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HOW TO ESTIMATE THE EFFECTIVE DOSE DUE TO INGESTION OF ^{14}C

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ABSTRACT

Systematic and continuous monitoring ^{14}C activity in atmospheric CO_2 and biological samples in the vicinity of the Krško Nuclear Power Plant (NEK) in Slovenia has been performed since 2006. The aim of the monitoring was to determine ^{14}C distribution in a close vicinity of the power plant and to estimate a possible contribution of NEK to the effective dose to the local population through food chain. This paper describes a model of food origins developed for the purpose of estimating the effective dose from ingestion of ^{14}C via local food. Some other approaches of diet estimation are also discussed. The slightly increased ^{14}C specific activity in plants very close to NEK could not change significantly the effective dose to the local population due to ingestion of ^{14}C when compared to the effective dose from the ingestion of natural ^{14}C.

1. INTRODUCTION

Radiocarbon (^{14}C) is a cosmogenic isotope that is incorporated into various carbon-bearing compounds, such as CO_2 in the atmosphere, organic molecules (plants and animals) in the biosphere, and various inorganic compounds. The equilibrium between the specific activity of ^{14}C (A^{14}C, expressed in units Bq/kg of carbon, Bq/kgC) of the atmosphere and the biosphere is almost immediately established [1,2]. This is the basis for the radiocarbon dating method [e.g. 2]. ^{14}C has relatively long half-life (5730 yr) and it decays by emitting low-energy (<150 keV) beta particles. It behaves in the same way as other carbon isotopes (^{12}C 99%, ^{13}C 1%) in the environment and in the body. The specific activity of ^{14}C is the same in terrestrial plants as in the atmospheric CO_2 used for photosynthesis [3]. Carbon, including ^{14}C, is thus a constituent of food and contributes to the natural irradiation of man through the food chain [3,4]. Average annual human exposure to natural sources of ionizing radiation is 2.4 mSv, and the average annual contribution of ^{14}C through food chain to this dose is about 12 μSv or about 0.5%.

Anthropogenic activities influenced the natural ^{14}C levels. Intense atmospheric nuclear and thermonuclear bomb tests after the World War II caused an increase in atmospheric A^{14}C by a factor of 2. After the atmospheric test ban treaty in 1963, mixing of the atmosphere with other carbon reservoirs (oceans and terrestrial biosphere) led to a decrease in the atmospheric A^{14}C that recently almost reached the pre-bomb levels [1,2,5]. Intense use of fossil fuels (oil, coal) mostly for traffic and as sources of energy for industry and heating caused an increase in the concentration of CO_2 in the atmosphere, but lowered the atmospheric A^{14}C because fossil fuels do not contain ^{14}C [1,2]

Anthropogenic sources that locally influence ^{14}C levels are various nuclear facilities. ^{14}C in gaseous effluents from a nuclear power plant is of the interest here. Plants can use the released CO_2 for photosynthesis and thus ^{14}C in the food chain may contribute to the effective dose to man due to ingestion. In case of operation of nuclear power plants, there is usually a very low annual release of typical fission products and therefore measurements could bring to the front an increase of ^{14}C in some local samples. Carbon ^{14}C is likely to be one of the most visible contributors to the effective dose in such a case. This paper presents various scenarios of the conservative effective dose estimation to the average inhabitant of the vicinity of the Krško Nuclear Power Plant (NEK in further text) based on the experience gained by continuous monitoring ^{14}C in the atmospheric CO_2 and in plants in the vicinity of NEK.

2. RESULTS OF MONITORING

NEK established an extensive effluent monitoring programme including also measurement ^{14}C in the air of the ventilation system [6], as well as the measurements of imissions in the surrounding [7]. Systematic and continuous monitoring of ^{14}C specific activity in atmospheric CO_2 and biological samples in the vicinity of NEK has been performed since 2006 [8,9]. Atmospheric CO_2 has been collected at two sampling locations (A and B, Figure 1) in two-month intervals, while during the refuelling outage periods the sampling period has been shorter, 2 – 4 weeks. Biological samples, mostly apples, vegetable, cereals, corn, etc., have been collected twice a year: in early summer (June/July) and in early autumn just before harvesting. The sampling locations are divided into two circles (Figure 1): the "inner circle" encompasses locations closest to the fences of the restricted area, while locations about 300 – 600 m off the fences form the "outer circle". The aim of the monitoring is to determine ^{14}C spatial and temporal distribution that might be influenced by atmospheric discharges in a close vicinity of the power plant. A control location Dobova, 12 km SE, has been chosen as a "clean-air" site with natural $A^{14}C$, i.e., it is not influenced by the gaseous effluent from NEK.

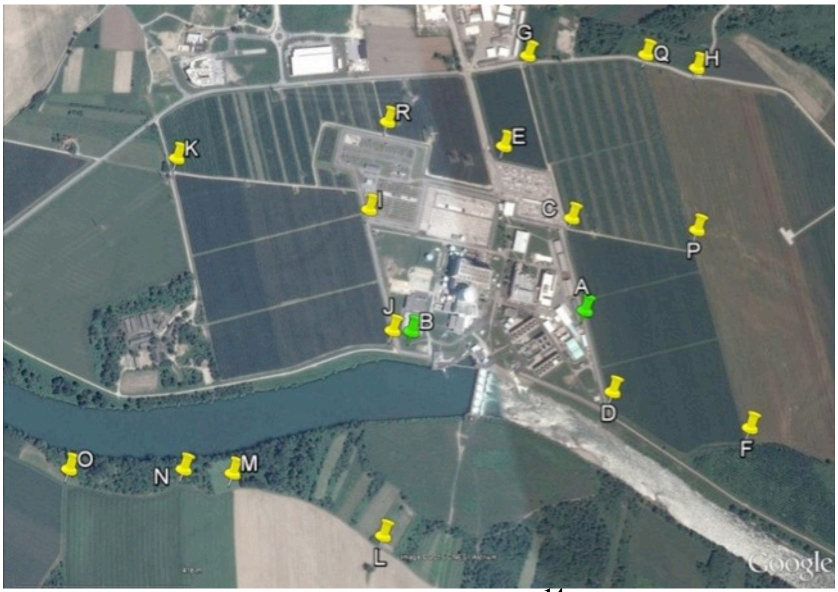

Figure 1. Sampling locations for monitoring ^{14}C in the vicinity of NEK. A, B – atmospheric CO_2; inner circle (E, C, D, J, I, R) – apples; outer circle (G, Q, H, P, F, L, M, N, O, K) – apples, cereals, corn.

Atmospheric CO_2 is collected in the field by absorption on saturated NaOH. The obtained Na_2CO_3 is then in the laboratory converted to benzene [8-11]. Biological samples are dried, carbonized and combusted in a stream of pure oxygen, and the obtained CO_2 is absorbed in a mixture of Carbosorb®E and Permafluor®E. The ^{14}C

specific activity is measured by liquid scintillation counter LSC Quantulus 1220 in both cases [10,11].

The main results of the ^{14}C monitoring program can be summarized as follows [9]. The influence of the released air-born ^{14}C activity is measurable in both atmospheric CO_2 and in plants; the higher the activity of gaseous effluent, the higher the atmospheric and plant $A^{14}C$. However, the influence is temporally and spatially limited. During and immediately after the refuelling outage periods, that are performed in 18-month intervals, the atmospheric ^{14}C specific activity at the sampling sites inside the NEK area is higher than that at the sites not influenced by the NEK effluents (e.g., Zagreb). It drops down to almost natural levels within 2-3 months after the most intense releases. Plants growing in the vicinity of NEK may take the released CO_2 for the photosynthesis during the vegetation period. Due to the 18-months period of the refuelling outage, we have to distinguish among the years when the refuelling outage takes place in spring (April-May) from those carried out in autumn or even without it ("other years" further in the text). The ^{14}C specific activities in plants after the spring refuelling outage, i.e., at the beginning of the vegetation period, are higher than those in other two cases (Figure 2). The type of the plant considered in the monitoring program does not have any influence on the $A^{14}C$ values at a certain location. Spatial distribution of biological $A^{14}C$ is determined by the distance from the gaseous release point and by the prevailing wind direction, SW – NE. The average $A^{14}C$ in plants at the control location Dobova does not differ from the average atmospheric ^{14}C specific activity in Zagreb.

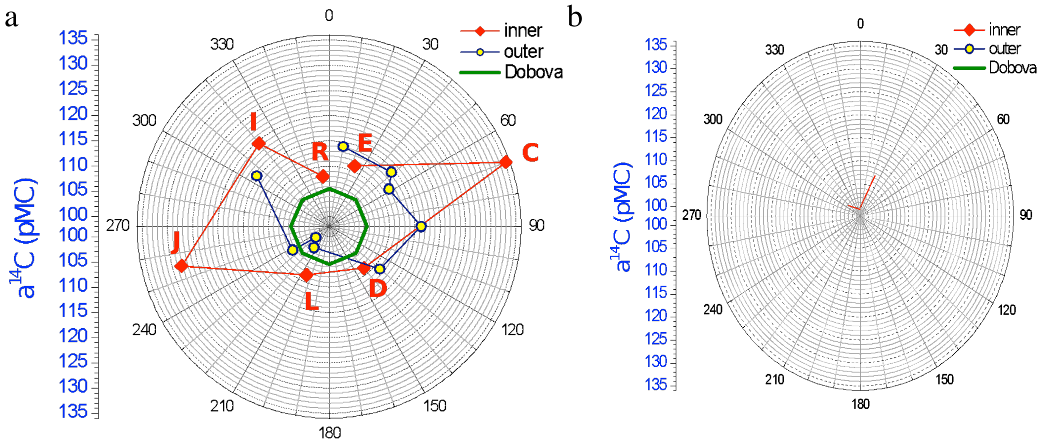

Figure 2. Examples of spatial distributions of relative specific activity of ^{14}C ($a^{14}C$) in inner and outer circles around NEK and at the control site Dobova. a) summer sampling 2009, after refuelling outage in spring, b) summer sampling 2007, before refuelling outage in autumn 2007

Table 1 summarizes the average values of ^{14}C activities in the inner and the outer circles and at the control location. Separately are given the average values for the years with the spring refuelling outage (2006, 2009, 2012, 2015) and the other years with the autumn refuelling outage or without it. The results are expressed as $a^{14}C$, the relative specific ^{14}C activity, in units pMC (percent Modern Carbon), and as the specific activity $A^{14}C$ (in Bq/kgC), where the relation between them is 100 pMC = 226 Bq/kgC.

3. DOSE ESTIMATION

Effective annual dose E (Sv) to a representative person from the ingestion of food having ^{14}C specific activity $A^{14}C$ (Bq/kgC) is [3,7,12]

$$E = e \times A^{14}C \times m \tag{1}$$

where m (kg) is the annual mass of carbon taken into an organism by ingestion and e is the ICRP dose coefficient [12], $e = 5.8 \times 10^{-10}$ Sv/Bq. Therefore, for the effective dose estimation it is important to know the $A^{14}C$ of the food and the annual intake of food m. Usually, the dose estimation due to ingestion of a radionuclide requires a detailed knowledge of dietary habits of either "a representative person" or "the most exposed person". Obtaining the detailed and relevant consumption data for particular areas may not be a simple task. Luckily, the specific activity $A^{14}C$ in all types of foodstuff (however, limited to terrestrial plants and animals) is the same and reflects the atmospheric ^{14}C activity in the area during the period of formation of a specific food item. In our case, the relevant values are presented in table 1.

Table 1. Comparison of the average values of $a^{14}C$ and $A^{14}C$ in the inner circle and the outer circle around NEK, separately for the years with the spring refuelling outage, and other years, as well as at the control location Dobova

Locations	**$a^{14}C$ (pMC)**		**$A^{14}C$ (Bq/kgC)**	
	spring refuelling outage	**other years**	**spring refuelling outage**	**other years**
inner circle (<300 m)	114.3 ± 5.2	107.0 ± 2.6	258 ± 12	241.8 ± 5.9
outer circle (<600 m)	108.1 ± 4.1	105.5 ± 1.5	244.3 ± 9.3	238.4 ± 3.4
control site (12 km)	103.6 ± 1.0		234.1 ± 2.3	

The data about daily, monthly or annual intake of food and the proportions of the food originating from the vicinity of the nuclear power plant are necessary to be determined. As carbon is an essential component of the diet and is found in all foods, it is likely that the intake of ^{14}C is insensitive to changes in the dietary composition. The annual intake of food for an average adult in Slovenia is about 250 kg [13], the estimated fraction of carbon is about 30% [3], meaning that about 75 kg of carbon is consumed annually. This value is in accordance with the UNSCEAR estimation [3] for Europe based on the detailed diet analysis (annual intake of carbon about 72 kg). Therefore, we will take the annual intake of m = 75 kg of carbon as an input value in eq. (1).

4. SCENARIOS

In the previous sections we defined all the quantities that enter eq. (1), but there is still a problem of combination of food from various locations. We have shown that there is a difference in ^{14}C specific activity of food from various locations around NEK. The $A^{14}C$ values are neither spatially nor temporarily uniformly distributed. Moreover, the years with the refuelling outage period in spring show distinctly different distributions (Figure 2) and different mean values (table 1) than the other years.

An assessment model of the ingestion dose should be "fit for purpose, i.e., it should not exclude, or poorly represent, any process that is known or suspected of having an important influence on radionuclide behaviour, and equally, it should not attempt to include detail that is not relevant to either the spatial or temporal scale of the assessment" [3]. How to determine a scenario (a model of ingestion) that should satisfy these requirements and is based on our data? In addition, if a contribution of ^{14}C atmospheric releases from a nuclear facility has to be assessed, the effective dose has to be compared to the natural background dose because ^{14}C is present everywhere in nature. We have developed several scenarios that may represent the combination of food from the "clean-air" sites and from the vicinity of NEK. Some of them are very conservative, unlikely and unrealistic, and some of them may be realistic, but still rather conservative.

In **Scenario A** only food from the control location is consumed throughout the year. The result is a natural effective dose due to ingestion of ^{14}C. Based on our data (table 1 and annual intake of 75 kg of C), the annual effective dose for location Dobova is 10.18 μSv. The value is in accordance with the UNSCEAR estimate of 12 μSv from the naturally occurring ^{14}C [4]. Seasonal variations (intra-annual) and inter-annual variations of atmospheric A^{14}C result in the natural variations in the affective dose of 1%.

Scenario B assumes consumption of food from any location in the inner circle, and none from the outer circle or the control site. It is the most conservative and unlikely scenario since one cannot expect that throughout the whole year a person consumes only the plant products (apples) from the very restricted area of inner circle. The scenario B is presented here only as the hypothetic worst possible case. Here and in all the following scenarios one has to distinguish the years with the spring refuelling outage and other years (autumn refuelling outage or none).

In **Scenario C** food from both inner and outer circles is consumed and the average A^{14}C for all locations in both inner and outer circles is taken as an input value in eq. (1). In **Scenario D** half of the food comes from the vicinity of NEK (having average A^{14}C for all locations in both inner and outer circles) and the other half from the "clean-air" site represented here by the control site Dobova. The scenario D seems to be realistic, but not probable because of the availability of the foodstuff on the market from other parts of Slovenia and from all over the world. However, a proportion of local and imported food in not known at this point.

Therefore, we suggest a more realistic, although still rather conservative, **scenario E**, in which a representative member of the public uses a certain small fraction of food produced locally, and much larger fraction of food "imported" from other locations. All "other locations" in our approach are represented by the control location and the A^{14}C measured there. The "imported food" may have even slightly lower ^{14}C specific activity if grown in the vicinity of a large industrial centre. We assume that a representative inhabitant in the vicinity of NEK uses local food from any location in either outer or inner circle for 2 months in the year and during the remaining 10 months food from the control site. Such an assumption is an equivalent to the proportion of 2/12 = 17% and 10/12 = 83% of local and imported food consumed, respectively. We believe that this is a conservative realistic assumption, likely to be close to the actual situation.

Scenario F takes into account that an average inhabitant of Slovenia annually consumes 25 kg of apples [13,14], which can be approximated by 3 kg of carbon or about 4% of the total annual carbon intake. For the inhabitant of the vicinity of NEK we can conservatively assume that all the apples come from any location around NEK. We also assume that all other foodstuffs come from the control site.

Table 2 shows the estimated annual effective doses E (eq. (1)) obtained by these scenarios, as well as the relative difference to the natural effective dose from ^{14}C obtained at the "clean-air" site (Scenario A). As input A^{14}C data in eq. (1) the average values for the years with the spring refuelling outage and for the other years are taken (Table 1) with m = 75 kg annual intake of carbon. The representative "clean-air" site in our case is the control location Dobova. Relative differences d are calculated as

$$d = (E_X - E_A)/E_A \qquad (2)$$

where X represents the scenarios B – F.

Table 2. Annual effective dose (E) and relative difference (d) from the natural ^{14}C background (scenario A)

Scenario	refuelling outage	E (μSv)	d, relative difference (%)	comment
A – natural ^{14}C background		10.18 ± 0.10	0	natural variations 1 %
B – only inner circle	spring	11.2 ± 0.5	10.3	unlikely, unrealistic
	other	10.5 ± 0.3	3.3	
C – both inner and outer circles	spring	10.9 ± 0.3	7.3	unlikely, unrealistic
	other	10.5 ± 0.2	2.6	
D – 50% from both inner and outer circle, 50% from Dobova	spring	10.6 ± 0.2	3.7	realistic, but not probable
	other	10.32 ± 0.11	1.3	
E – 2 months from both inner and outer circles, 10 months from the control site	spring	10.31 ± 0.12	1.2	most realistic, still conservative
	other	10.23 ± 0.11	0.4	
F – 25 kg of apples from vicinity of NEK, other food from the control site	spring	10.22 ± 0.11	1.2	based on annual apple consumption rate estimate
	other	10.20 ± 0.11	0.4	

5. CONCLUDING REMARKS

The annual effective dose in Table 2 is calculated based on the annual intake of 250 kg of food, or 75 kg of carbon. A different estimate of total annual intake of food in Slovenia is 330 kg, resulting in about 100 kg of carbon intake, is used for the dose assessment in off-site radiological monitoring around NEK [14]. This certainly changes the absolute values of the effective dose, both at the control site (scenario A, 13.38 μSv) or in any other case (e.g., for the most unlikely scenario B – spring refuelling outage, 14.93 μSv). However, the relative difference d remains the same.

The estimates of the contribution of the atmospheric releases from NEK to the annual effective dose to a representative person due to ingestion of ^{14}C does not depend on the detailed knowledge of the diet, even not on the exact knowledge of the total carbon intake – it depends on the measured ^{14}C activities in the plants and on the scenario describing the proportion of food from various origins. Here we presented several scenarios of the food origin, from the most unlikely (consuming fruits the inner circle only throughout the year, scenario B) to a more realistic one. The refuelling outage period of NEK is 18 months, meaning that every third year the refuelling outage takes place in spring. The released CO_2 can be taken for photosynthesis during the vegetation period and we have therefore separately discussed years with the spring refuelling outage and years with the autumn refuelling outage or without it. In all scenarios B – F the effective dose in the year with the spring refuelling outage is higher than in other years, as it was expected from the ^{14}C specific activity data. If we consider only the most realistic scenario E, the increase in the annual effective dose is not higher than 0.13 μSv or < 1.2% relative to the natural ^{14}C background in the worst case of the years with the spring refuelling outage. Such an increase is comparable to the natural variations in the natural ^{14}C background effective dose (Table 2). For other years, the difference between the effective dose given by scenario E and the natural ^{14}C effective dose is lower that the annual (both intra-annual and inter-annual) variations of the natural effective dose due to ingestion of ^{14}C by food from the "clean-air" sites. Based on a comprehensive ^{14}C monitoring program in the period 2006 – 2016, we may conclude that the contribution of NEK to the effective dose to a representative person of the inhabitant from the vicinity of NEK due to ingestion ^{14}C via the food chain is negligible.

The uncertainties presented in Table 2 are derived from the uncertainties in the measured A^{14}C values. If the uncertainties of the proportions of the food from different origins are taken into account, the resulting uncertainties in the effective doses would be much higher for the scenario E. However, this will not change the main conclusion of the negligible influence of NEK to the effective dose due to ingestion of ^{14}C.

Similar results were obtained in some other environmental ^{14}C monitoring studies [15,16]. A negligible total effect, close to the statistical limit, of the NPP Jaslovske Bohunice (Slovakia) on the environmental ^{14}C activity was determined [15]. A negligible increase of the annual effective dose from ingestion of 1.3% was attributed to the effect of gaseous releases of French nuclear power plants [16].

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KAKO PROCIJENITI EFEKTIVNU DOZU ZBOG INGESTIJE ^{14}C

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SADRŽAJ

Sustavni monitoring specifične aktivnosti ^{14}C u atmosferskom CO_2 i u biološkim uzorcima iz neposredne okolice Nuklearne elektrane Krško (NEK) u Sloveniji provodi se od 2006. godine. Cilj monitoringa je odrediti prostornu raspodjelu specifične aktivnosti ^{14}C u neposrednoj okolici NEK-a, te njenu ovisnost o razdobljima promjene goriva koja se obavlja svakih 18 mjeseci. Na osnovu izmjerenih specifičnih aktivnosti ^{14}C može se procijeniti doprinos NEK-a efektivnoj dozi koju lokalno stanovništvo primi zbog konzumacije (ingestije) hrane porijeklom iz okolice NEK-a. Budući da je ugljik, pa time i ^{14}C, sastavni dio sve hrane, nije potrebno poznavati detaljne prehrambene navike lokalnog stanovništva. Međutim, potrebno je poznavati ili procijeniti udio hrane iz okolice NEK-a u ukupnoj prehrani. U ovom radu opisano je nekoliko scenarija za porijeklo hrane, počevši od vrlo konzervativnog i malo vjerojatnog scenarija konzumacije samo hrane iz neposredne okolice NEK-a do realističnijeg (iako još uvijek konzervativnog) scenarija od 17% hrane iz okolice NEK-a, a ostalih 83% s kontrolne točke na kojoj nije opažen utjecaj ^{14}C iz NEK-a. Malo povišenje specifične aktivnosti ^{14}C u biljkama iz neposredne okolice NEK-a, koje je opaženo tijekom monitoringa, ne utječe značajno na procjenu efektivne doze zbog ingestije ^{14}C, jer je razlika u odnosu na efektivnu dozu zbog ingestije "prirodnog" ^{14}C usporediva ili manja od prirodnih varijacija.