center of Zagreb, while Jakovlje is shielded from these winds by Medvednica Mt (1035 asl) (see Fig 1).

Activity measurements of plants show no significant variations. They

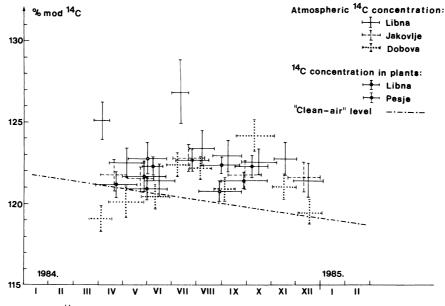


Fig 2.  $^{14}\mathrm{C}$  activity (% modern) of atmospheric  $\mathrm{CO}_2$  and vegetables in 1984 at 4 sampling points

agree, within the error, with the activity of the atmospheric  $CO_2$  in the Northern Hemisphere. The slightly lower values of <sup>14</sup>C activity of plants *vs* atmospheric  $CO_2$  sampled around the power plant can be explained by the fact that  $CO_2$  in the atmosphere was collected 24 hours a day, while plants absorb  $CO_2$  only during daylight, when atmospheric stability is lower and mixing is enhanced, which brings fresh, non-contaminated air in the monitored area. Both values are slightly above the clean-air level at the reference point.

<sup>14</sup>C activity in linden (*Tilia* sp) tree rings follow the general activity trend in the Northern Hemisphere. Results are compared with those in Germany (Levin, Munnich & Weiss, 1980) (Fig 3).

To calculate the <sup>14</sup>C release in Bq (Ci) per year from the observed increase of the <sup>14</sup>C concentration in the vicinity of the Nuclear Power Plant Krško, a record of the actual atmospheric dispersion conditions is needed. Source strength (in Bq/s) is given as:

$$Q = \frac{c}{f}$$

(Levin, Munnich & Weiss, 1980) where c is the <sup>14</sup>C excess (in Bq/m<sup>3</sup>) and f is the dispersion factor. The composite annual average value of the dispersion factor at 1500m from the smokestack, calculated from long-term synoptic records at the Nuclear Power Plant is  $10^{-6}$  s/m<sup>3</sup> (FSAR, 1981). For a 2.2% excess, an average <sup>14</sup>C release of 0.1 TBq (2.7 Ci) per year was estimated which agrees with data obtained for other power plants (Winkelmann *et al*,

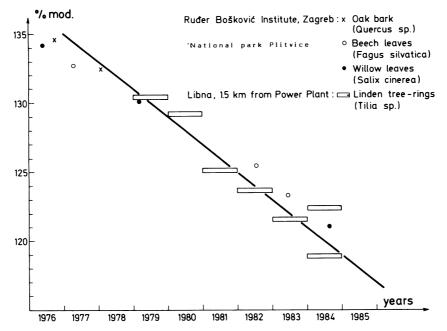


Fig 3. <sup>14</sup>C activity (% modern) of tree rings, oak bark, willow, and beech leaves at several sampling sites, 1976–1985. Full line represents the mean <sup>14</sup>C activity in the Northern Hemisphere according to Levin, Münnich and Weiss (1980). See text for details about selection of sampling sites.

1982). This preliminary result can give only the order of magnitude of the actual <sup>14</sup>C emission because the long-term dispersion factor is based on the mean dispersion conditions during the whole year and the excess of <sup>14</sup>C activity was observed only in March and July 1984. Such calculation was useful because no direct emission measurement in the power plant stack was done up till now.

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