IMPACT OF REFUELLING OF THE KRŠKO NUCLEAR POWER PLANT ON THE ^{14}C ACTIVITY IN THE ATMOSPHERE AND PLANTS

krajcar@irb.hr

I. Krajcar Bronić[1], B. Obelić[1], N. Horvatinčić[1], A. Sironić[1], J. Barešić[1], A. Rajtarić, B. Breznik[2], A. Volčanšek[2]

[1]Ruđer Bošković Institute, Zagreb, CROATIA

[2]Nuclear Power Plant Krško (NEK), Krško, SLOVENIA

INTRODUCTION

^{14}C is formed in upper layers of the atmosphere and as CO_2 it is distributed uniformly throughout the atmosphere. The equilibrium between radioactive decay of ^{14}C and its production rate has been established. The natural ^{14}C concentration (specific activity of ^{14}C) in the Earth's atmosphere and biosphere equals 226 Bq/kg C.

In the second half of the 20th century the natural ^{14}C distribution was disturbed by atmospheric bomb tests. In 1963 the atmospheric specific activity of ^{14}C reached a maximum of twice the natural activity, and after the ban of atmospheric bomb tests it approaches the natural (non-disturbed) values.

^{14}C is produced also in nuclear power plants. The emitted CO_2 enters the carbon cycle and through food chain it may contribute to the dose of the local population. The aim of the monitoring ^{14}C is to determine its distribution in a close vicinity of the Nuclear Power Plant Krško (NEK) in Slovenia (Figure 1) and to estimate possible contribution of NEK to the effective dose of the local population through food chain.

Figure 1. Sampling sites.

Figure 2. Sampling locations around NEK Locations **A** and **B**: atmospheric CO_2. Locations **C – R**: biological samples

SAMPLING AND MEASUREMENTS

Atmospheric CO_2 was collected regularly every two months at locations A and B inside the NEK area (Figures 2 and 3) by absorption of CO_2 in saturated NaOH solution. In the laboratory, CO_2 was produced and used for synthesis of benzene to be measured by LSC Quantulus. Results are expressed in mBq/m³ of air.

Biological samples (apples (mostly), corn, wheat, various vegetables and grass) were collected twice a year (in June/July and September/October) on several locations very close to the plant (200 – 400 m from the release point, inner circle) and in a radius of about 1 km (outer circle), as well as on the control site at Dobova, 12 km from the plant (Figures 1 and 2). Samples were dried, carbonized at 600°C, combusted to CO_2 and then absorbed in mixture of Carbosorb E and Permafluor E. The cocktail was measured by LSC Quantulus. Results are expressed as relative specific activity $a^{14}C$ in units percent Modern Carbon (pMC), where 100 pMC = 226 Bq/kg C. Results are normalized to $\delta^{13}C$ by using the mean value of measured $\delta^{13}C$ values of 71 samples [Sturm et al, J. Env. Radioact. 2012].

Location **B**, CO_2, Sept. 2006

Location **J**, July 2006, apples

Figure 3. Sampling locations around NEK

Figure 5. Wind rose in the vicinity of NPPK, measured on a 10 m high column (according to the Environmental Agency of the Republic of Slovenia)

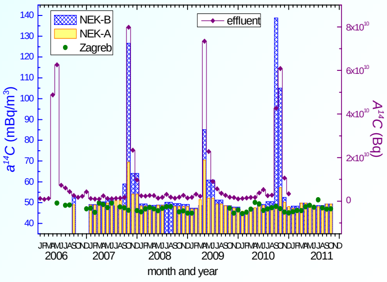

Figure 4. ^{14}C activity concentration in atmospheric CO_2 at locations **A** and **B** and comparison with the results obtained in Zagreb (40 km E) (left ordinate) and monthly ^{14}C activity in effluent released through the plant ventilation system (right ordinate) [*]. Atmospheric ^{14}C activity at the location **B** is always slightly higher than that at the location **A** since it is located at local prevailing wind direction (Figure 5). The increase in the refuelling outage periods and immediately thereafter is evident.

* V. Stibilj, B. Svetek, Z. Trkov, A. Volčanšek, B. Breznik, *Measurement of ^{14}C activity in exhaust air at the Krško Nuclear Power Plant,* In: Proc. 8th Symp. CRPA, Krk, 13.-15.4.2011., Eds: Krajcar Bronić I., Kopjar N., Milić M., Branica G. HDZZ, Zagreb. 2011. pp. 341-346, www.hdzz.hr

Table 1. Mean values of $a^{14}C$ (pMC) in biological samples in all sampling campaigns.
Inner circle – locations **C, D, E, I, J, L** and **R**
Outer circle – locations **F, G, H, K, M, N, O, P** and **Q**

Collection month/year	$a^{14}C$ (pMC) **inner circle**	$a^{14}C$ (pMC) **outer circle**	$a^{14}C$ (pMC) **Control site (Dobova)**
07 / 2006	120.6 ± 11.0	108.3 ± 3.0	103.2 ± 1.5
10 / 2006	112.3 ± 12.0	105.1 ± 2.0	104.0 ± 1.5
07 / 2007	103.7 ± 3.9	103.7 ± 2.8	105.6 ± 1.9
09 / 2007	106.8 ± 1.7	105.7 ± 2.6	103.8 ± 1.8
07 / 2008	109.6 ± 3.5	107.3 ± 1.7	104.1 ± 2.3
10 / 2008	109.1 ± 3.3	108.4 ± 3.5	104.4 ± 2.7
06 / 2009	117.0 ± 11.2	110.5 ± 2.0	105.4 ± 1.4
09 / 2009	112.0 ± 8.5	104.8 ± 2.5	102.0 ± 1.6
07 / 2010	105.0 ± 4.3	104.9 ± 2.3	103.7 ± 1.6
09 / 2010	107.2 ± 2.4	106.4 ± 3.4	102.4 ± 2.5
07 / 2011	107.8 ± 4.8	107.0 ± 4.8	104.0 ± 1.6
09 / 2011	103.2 ± 3.6	104.5 ± 1.9	106.8 ± 2.5

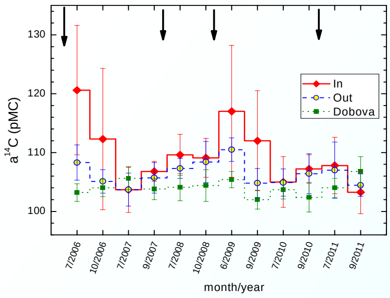

Figure 6. Mean values of $a^{14}C$ in biological samples from the vicinity of NEK for all sampling campaigns 2006 - 2011. Data from Table 1. Arrows indicate the refuelling outage periods (4/2006, 10/2007, 4/2009, 10/2010).

RESULTS

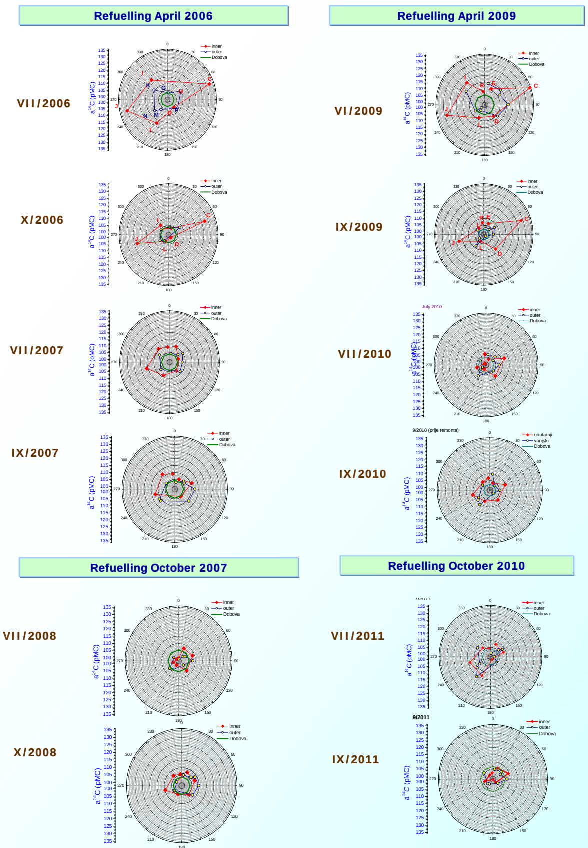

Figure 7. Spatial distributions of ^{14}C activities in biological samples around NEK for all campaigns since 2006 showed the dependence on the distance and the orientation. Increased activities were observed in SW – NE direction of most pronounced winds. As expected, higher activities of ^{14}C were measured in biological material which used CO_2 during and immediately after the refuelling outage periods. In 2006 and 2009 the power plant was refuelled in spring period (April), so the plants, having their vegetation period immediately afterwards, used more active CO_2. Therefore, at all locations the highest ^{14}C activity was in July 2006 and June 2009 (Table 1), since both campaigns were performed after the refuelling in April of the corresponding year. On the other hand, in 2007 and 2010 the refuelling was in autumn after harvesting. The mean ^{14}C activities in both sampling periods in 2007 and 2010 were similar to the ^{14}C activity measured at the control point Dobova.

DISCUSSION

In order to estimate the contribution of the ^{14}C in the NEK effluents to the effective dose of the local population through food chain, it is necessary to establish also the dose due to naturally occurring ^{14}C. Furtermore, it is difficult to realistically estimate the proportion of consumption of local products.

The most conservative model of ingestion takes "the most exposed adult person" who would consume only products (apples) from the inner circle NEK throughout the year. In that case, and for year 2006 with the highest ^{14}C activity measured, the increase of the total annual dose due to ingestion of ^{14}C is about 0.1%. Since this unrealistic approach overestimates the dose, we propose a more realistic model (however, still rather conservative) of ingestion of the food from both inner and outer circle during 2 months of the year, and the rest from the remote (control) site. The increase of the natural dose due to ingestion of ^{14}C is then less than 0.015% in worst case, and is practically indistinguishable from the natural dose, i.e., the difference in doses is within the combined experimental and model errors.

Acknowledgement: The work performed under the Project 098-0982709-2741 by the Ministry of Science, Education and Sport, Republic of Croatia and financed partly by NEK.

CONCLUSION

Increase of ^{14}C activity in atmospheric CO_2 was always observed during and immediately after the refuelling of the nuclear power plant, which has been performed every 18 months, and it is more pronounced on the location in the SW-NE direction that coincided with the most pronounced wind directions. ^{14}C activity in plants collected close to the Krško NPP is always higher than the activities on the control point, and depends both on the distance from the exhaust of the plant ventilation system and on wind direction. Significantly higher activities in plants collected after the spring refuelling in 2006 and 2009 were measured. This can be explained by the influence of plant effluents during the summer when the process of photosynthesis is the most prominent, while autumn effluents (2007 and 2010) do not significantly influence the ^{14}C activity in plants. The maximum increase of total annual dose to local population due to the release of ^{14}C from the Krško power plant (NEK) in the years of spring refueling was estimated to be 0.015 %, which is negligible and within the errors of the estimate.