J Radioanal Nucl Chem (2012) 292:689–695
DOI 10.1007/s10967-011-1495-y

^{14}C studies in the vicinity of the Czech NPPs

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Received: 27 September 2011 / Published online: 22 October 2011

Abstract The Czech Republic has two nuclear power plants (NPPs) equipped with light water pressurized reactors (LWPR). Annual sampling of biota for ^{14}C activity monitoring by Nuclear Physics Institute in cooperation with the National Institute of Radiation Protection started in 2002. We present the results of biota monitoring covering two sampling periods 2002–2005 and 2007–2008. The considerable problem in the case of biota sampling for monitoring purpose is given by a relatively short period of biota accumulation for prevailing types of biota samples (leaves of deciduous trees or agricultural plants), which usually lasts from several weeks to 2 months. The short period of sample accumulation can also be partly overlapped by a service period of reactor outage in a given NPP. On the base of our several years' experiences we have changed a type of the sampled material to reduce variations of observed activities and to precise reference levels in the exposed and reference sites.

Keywords Radiocarbon in biota · Nuclear power plants · ^{14}C monitoring · Sampling material selection

Introduction

The Czech Republic has two nuclear power plants (NPPs) equipped by light water pressurized reactors (LWPR), Temelín and Dukovany, with the installed power output 2 × 1,000 MW and 4 × 440 MW, respectively. The monitoring in the surrounding environment of Czech nuclear power plants Dukovany and Temelín consists of routine determinations, performed by NPP's staff, and also extended sampling which is performed by research institutions [1–3].

Environmental compartments contain a mixture of two stable carbon isotopes (^{12}C and ^{13}C) and one radioactive isotope ^{14}C (radiocarbon). This radionuclide of global occurrence and a half-life of 5,730 year is partly of anthropogenic origin. In the nature, ^{14}C is produced by nuclear reactions generated by cosmic rays in the atmosphere [4, 5].

At present, the most significant artificial sources of radiocarbon in the environment are effluents from nuclear power facilities, even though it is a minor contribution in comparison with its natural production. Nevertheless, radiocarbon is responsible for dominant contribution to the collective effective dose from all radionuclides released by nuclear power plants (NPP) with light-water pressurized reactors (LWPR) during normal operation [6]. Part of ^{14}C is discharged by NPPs into the surrounding environment during normal operation as gas effluents, in the case of LWPR it reaches about 95% of released ^{14}C [7, 8]. Radiocarbon from the gas releases of the NPP can be captured in the surrounding or dissipated in the atmosphere depending on its chemical form. The stable chemical forms are hydrocarbons with prevailing $^{14}CH_4$ [7], which is not significantly captured in the vicinity of the NPP and contributes to the increase of the ^{14}C activity level more on a regional or global scale [9]. Abundance of $^{14}CO_2$ in airborne effluents from LWPR varies between 5 and 43% [7, 8, 10–13]. The biota in the surrounding of NPPs intakes $^{14}CO_2$, especially during: calm, rainfall, haze, or

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atmospheric inversion. Radiocarbon in the form of CO_2 is assimilated by plant photosynthesis and afterwards transferred also into the food chain [6].

In the last century, nuclear weapons tests were important sources of anthropogenic ^{14}C. Consequently, ^{14}C activity in the atmosphere of the Northern Hemisphere was double the natural level in 1963 [14, 15]. Since the nuclear moratorium on atmospheric nuclear bomb tests was signed in 1963, the ^{14}C concentration in the atmosphere has been decreasing due to its intensive transfer to oceanic and terrestrial carbon reservoirs [16–23]. Currently, ^{14}C activity in the atmosphere is gradually approaching the level that was seen before the nuclear age.

There is another significant anthropogenic influence on ^{14}C levels in the atmosphere and biosphere - the Suess effect [24]. This effect causes a decrease of the ^{14}C activity on global, regional, and local scales as a result of the dilution of the carbon isotopic mixture by fossil carbon [5, 17, 18, 25–33].

Time behavior of atmospheric $^{14}CO_2$ activity can be characterized by linear interannual decrease since the beginning of 1990s. This long-term trend is caused by global Suess effect. Seasonal fluctuations with minima during cold parts of year are amplified by more intensive local and regional Suess effect, what is evident from the time series of atmospheric $^{14}CO_2$, see Fig. 1 [22, 29, 34].

Materials and methods

Monitoring of ^{14}C surrounding the NPPs and in reference localities can be performed by two possible ways:

1. Monitoring of atmospheric air. This monitoring can be performed during whole year, without limitations given by vegetation period. Drawbacks of such monitoring are greater time and economy requirements and also limited number of monitoring facilities in fixed localities. Such monitoring can provide the samples with exactly known duration of sampling period [28, 34–39].
2. Monitoring of ^{14}C activity in biota [39–47]. Time interval of ^{14}C activity record depends on period of biomass accumulation in a given plant. Samples (parts of plants) should be selected with care to avoid contamination by biomass originated in previous years. During sampling, a great number of biota samples can be collected and the NPP surrounding area can be covered by dense network of sites, if necessary. Positions of sampling localities can be changed easily according to atmospheric dissipation conditions in the given year. Obviously, ^{14}C monitoring in biota is restricted on the part of vegetation period.

Annual sampling of biota and agricultural products for ^{14}C activity monitoring in the surrounding of both Czech NPPs and in reference areas was launched in 2002 by

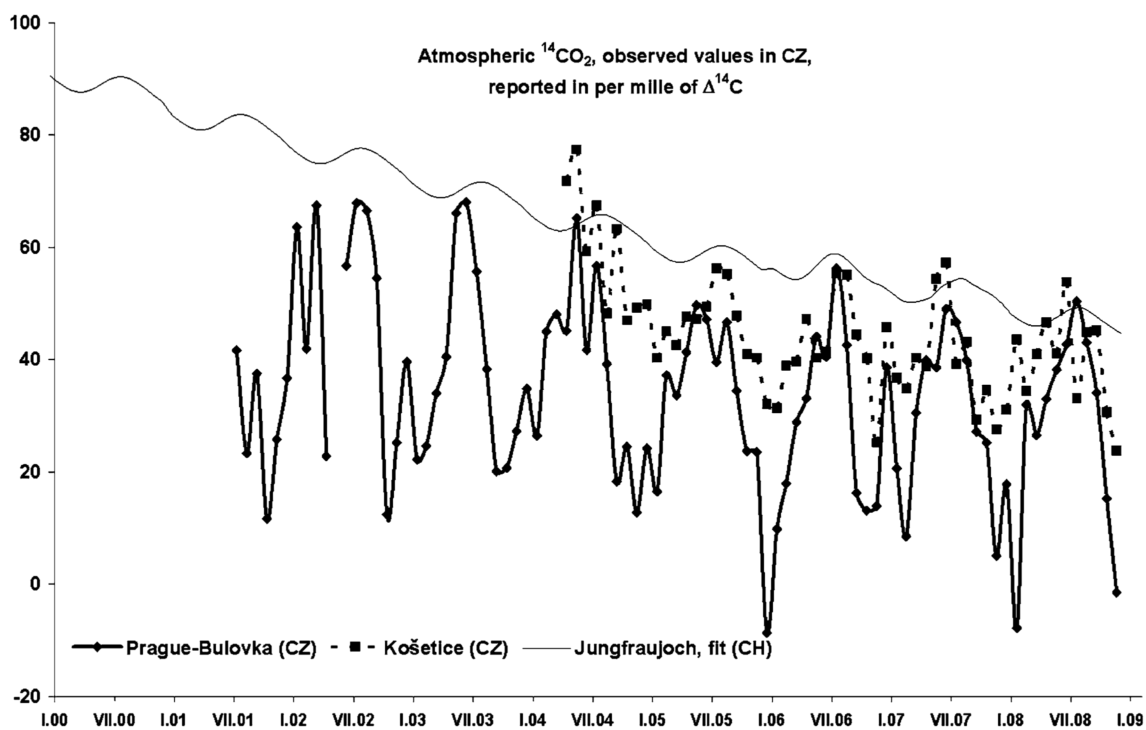

Fig. 1 Time series of atmospheric $^{14}CO_2$: Prague-Bulovka (*local and regional Suess effect*), Košetice (*regional Suess effect*), and Jungfraujoch (clean-air Alpine monitoring station, simulation). ^{14}C activities are reported (y-axis) in ‰ of Δ^{14}C [54]. Interannual decrease of ^{14}C activities is visible in all time series [22, 29, 34]

Nuclear Physics Institute AS CR in the cooperation with the National Institute of Radiation Protection.

Sampling of biota was performed in the sites of Temelín and Dukovany NPPs at the distance from 0.5 to 9 km during the period of 2002–2005. Prevailing part of biota samples were leaves of deciduous trees, analogically to published studies performed in the vicinity of other NPPs [41–47]. Leaves of *Sambucus nigra* (pipe tree) were preferred, because this tree is widespread in the Czech Republic and it can be easily identified. Agricultural plants (spikes of wheat and barley) made a smaller part of collected samples [1]. In the vicinity of NPPs Temelín and Dukovany several roads are situated and there are also some smaller cities and villages [1]. The influence of local Suess effect could be estimated/quantified with difficulties, due to the lack of data on local fuel combustion and density of surrounding traffic. To compare ^{14}C activities in the NPPs surroundings with relevant ^{14}C activity level in the environment two types of areas with different load of Suess effect were selected. It can be supposed that actual size of Suess effect influencing NPPs surrounding will be in the interval demarked by these two types of reference areas [1, 29]. (A) Localities, in greater distances from fossil carbon sources, where only small local influence of Suess effect was supposed (Košetice, Kleť, Sudoměřice u Bechyně, Krokočín). (B) Localities where extended local Suess effect influence can be expected (bordering parts of Prague).

Since 2007 the type of sampling material has been changed to above ground parts of *Urtica dioica* (stinging nettle) collected in November. Stinging nettle or common nettle is a herbaceous perennial flowering plant, native to Europe, Asia, northern Africa, and North America, and is the best-known member of the nettle genus *Urtica* [48]. This perennial plant has only exiguous annual biomass supply and thin root system. Vegetation period of this plant is between the end of March and beginning of November in the Central Europe. Besides, this herb forms internodia with relatively uniform rate. On each internodium biomass cumulates from its origin till the end of vegetation period [49]. Hence, more detailed time resolution of ^{14}C activity advancement could be achieved during vegetation period using this plant, if necessary.

Dusty biota samples were washed with 10% HCl and distilled water, dried (105 °C) and homogenized. Washing with diluted HCl was suspended if presence of dust on a sample surface was not evident. Dried samples were combusted and the produced CO_2 was purified. In the NPI AS CR a routine of sample processing based on benzene synthesis was followed [1, 29, 30, 50–52]. ^{14}C activity was measured by liquid scintillation spectrometer Quantulus 1,220 in 3 mL low-background Teflon vials. Total counting time was about 3,000 min per sample. Benzene distributed by Sigma-Aldrich (spectrophotometric grade) was used as a blank sample. Calibration was performed using oxalic acid NIST (NBS) HOX II, SRM 4990-C [53]. Resulting activities were reported in ‰ of Δ^{14}C following Stuiver-Polach convention [54]. Combined uncertainties of observed values (in the interval 6.1–8.1 ‰ of Δ^{14}C) include the individual uncertainties of measured sample, blank sample, calibration, quenching corrections, and uncertainty of the δ^{13}C value [55].

Results

During the period of 2002–2005, 77 biota samples for ^{14}C analyses were collected in the vicinity of NPPs Dukovany (EDU) and Temelín (ETE). Likewise, 30 samples were collected in reference areas influenced with slight (A) and extended (B) local Suess effect. Basic statistical parameters of results (EDU, ETE, A, B) are reported in Table 1. Standard deviations of couples EDU-B, ETE-A, and ETE-B are equal on the base of F tests performed (Fischer-Snedecor test). In the next step results from each type of area were compared utilizing t test (student test, unpaired, probability of first kind of observation error 5%), see Table 2.

In the period of 2007–2008 samples of *Urtica dioica* were collected in the surrounding of $EDU_{7,8}$ and $ETE_{7,8}$ (18 samples, adjacent – in distances from 0.7 to 1.2 km, without preferred direction). In the comparison with previous sampling, distances were reduced to achieve areas with expected maximal possible ^{14}C activity surplus [41, 43, 44]. Reference samples (11 samples, 10–20 km from a given NPP, without preferred direction) from more distant surrounding were collected in localities with estimated densities of roads and traffic loads similar to those in the

Table 1 Basic statistical parameters of biota samples collected in the vicinity of NPPs Dukovany (EDU) and Temelín (ETE) and in reference localities with slight (*A*) and extended (*B*) local Suess effect

	EDU	ETE	Ref. localities A	Ref. localities B
Average	60.1	61.0	56.2	47.4
Median	58.3	60.4	56.2	45.7
Standard deviation	13.2	9.0	6.5	7.3
Variation	173	81	42.1	53.5
Number of observations	27	50	21	9
Observed maximum	95.9	84.4	67.9	58.7
Observed minimum	39.8	41.7	44.0	38.0

Sampling period 2002–2005. Activities are reported in ‰ of Δ^{14}C [54]

Table 2 Comparisons of activities of observed results from each type area (group of the data), values of T reported in table: T_o (observed) and T_c (critical); probability of first kind of observation error 5%

Couple compared	T_o	T_c	t test, commentary
A-B	2.621	2.201	$T_o > T_c \Rightarrow$ difference is significant
EDU-A	1.479	2.024	$T_o < T_c \Rightarrow$ difference is not significant
EDU-B	2.507	2.037	$T_o > T_c \Rightarrow$ difference is significant
ETE-A	2.336	1.996	$T_o > T_c \Rightarrow$ difference is significant
ETE-B	3.913	2.004	$T_o > T_c \Rightarrow$ difference is significant
EDU-ETE	0.305	1.993	$T_o < T_c \Rightarrow$ difference is not significant

Sampling period 2002–2005

Table 3 Basic statistical parameters of biota samples collected in the direct neighborhood of NPPs Dukovany ($EDU_{7,8}$), Temelín ($ETE_{7,8}$) and in corresponding reference localities (refEDU, refETE) with similar estimated local Suess effect, sampling period 2007–2008

	$EDU_{7,8}$	ref.EDU	$ETE_{7,8}$	ref. ETE
Average	37.9	33.2	35.4	30.0
Median	35.5	33.4	34.5	30.3
Standard deviation	7.2	4.7	4.6	2.4
Variation	52.2	22.1	21.3	5.9
Number of observations	10	6	8	5
Observed maximum	52.7	39.7	42.9	32.5
Observed minimum	30.0	26.6	28.8	27.0

Activities are reported in ‰ of $\Delta^{14}C$ [54]

Table 4 Comparisons of activities of observed results from each type area (group of the data), values of T reported in table: T_o (observed) and T_c (critical); probability of first kind of observation error 5%, sampling period 2007–2008

Couple compared	T_o	T_c	t test, commentary
$EDU_{7,8}$-ETE	0.868	2.120	$T_o < T_c \Rightarrow$ difference is not significant
$EDU_{7,8}$-refEDU	1.422	2.145	$T_o < T_c \Rightarrow$ difference is not significant
$ETE_{7,8}$-refETE	2.359	2.201	$T_o > T_c \Rightarrow$ difference is significant
refEDU-refETE	1.352	2.262	$T_o < T_c \Rightarrow$ difference is not significant

direct vicinity of NPPs [41]. Basic statistical parameters of results ($EDU_{7,8}$, $ETE_{7,8}$, refEDU, refETE) are reported in Table 3. Variances for all couples $EDU_{7,8}$-$ETE_{7,8}$, $EDU_{7,8}$-refEDU, $ETE_{7,8}$-refETE, and refEDU-refETE do not differ significantly on the base of F-tests performed. In the next step results from each type of area were compared utilizing t-test (unpaired, probability of first kind of observation error 5%), see Table 4.

Discussion

For the period of 2002–2005, statistical evaluation of the results confirmed significantly greater ^{14}C activity level in biota from both NPPs surroundings in comparison with reference area B (greater load from fossil fuel combustion – bordering parts of Prague), see Table 2. Significant differences were found also between reference localities A and B. Likewise, the difference of ^{14}C activity level between biota from NPP Temelín vicinity and biota from reference area A (minor load from local fossil fuel combustion) was found to be statistically significant also. In the point of view of local Suess effect, it can be supposed that relevant reference ^{14}C activity level for NPPs surroundings with relatively traffic-loaded roads is situated in the interval between reference areas A and B.

Observed values of ^{14}C activity for each type of locality are charged with relatively great variations, probably caused by local Suess effect from surrounding roads, for samples from NPPs surroundings namely. Another reason of fluctuation can be caused also by relatively short time interval of biomass accumulation in leaves of deciduous trees (about four or 5 weeks in April and May). At this part of year, the activity of atmospheric $^{14}CO_2$ changes rather quickly, what is visible also from Fig. 1 [17, 22, 25, 28, 29, 56]. The exact duration of the period of plant biomass accumulation in tree leaves is depending on the local microclimatic conditions (atmospheric precipitations, soil moisture, and sunlight exposure). Hence, the local microclimatic differences can cause small time shift of the period for dominant atmospheric $^{14}CO_2$ intake by tree leaves and thus also differences in resulted ^{14}C activities. Additional reason of ^{14}C activity variations in NPPs surroundings is given by relatively greater variation of distances from NPPs stacks (below 9 km) in certain sample collection sites. It can be supposed, that ^{14}C activity surplus in biota at distances exceeding 10 km is minimal [41]. Potentially most influenced zones around NPPs can be probably found in the distance up to 2 km, on the basis of ^{14}C dissipation model [41, 57]. Direct results of ^{14}C of biota monitoring performed in the vicinity of NPPs with boiling water reactors (BWR)[1] also confirm similar distances from stacks for maximal surplus of ^{14}C activity in biota [43, 44, 58].

In the case of nettle samples (2007–2008), observed activities of ^{14}C seem to be more uniform, namely due to longer biomass accumulation period, probably reducing influence of microclimatic variations. In comparison with previous types of samples, reduced activities of nettle samples are evident for each group. Such difference is

[1] Compared with LWPR releases from BWR contain considerably greater percentage of $^{14}CO_2$, above 90% [6, 8]. This chemical form of released ^{14}C can be assimilated by plant photosynthesis and hence greater ^{14}C activity excess can be observed in the surrounding biota of NPPs with BWR [43, 44].

given particularly by extended period of biomass accumulation (end of March till beginning of November). During colder seasons, the activity of atmospheric $^{14}CO_2$ decreases as a result of regional and local Suess effect, see Fig. 1 [5, 17, 18, 25–29]. Likewise, the difference between mean values of ^{14}C activities observed in periods of 2002–2005 and 2007–2008 is partly given also by the interannual decrease of atmospheric $^{14}CO_2$ activity. This interannual decrease of atmospheric $^{14}CO_2$ activity is estimated to be about 5 ‰ of $\Delta^{14}C$ [34]. Due to large variations of activities, the interannual decrease was not observed (insignificant statistically) when leaves of deciduous trees were used in the period of 2002–2005.

In the case of nettle sampling, difference of several per mille in each group of samples seems to occur between sampling years 2007 and 2008, including reference samples. The corresponding reference areas were selected in the distances 10–20 km from a given NPP, supposing minimal ^{14}C activity surplus at such distances [41, 43, 44]. Utilizing Student test (unpaired, probability of first kind of observation error 5%), a significant difference between vicinity of ETE ($ETE_{7,8}$) and corresponding reference localities (ref.ETE) was found. Difference of 5.4 ‰ of $\Delta^{14}C$ ($ETE_{7,8}$ - ref.ETE) for nettle sampling is in a good agreement with observed difference 4.8 ‰ of $\Delta^{14}C$ (ETE - A, significant for 5% probability of the first kind of observation error) obtained on the base of previous sampling campaign.

Application of nettle plants as a sampling material seems to reduce variations of ^{14}C activities caused by microclimatic differences. Likewise, this sampling material is widespread and can assure relatively long period of ^{14}C activity record.

Due to formation of nettle internodia with relatively uniform rate, time resolution of ^{14}C activity changes (in the surrounding air) in a given year with precision of several weeks can be achieved, if necessary. To validate such possibility, small outdoor experiments are intended in 2011. The group of nettle plants will be partly sealed by a polyethylene bag and exposed to $^{14}CO_2$ (about several kBq) for several hours in June or July. During November the plants will be collected and the material will be cut node to node. The response curve of ^{14}C activity vertical distribution (in dependence on internodium number) will be compared with the data of a model of accidental release of $^{14}CO_2$.

Conclusion

Results of ^{14}C monitoring in the biota of the surroundings of NPPs Dukovany and Temelín were briefly reported. On the base of biota monitoring around Czech NPPs a small surplus of ^{14}C activity level in the close surrounding of NPP Temelín was observed for both sampling campaigns (2002–2005, utilizing mainly leaves of deciduous trees, and 2007–2008, sampling of nettle plants). In comparison with reference areas minimally locally loaded by fossil fuel combustion (A), the mean numeric value of the excess is 4.8 ‰ of $\Delta^{14}C$, (*t* test, unequal variations, probability of first kind of observation error 5%). Applying a new sampling routine in the period of 2007–2008, the surplus of 5.4 ‰ of $\Delta^{14}C$ was observed in the neighborhood of ETE compared with reference sites in distance 10–20 km from the NPP (*t* test, equal variations, probability of first kind of observation error 5%). Nevertheless, this excess of ^{14}C activity was not significant for 1% probability of first kind of observation error. Low/insignificant ^{14}C activity surplus in biota around these NPPs with LWPRs is given by small abundance of $^{14}CO_2$ form in gas releases of ^{14}C from both NPPs (about 5%). Other ^{14}C chemical forms, with prevailing $^{14}CH_4$, are not responsible for radiocarbon intake by surrounding biota.

During campaign in 2007–2008, samples of nettle plants were collected. With regard to the extended period of biomass accumulation by this plant, resulting activities in each group of samples seems to be charged by smaller variations compared to classical types of biota samples. Several week period of biomass accumulation in tree leaves can be also in the partial coincidence with the several weeks of service period of reactor outage in a given NPP (^{14}C releases are minimal at such case).

Campaign performed with a new type of sampling material covers only 2 years and two localities in Czech Republic. Due to a widespread occurrence of the nettle plants, a validation of this unconventional sample material suitability also in other geographical positions and slightly different climatic conditions would be a benefit.

Acknowledgments This work was funded by internal grant of the Nuclear Physics Institute AS CR (No. AV0Z 10480505) and by National Radiation Protection Institute (grants No. JC 03/2006 and JC 05/2008). The authors acknowledge for the ^{13}C determinations of nettle samples performed by Dr. István Futó from the Institute of Nuclear Research HAS (ATOMKI) in Debrecen, Hungary.

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