^{14}C in biological samples from the vicinity of NPP Krško

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Abstract

Monitoring ^{14}C activity in the atmospheric CO_2 and in biological samples (fruits and vegetables) in the close vicinity of the NPPK was performed regularly since 2006 to estimate the possible influence of the plant on environmental ^{14}C levels and the possible contribution to the effective dose of local population through food chain.

Mean values of ^{14}C activity in atmospheric CO_2 in the years when the refuelling was not performed represent a barely visible increase in relation to the activities of ^{14}C in the atmosphere at the control point. Immediately after refuelling the atmospheric ^{14}C activity was increased for a short period of time. Spatial distributions of ^{14}C activities in biological samples show the dependence on distance and orientation. In all campaigns the highest activities were obtained at the same locations in the SW-NE direction that coincided with the most pronounced wind directions. Estimated contribution of ^{14}C released by NPPK to the natural dose is less than 1 μSv.

Introduction

^{14}C is formed in upper layers of the atmosphere in a reaction of neutrons from the cosmic rays and nitrogen atoms. All carbon isotopes take part in chemical reactions, hence ^{14}C is also oxidized into $^{14}CO_2$ and distributed uniformly throughout the atmosphere. Plants assimilate $^{14}CO_2$ through the photosynthesis process, and animals are fed by plants, thus the ^{14}C isotope is a part of the natural carbon cycle. In the past, the equilibrium between radioactive decay of ^{14}C and its production rate has been established, and the natural ^{14}C concentration, or better to say, specific activity of ^{14}C, in the Earth's atmosphere and biosphere is approximately constant and it equals 226 Bq/kg of carbon. Assuming atmospheric CO_2 concentration of 370 ppm, this equilibrium specific activity of ^{14}C in the atmosphere corresponds to 0.037 Bq/m^3 of air.

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

^{14}C is produced also in nuclear power plants. The emitted $^{14}CO_2$ enters the carbon cycle, and through food chain it may contribute to the dose of the local population. The aim of this study was to determine distribution of ^{14}C in a close vicinity of the Nuclear Power Plant Krško (NPPK) in Slovenia close to the border with Croatia, and to estimate possible contribution of NPPK to the effective dose of local population through food chain.

Sampling and measurements

Atmospheric CO_2 was collected at two locations inside the NPPK area, marked as **A** and **B** in Figure 1, close to the release point of the plant ventilation system. Integral samples were collected since 2006 on a regular bimonthly basis by absorption of CO_2 in saturated NaOH solution. Na_2CO_3 thus produced was in the laboratory dissolved in HCl, and the obtained CO_2 was used for benzene synthesis. Benzene samples were measured by a liquid scintillation counter *Quantulus* (Horvatinčić et al. 2004).

Specific ^{14}C activity of biological samples (apples, corn, cereals, borecole, grass) was measured in immediate vicinity (locations **C**, **D**, **E**, **I**, **J**, **L** and **R**, 200 – 400 m from the NPPK release point), and in a wider environment (locations **F**, **G**, **H**, **K**, **M**, **N**, **O**, **P** and **Q**, about 1000 m) of NPPK (Fig. 1). As a control site, where no influence of the plant is expected, we have chosen village Dobova, about 10 km SE from NPPK. Two sampling campaigns have been performed each year since 2006, in June/July and in September/October. Samples were dried, carbonized (pyrolysis at 600°C), and combusted in a stream of oxygen. The obtained CO_2 was absorbed in a mixture of Carbosorb®E and Permafluor®E. ^{14}C activity was measured by the LSC *Quantulus 1220* (Horvatinčić et al, 2004; Krajcar Bronić et al, 2009).

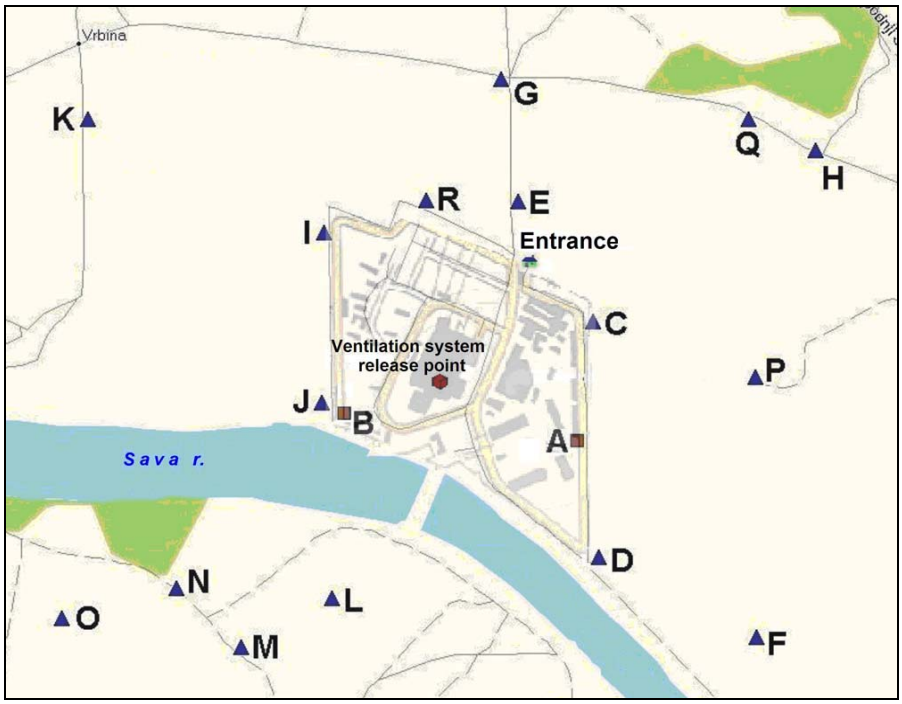

Fig. 1. Sampling locations in the vicinity of Nuclear Power Plant Krško. Locations A and B for atmospheric CO_2. Locations C – R for biological samples.

Results

The results are usually expressed as the relative specific activity $a^{14}C$, defined as the ratio of the specific activity of the sample and that of the atmosphere undisturbed by anthropogenic influence, in "percent of Modern Carbon", or pMC. Consequently, relative specific activity of 100 pMC corresponds to specific activity of 226 Bq/kgC or to the $^{14}CO_2$ activity concentration of 0.037 Bq/m^3 of air.

Activity in atmospheric $^{14}CO_2$

^{14}C activity concentrations in the atmosphere at two locations (**A**, **B**) inside the NPPK are shown in Figure 2. For comparison, a part of long-term atmospheric ^{14}C data in the city of Zagreb is shown (Krajcar Bronić et al, 2009), as well as the integrated monthly ^{14}C activities released through the plant ventilation system. Atmospheric ^{14}C activity at the location **B** is always slightly higher than that at the location **A**. This difference can be explained by the position of location **B** relative to the release point and the local prevailing wind direction SW – NE (Fig. 3), and is particularly evident in the refuelling outage periods and immediately thereafter. The half-life of the released ^{14}C in the atmosphere is estimated to app. 1.5 months. During the refuelling in October 2007 higher $a^{14}C$ values at the site **B** were observed (102 mBq/m^3) than during the refuelling in April 2009 (69 mBq/m^3) probably due to the of shorter sampling period (ten days in 2007 in comparison to three weeks in 2008), which coincided better to the highest ^{14}C release to the atmosphere.

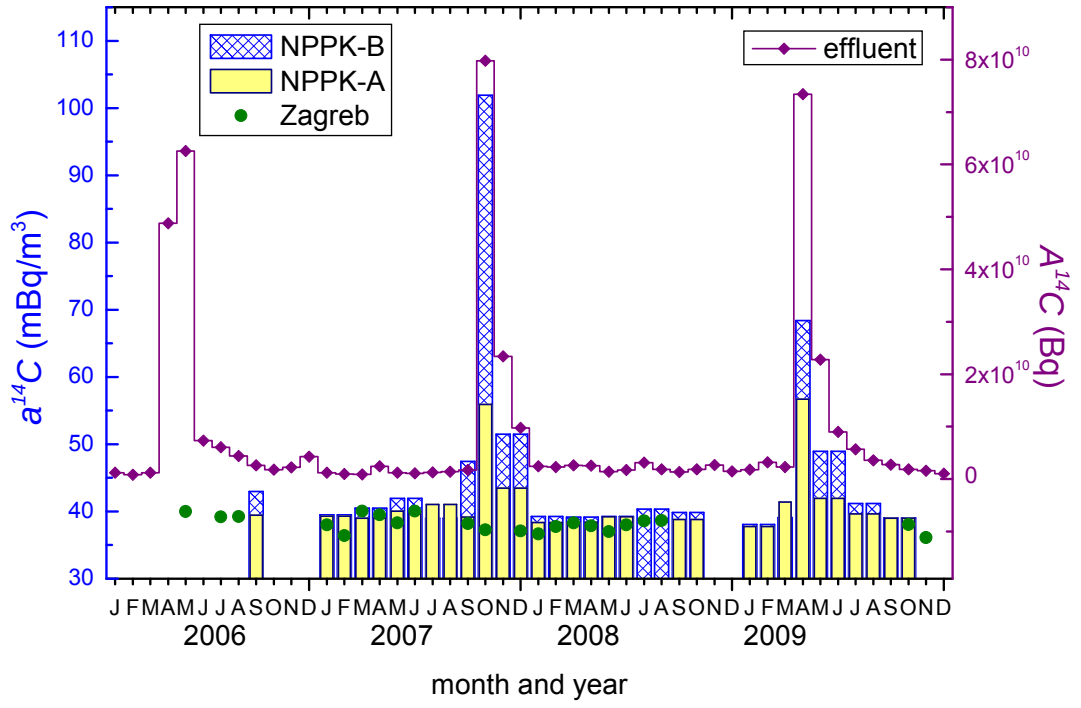

Fig. 2. ^{14}C activity concentration in atmospheric CO_2 at locations A and B and comparison with the results obtained in Zagreb (40 km E) (left ordinate). Monthly ^{14}C activity in effluent released through the plant ventilation system (right ordinate).

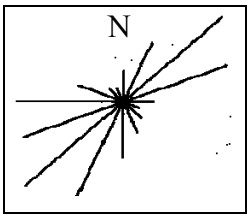

Fig. 3. Wind rose in the vicinity of NPPK, measured on a 10 m high column (according to the Environmental Agency of the Republic of Slovenia)

Mean values of $a^{14}C$ in atmospheric CO_2 in the years when the refuelling was performed (2007 and 2009) are 41.7 and 41.5 mBq/m^3 at the location **A**, and 47.9 and 44.2 mBq/m^3 at the location **B**. In 2008, when no refuelling was performed these values were 38.7 (location **A**) and 39.6 mBq/m^3 (location **B**). They represent a barely visible increase in relation to the activities of ^{14}C in the atmosphere measured in Zagreb (38.3 mBq/m^3 in 2007 and 37.8 mBq/m^3 in 2008), where the effect of reducing ^{14}C atmospheric activity is possible due to combustion of fossil fuels.

Activity in biological samples

In Figure 4. we present the spatial distributions of measured ^{14}C activities (expressed as specific activities in pMC) for all eight sampling campaigns, as well as the corresponding polar diagrams that show the dependence on distance and orientation. In polar diagrams we have separated closer locations **C**, **D**, **E**, **I**, **J**, **L** and **R** (inner circle) and farther locations **F**, **G**, **H**, **K**, **M**, **N**, **O**, **P** and **Q** (outer circle), and have also shown $a^{14}C$ values at the control point Dobova. Increased activities were observed in all sampling campaigns in SW – NE direction of most pronounced winds. As expected, higher activities of ^{14}C were measured in biological material which used CO_2 in the refuelling outage period which was performed during and immediately after the vegetation period. In 2006 and 2009 the power plant was refuelled in spring period, so the apples, having their vegetation period immediately afterwards, collected more active CO_2. Therefore, at all locations the highest ^{14}C activity was in July 2006 and June 2009, since both campaigns were performed after the refuelling in April of the corresponding year. At the other hand, in 2007 the refuelling was in autumn after the apples were harvested, and the mean ^{14}C activities in both sampling periods in 2007 (July and September) were similar to the activity measured at the control point Dobova (Table 1).

Based on predetermined spatial distribution of ^{14}C activities in biological samples around the plant, it is clear that important results could be obtained only within a few hundred meters distance from the plant (Breznik et al, 2008). This close distance is not very convenient for testing the dispersion model but enabled to detect higher activities in a shorter time interval.

Table 1. Mean values of $a^{14}C$ (pMC) in biological samples in all eight 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. * Measurements of samples from 9/2009 have not yet been completed.

Collection time	$a^{14}C$ (pMC)	$a^{14}C$ (pMC)	$a^{14}C$ (pMC)
	Inner circle	Outer circle	Control point
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	109.1 ± 3.0	104.4 ± 2.7
06 / 2009	117.0 ± 11.2	110.5 ± 2.0	105.4 ± 1.4
09 / 2009	115.9 ± 11.4*	103.2 ± 2.3*	102.0 ± 1.4

Discussion

Equivalent annual dose E received by an average adult person by ingestion of food of specific ^{14}C activity $a^{14}C$ (Bq/kgC) can be estimated as

$$E = e \times a^{14}C \times m \times t$$

where t is a 1-year period (365 days), m is a mass of carbon ingested daily by food (0.3 kg, ICRP, 1996), $a^{14}C$ is measured ^{14}C specific activity (Bq/kg C), and e is the ingestion dose coefficient for ^{14}C, i.e., expected effective dose per unit ^{14}C activity, 5.8×10^{-10} Sv/Bq (ICRP, 1996).

It is difficult to realistically estimate a possible increase of the annual dose in case of ingestion of fruit grown in the vicinity of NPPK. As we have shown here, there are spatial and temporal variations in $a^{14}C$. It is also difficult to estimate the fraction of total ingested food that originates from the close vicinity of NPPK. It is reasonable to assume that the local products are dispersed in the general food supply system, and not all consumed by local population. The simplest and the most conservative model of ingestion takes "the most exposed adult person" who would consume only apples from the close environment of NPPK throughout the year. In that case, and for the year 2006 (highest mean ^{14}C activity) the increase of the total annual dose is about 0.1%. Since this the most conservative estimate is unrealistic and not probable, we propose the following more realistic although still rather conservative model. We suppose that: i) in the daily consume of 0.3 kg of carbon, one half comes from the fruits; ii) 6 months the fruits from the environment of NPPK are not available, and during this period one can ingest only the food which has $a^{14}C$ as it is at the control site; iii) during the other 6 months, ⅓ of the total ingested carbon comes from fruits from any point of the monitored area around the NPPK, and the rest ⅔ come from the control site. Therefore, this model is equivalent to the assumption of ingestion of the apples from the vicinity of the power plant during 1 month in a year, while during the rest of the year carbon comes from the wider area. Taking into account these assumptions, we can calculate the increase of the equivalent ^{14}C dose relative to the natural ^{14}C dose measured at the control point, as well as relative to the total natural dose in our country (1.22 mSv). The results for each year since 2006 are shown in Table 2.

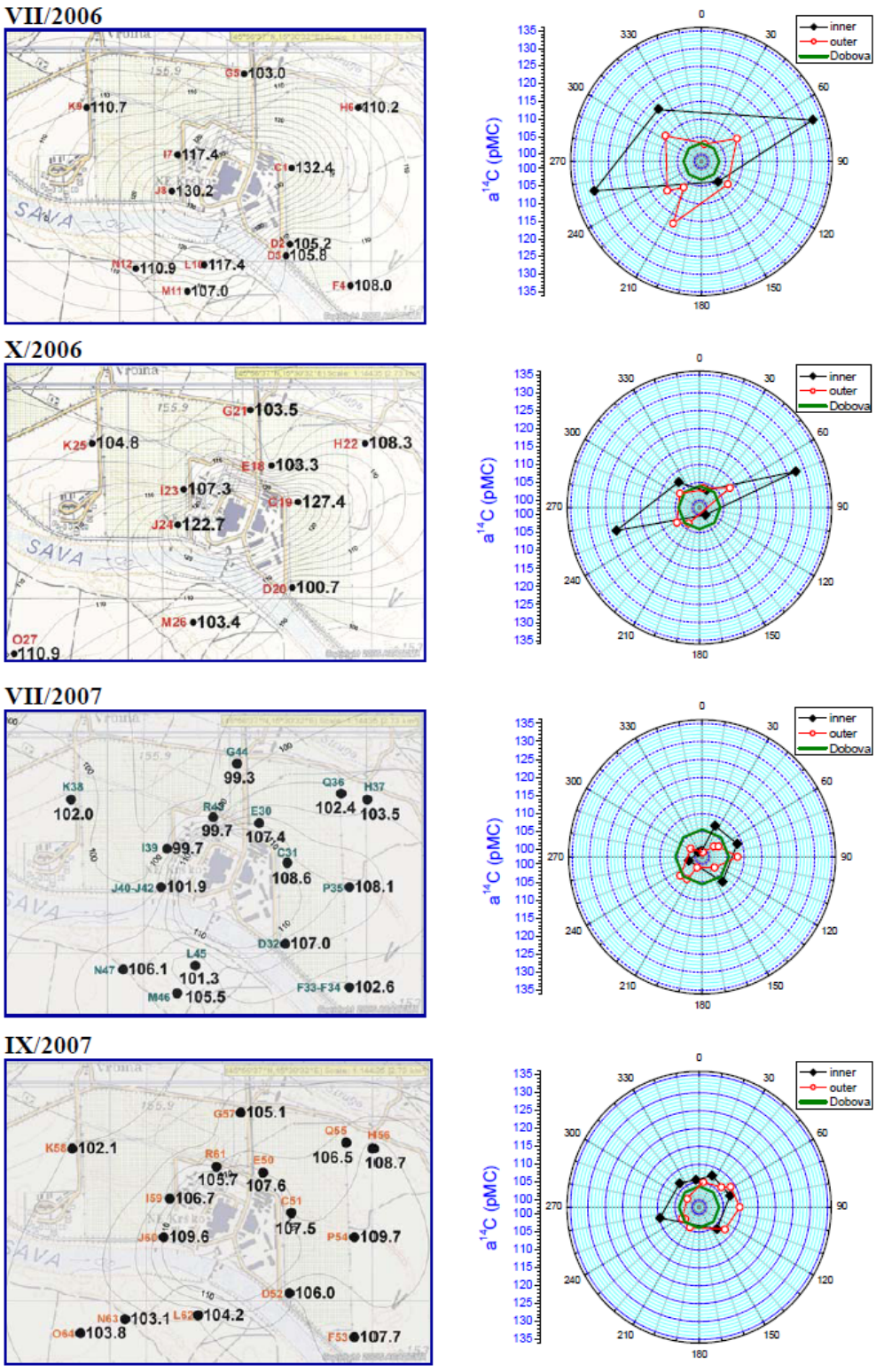

Figure 4. Spatial distribution and polar diagrams of relative specific ^{14}C activity (expressed in pMC)

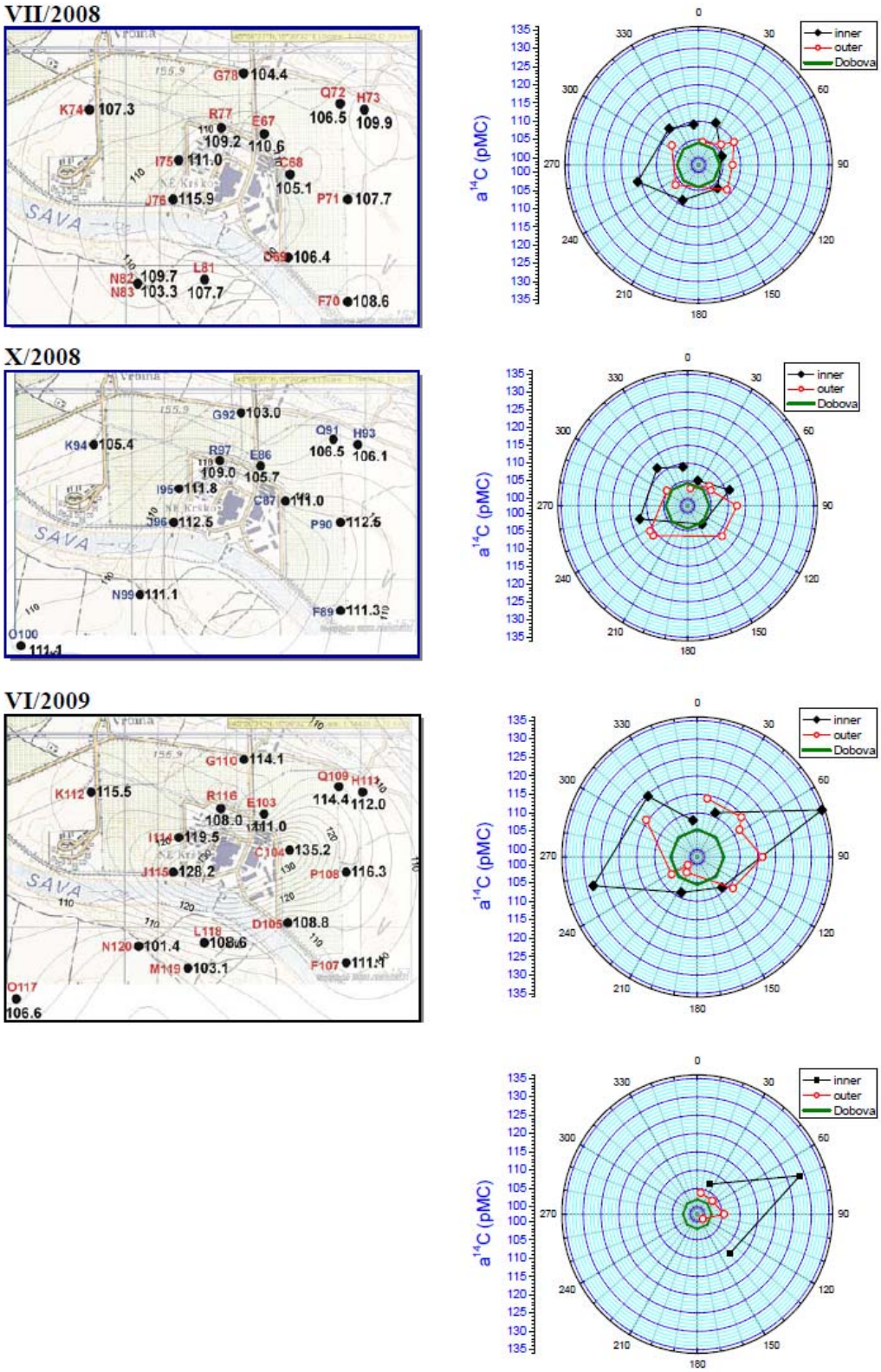

Figure 4 continued. Spatial distribution and polar diagrams of relative specific ^{14}C activity (expressed in pMC). Data for the sampling campaign in September 2009 not yet completed.

Table 2. Increase of equivalent ^{14}C dose relative to the natural ^{14}C dose measured at control point, as well as well as the increase relative to the total natural dose in our country (1.22 mSv)

Year	$a^{14}C$ [pMC] NPPK *	$a^{14}C$ [pMC] Dobova	^{14}C dose [μSv] NPPK	^{14}C dose [μSv] Dobova	Increase of ^{14}C dose (%)	Increase of total dose (%)
2006	111.6	103.5	14.95	14.86	0.65	0.0079
2007	105.0	104.7	15.03	15.03	0.02	0.0003
2008	108.8	104.3	15.02	14.97	0.35	0.0044
2009	110**	103.8	14.97	14.89	0.50	0.0061

* mean value of ^{14}C activity in inner and outer circle of NPPK
** data for September 2009 not yet complete

Conclusion

Monitoring of $a^{14}C$ in biological samples in immediate vicinity and wider environment of the Nuclear Power Plant Krško in Slovenia resulted in spatial distribution of $a^{14}C$ that is clearly determined by the dominant wind directions (SW – NE) and influenced by the outflow of ^{14}C from the release point of the plant ventilation system. The difference to the natural present-day $a^{14}C$ is determined relative to the control site Dobova, about 10 km SE from NPPK. At most sites the difference ranges from 0 to several pMC, being the highest 29 pMC at site **J** in July 2006. However, these differences do not affect significantly the total annual dose due to natural radiation sources, according to our model of ingestion.

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