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MONITORING OF ATMOSPHERIC EXCESS ^{14}C AROUND PAKS NUCLEAR POWER PLANT, HUNGARY

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ABSTRACT. The activity of radiocarbon in $^{14}CO_2$ and $^{14}C_nH_m$ chemical forms is measured in the vicinity of Paks nuclear power plant (NPP), Hungary, by sampling environmental air. Four differential sampling units at different sites collected samples less than 2 km away from the 100-m-high stacks of Paks NPP, and for reference a sampler was operated at a station ~30 km away from Paks NPP. We present the results of continuous observations at the 5 stations covering the time span from 2000 to 2005. The samples have been analyzed by a proportional counting technique. During a cleaning tank incident at unit 2 of Paks NPP in April 2003, a significant release of radioactive isotopes took place from the damaged fuel assemblies, and gaseous products escaped through the chimney. We evaluate the possible short- and long-term impact of this incident on the ^{14}C content of the atmosphere in the surroundings of Paks NPP. Comparing our $^{14}CO_2$ measurements with data sets from Jungfraujoch and Schauinsland, as well as from Košetice (Czech Republic), we demonstrate that the incident had no definite influence on the ^{14}C content of the atmosphere.

INTRODUCTION

Atmospheric radiocarbon distributions have been extensively investigated in the Northern Hemisphere, Southern Hemisphere, and in the tropics for a number of decades (e.g. Manning and Melhuish 1994; Meijer et al. 1995; Rozanski et al. 1995; Levin et al. 2003; Levin and Kromer 2004). Systematic global observations of $^{14}CO_2$ in the troposphere were made during and after atmospheric nuclear weapon tests in the 1950s and 1960s by several laboratories (e.g. Hertelendi and Csongor 1982; Nydal and Lövseth 1983; Levin et al. 1985; Manning et al. 1990). At present, the monitoring of ^{14}C in regions adjacent to nuclear power plants (NPPs) or sites of land-filled radioactive wastes has a growing importance in determining the frequency and activity of anthropogenic ^{14}C released into the environment. Levin and Kromer (2004) published annual mean $^{14}CO_2$ values of 148.3–102.5‰ from Schauinsland from 1990 to 1996, which can be accepted as typical Northern Hemisphere atmospheric ^{14}C concentrations. Vokal and Kobal (1997) measured $\Delta^{14}C$ values (180–200‰) in tree rings grown in 1990–1996 located 1.5 km from Krško NPP. Excess of ^{14}C has been found in tree rings and air samples around Barsebäck NPP, Sweden (~140‰ in 1996, Stenström et al. 1998). In the environment of Ignalina NPP (Republic of Belarus), slightly elevated levels of ^{14}C (120–150 pMC) were observed in vegetation and waters of Lake Drisvyaty that are connected by canals with the NPP (Mikhajlov et al. 1999, 2004).

The estimated ^{14}C release into the environment by NPPs with WWER (water-water energetic reactor) light-water pressurized reactors (PWRs) operating under standard conditions is about 1 TBq GW_e^{-1} yr^{-1} (Otlet et al. 1992). In PWRs, ^{14}C is produced by neutron activation of oxides in the fuel, moderators, and coolant in the $^{17}O(n,\alpha)^{14}C$ reaction and in the $^{14}N(n,p)^{14}C$ reaction for nitrogen in the fuel, moderator, and coolant (Davis 1979). The mean values of ^{14}C production rates for PWRs are 0.57 TBq GW_e^{-1} yr^{-1} in fuel, 0.97 TBq GW_e^{-1} yr^{-1} in structural materials, and 0.19 TBq GW_e^{-1} yr^{-1} in the heat carrier (Kunz 1985). Lately, smaller normalized releases have been reported for PWRs of different constructions (Van der Stricht and Janssens 2001; Roussel-Debet et al. 2006).

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The ^{14}C produced in the coolant is released into the environment mainly via the stack, with ^{14}C activity discharged in liquid and solid wastes being <5% of the gaseous discharge.

In Paks NPP, 4 WWER-440/213 units are operating with a total nominal power of 1860 MW (electrical). Paks NPP is located in the middle of Hungary (46°35′N, 18°51′E), 120 km from Budapest (Figure 1a). More than 40% of the electrical energy generated in Hungary is produced here. ^{14}C monitoring around Paks NPP involves the determination of ^{14}C concentrations in the ventilation stacks, environmental air, nuclear waste, and primary water (Hertelendi et al. 1989b; Uchrin et al. 1989, 1992, 1998; Veres et al. 1995). ^{14}C concentration of air in the stacks of Paks NPP varied between 80 and 200 Bq m^{-3}. The average normalized yearly discharge rates for 1988–1993 were 0.74 TBq GW_e^{-1} yr^{-1} for hydrocarbons and 0.06 TBq GW_e^{-1} yr^{-1} for CO_2 (Veres et al. 1995). Due to technological improvements, the total annual discharge rate decreased from 0.92 TBq/yr in 2000 (Paks NPP 2003) to 0.69 TBq/yr in 2005. During a cleaning process of fuel assemblies in April 2003, a cooling deficiency occurred inside the cleaning tank, resulting in damage to the fuel assemblies inside the tank (http://www.npp.hu; Doma et al. 2005; Vajda et al. 2005). Consequently, significant amounts of noble gases were released (Hózer and Szabó 2005; Molnár et al. 2005) but, according to the measurements in the stacks' air (Paks NPP 2004), the damage had no effect on the total ^{14}C emissions.

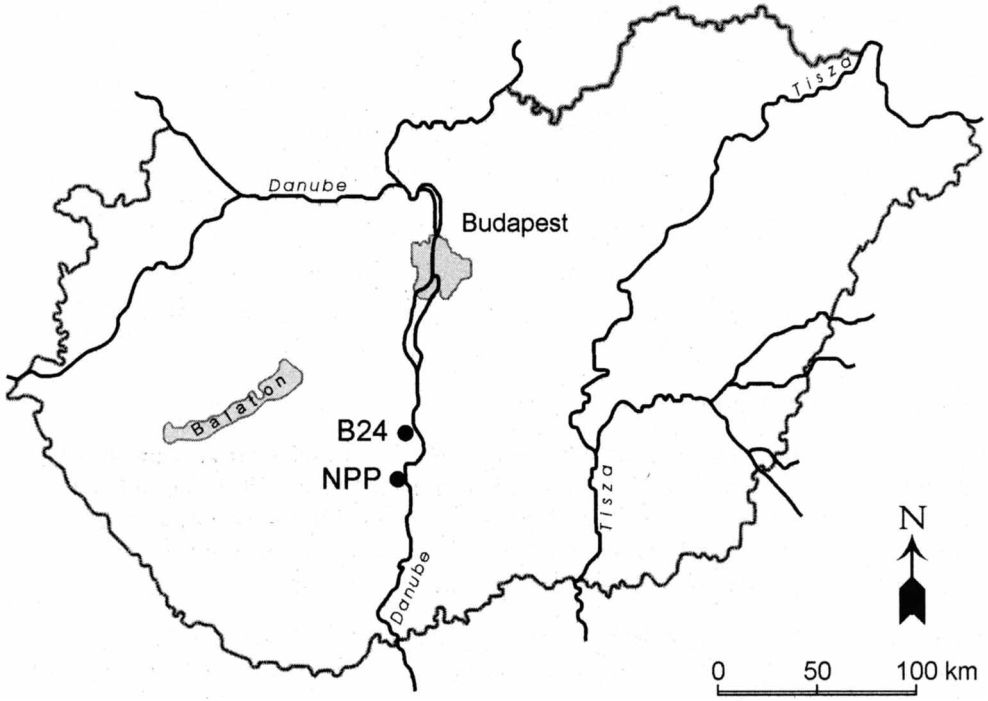

Figure 1a Location of Paks NPP and the B24 background monitoring station

We present here the $\Delta^{14}C$ values measured in the vicinity of the Paks NPP during the time span from 2000 to 2005. The investigation was designed to:

- Determine whether station B24 can be used as a "background" site unaffected by Paks NPP discharges (where background is defined as the regional atmospheric ^{14}C activity due to natural production, atmospheric nuclear weapons testing, and dilution due to fossil-fuel combustion).

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- Assess ^{14}C concentrations in the territories adjacent to Paks NPP under regular operating conditions, and
- Investigate the effect of the mentioned minor incident in 2003 on the environment.

Samples were collected on a monthly basis from 2000 to 2005 using differential ^{14}C samplers located about 2 m above ground level. Sample sites are the following: Site B24 (46°47′N, 18°57′E), 30 km from Paks NPP (Figure 1a); and sites A1, A4, A6, and A8 located <2 km from the 100-m-high stacks of Paks NPP (Figure 1b).

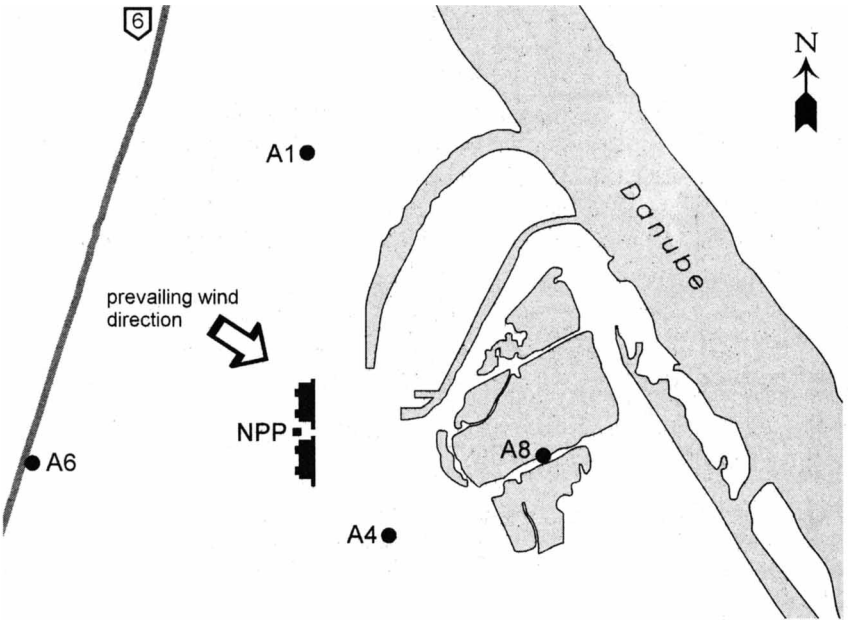

Figure 1b The location of the atmospheric monitoring stations (A-type) in the vicinity of Paks NPP.

MATERIALS AND METHODS

Differential ^{14}C samplers (Figure 2) have been developed to obtain integrated samples for measuring ^{14}C in chemical forms, such as CO_2, CH_4, and other hydrocarbons. ^{14}C is trapped in the form of CO_2 for all chemical species since hydrocarbons are oxidized by a Pt-Pd catalyst heated at 450 °C. The CO_2 is trapped in bubblers filled with 500 mL of 3M NaOH solution. The flow rate and the temperature of the catalyst were controlled by the sampling unit.

Samples were collected over a period of 4 weeks at a stable flow rate of ~10 L/hr. Discharged CO_2 (99.9%) was absorbed in a 3M NaOH solution using a specially designed bubbler-type trap. The conversion efficiency of hydrocarbons into CO_2 form exceeded 99.7%. A detailed description of the sampling devices can be found in Uchrin and Hertelendi (1992).

CO_2 was extracted from the samples by adding H_2SO_4 to the NaOH solution. Liberated CO_2 gas was purified over charcoal, then cryogenically separated from other gases using liquid N_2 (Csongor et al. 1982). Sample ^{14}C activity was measured using a gas proportional counting method (Csongor and Hertelendi 1986; Hertelendi et al. 1989a). The standard deviation applying this method of a single modern $\Delta^{14}C$ measurement was 5‰ (Hertelendi 1990). $\delta^{13}C_{PDB}$ (‰) values were measured using a

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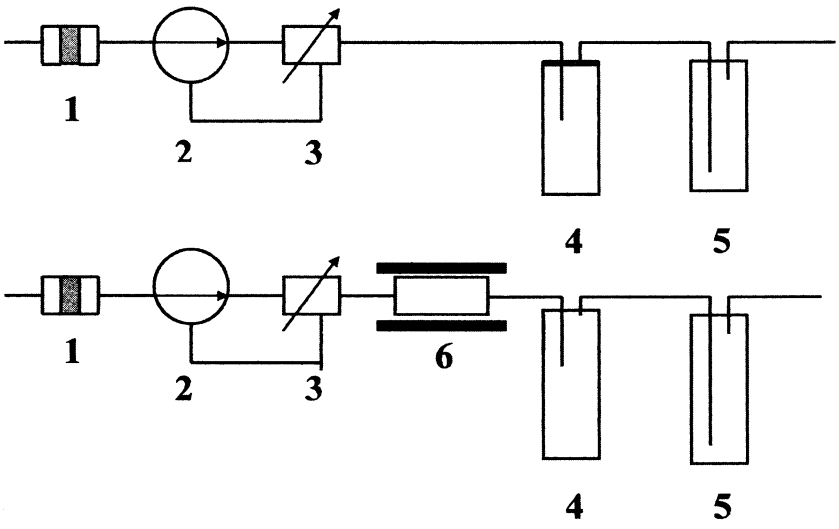

Figure 2 Layout of ^{14}C sampler developed and used for monitoring ^{14}C discharges in the form of CO_2 and C_nH_m: 1) filter; 2) air pump; 3) flow controller; 4) puffer; 5) bubbler with 500 mL of 3M NaOH; 6) converter (Pt-Pd catalyst heated at 450 °C).

stable isotope mass spectrometer developed in the ATOMKI (Hertelendi et al. 1987) until 2002 and by a ThermoFinnigan Delta^{PLUS} XP mass spectrometer from 2002 to 2005. $\delta^{13}C$-corrected $\Delta^{14}C$ data are given relative to NBS oxalic acid activity, corrected for decay (Stuiver and Polach 1977).

RESULTS AND DISCUSSION

$\Delta^{14}C$ values were determined on both CO_2+C_nH_m (hereafter "sum fraction") and CO_2 fractions of the monthly collected samples. Results are presented in the Appendix.

B24 is a true continental station situated in an agricultural plain ~100 m asl. To judge whether B24 is influenced or not by Paks NPP, we compared the $\Delta^{14}C$ time series at B24 with "clean air" $^{14}CO_2$ data from Jungfraujoch (Levin and Kromer 2004) and from an agricultural/woody site at Košetice, Czech Republic (Světlík et al. 2006). We calculated the linear prediction for Jungfraujoch data to obtain reference background values until the end of 2005 (Figure 3). The uncertainties of the predicted values for Jungfraujoch correspond to the maximal differences between the linear trend and observed values during the period from January 2000 to July 2003 (data for Jungfraujoch are not available after July 2003). The slopes of the linear fits on data at the 2 continental stations (B24 and Košetice) and at Jungfraujoch are equal within the uncertainties. Subtracting the data measured at the 2 continental stations from the predicted data for Jungfraujoch, we obtained detrended $\Delta^{14}C$ time series for these localities. The curves plotted in Figure 3 show the weighted smooth fits on these time series. The time response of the fits allows the fit curves to follow intra-annual variations (the smoothing factor is 5 months). The $\Delta^{14}C$ time series for B24 and Košetice are close and embody similar seasonal trends, especially the annual averages for 2005, which are in very good agreement. Monitoring at Košetice started in 2004, and so a longer time series does not exist from this locality. The B24 values for both fractions show relatively high seasonality typical for continental stations, but the $\Delta^{14}C$ values are generally lower than the Jungfraujoch, by an average of 13 ± 1‰ for the CO_2 and 5 ± 1‰ for the sum fraction. The differences between the Jungfraujoch and B24 data are greater during the autumn/winter period; this is probably due to the extended Suess effect during the heating season and/or atmospheric inversion effects.

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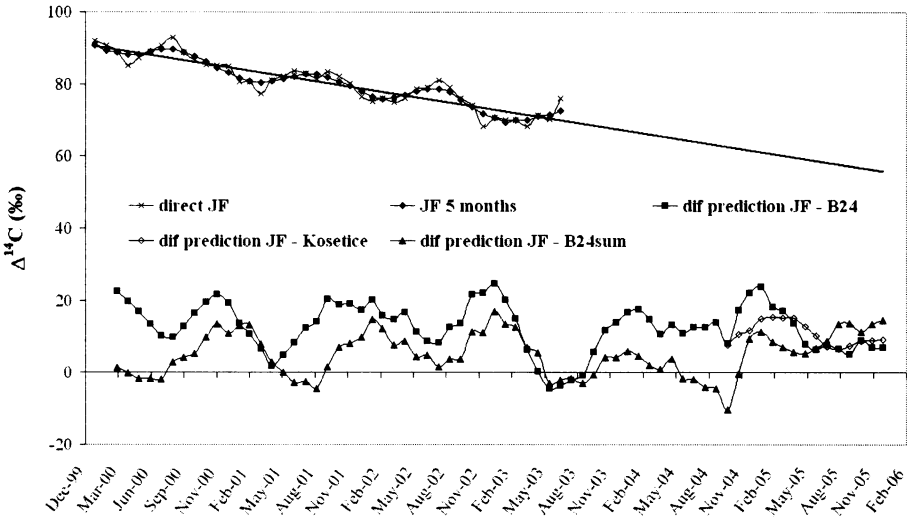

Figure 3 $\Delta^{14}C$ time series observed at B24 and Košetice (Czech Republic) subtracted from predicted (linear trend) "clean air" data from Jungfraujoch (Switzerland).

The curve fitted to the $\Delta^{14}C$ values measured on the sum fraction at B24 has a seasonal trend similar to the curve fitted to the $\Delta^{14}CO_2$ values, but the ^{14}C activities of the sum fractions are significantly higher by an average of 6.9‰ (Table 1, *t* test, $\alpha = 0.05$, $t_{obs} > t_{crit} \Rightarrow \mu_1 \neq \mu_2$). The difference between the CO_2 and sum fractions at B24 may show the effect of Paks NPP, but the effect cannot be firmly stated. From the point of view of reference background of atmospheric ^{14}C in hydrocarbon chemical forms, there are only sporadic data regarding the ^{14}C content of atmospheric methane (e.g. Steele et al. 1987; Wahlen et al. 1989; Quay et al. 1991; Brown et al. 1994; Eisma et al. 1995). The ^{14}C-free methane released by the production and use of fossil fuels lowers the ^{14}C concentration, whereas the emission of $^{14}CH_4$ by NPPs raises it. As a result, the ^{14}C concentration of atmospheric methane rose by ~1.4 pMC/yr in 1987–1989 (Quay et al. 1991) and by 0.8 pMC/yr in 1988–1992 (Brown et al. 1994). Levin et al. (1992) observed an increase of 0.9 ± 0.6 pMC in atmospheric $^{14}CH_4$ at the maritime site (Izaña) from 1987 to 1989 and estimated a global mean atmospheric $^{14}CH_4$ increase rate for 1988 of 3 pMC/yr. In the light of these findings, the average surplus of 6.9‰ $\Delta^{14}CH_4$ in B24 might be a regional or global and not a local phenomenon. The regional Suess effect observed for $^{14}CO_2$ in B24 and Košetice stations is similar.

Table 1 Statistical comparison (Goulden 1956) of $\Delta^{14}C$ values (CO_2 and sum of carbon fractions) observed in station B24.

	Sum of fractions	$^{14}CO_2$
Mean (μ)	65.6	58.7
Variance (σ^2)	248.3	174.4
Minimum	32.0	25.1
Maximum	97.2	88.3
Number of observations	69	69
2-sample paired *t* test for equal means, $\alpha = 0.05$.		
t_{obs}		5.0196
Critical value for *t* (bilateral)		1.9955

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Atmospheric monitoring stations A1, A4, A6, and A8 collected air samples from 2 m above the surface level in the vicinity of Paks NPP. $\Delta^{14}C$ values were measured both on the sum and the CO_2 fractions (Figure 4a,b). These curves show the same seasonality as the reference background at B24. Large fluctuations are observed at A4 (Figure 4a); these fluctuations are mainly caused by variations in the activity of emissions from Paks NPP and, to a lesser extent, changes in wind strength and direction. High summer peaks are probably due to significant emissions from Paks NPP during the shutdown events that occur before the yearly maintenance of the reactor units.

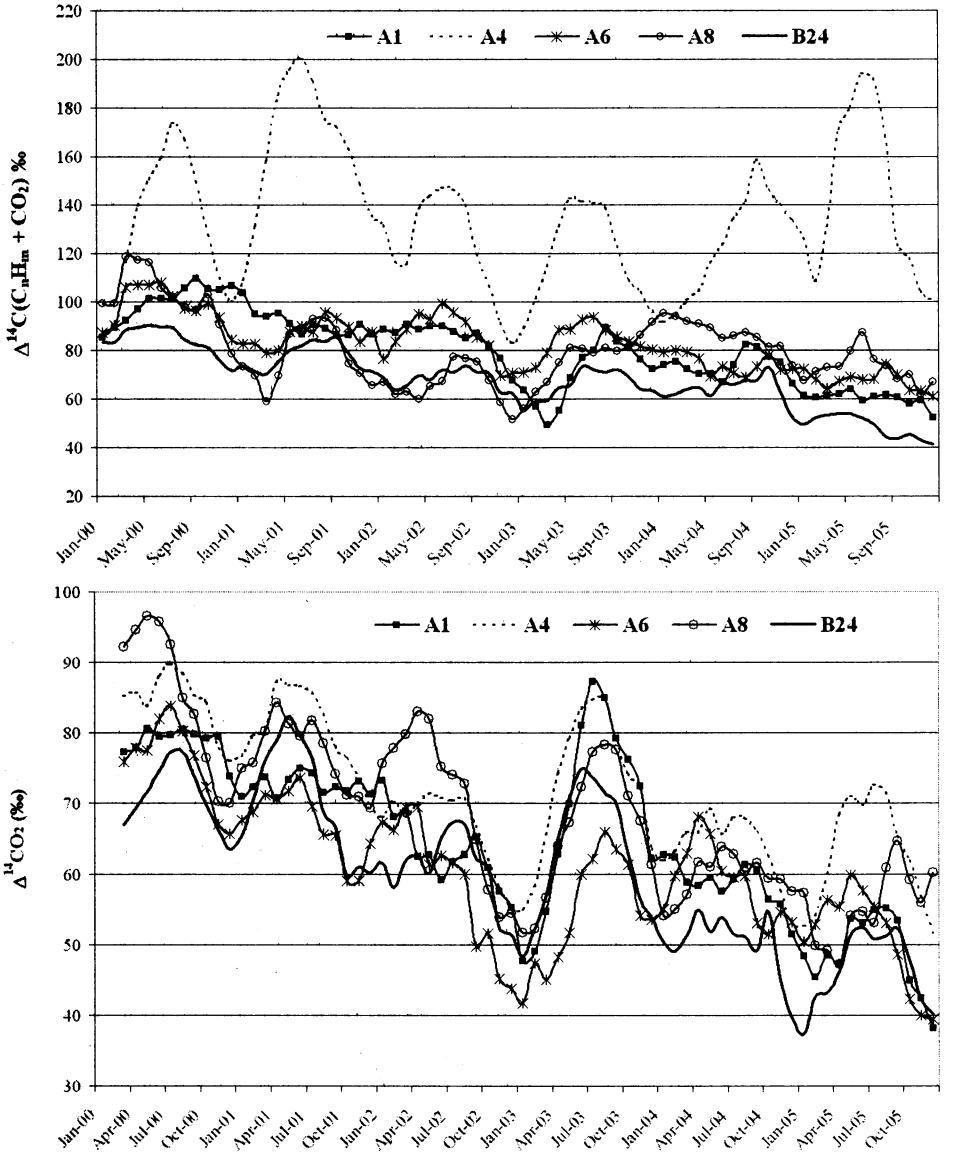

Figure 4 $\Delta^{14}C$ values measured on the (a) C_nH_m+CO_2 and (b) CO_2 fractions, smoothing over 5 months

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To assess the effect of Paks NPP, we subtracted background values obtained from site B24 from the measured data from each monitoring station. This gives a value for "excess $\Delta^{14}C$" (Figure 5a,b), which is due to Paks NPP emissions. The smoothing factor for the excess $\Delta^{14}C$ values was 1 yr, allowing the curve to follow slow, trend-like changes, but preventing it from following the intra-annual variations.

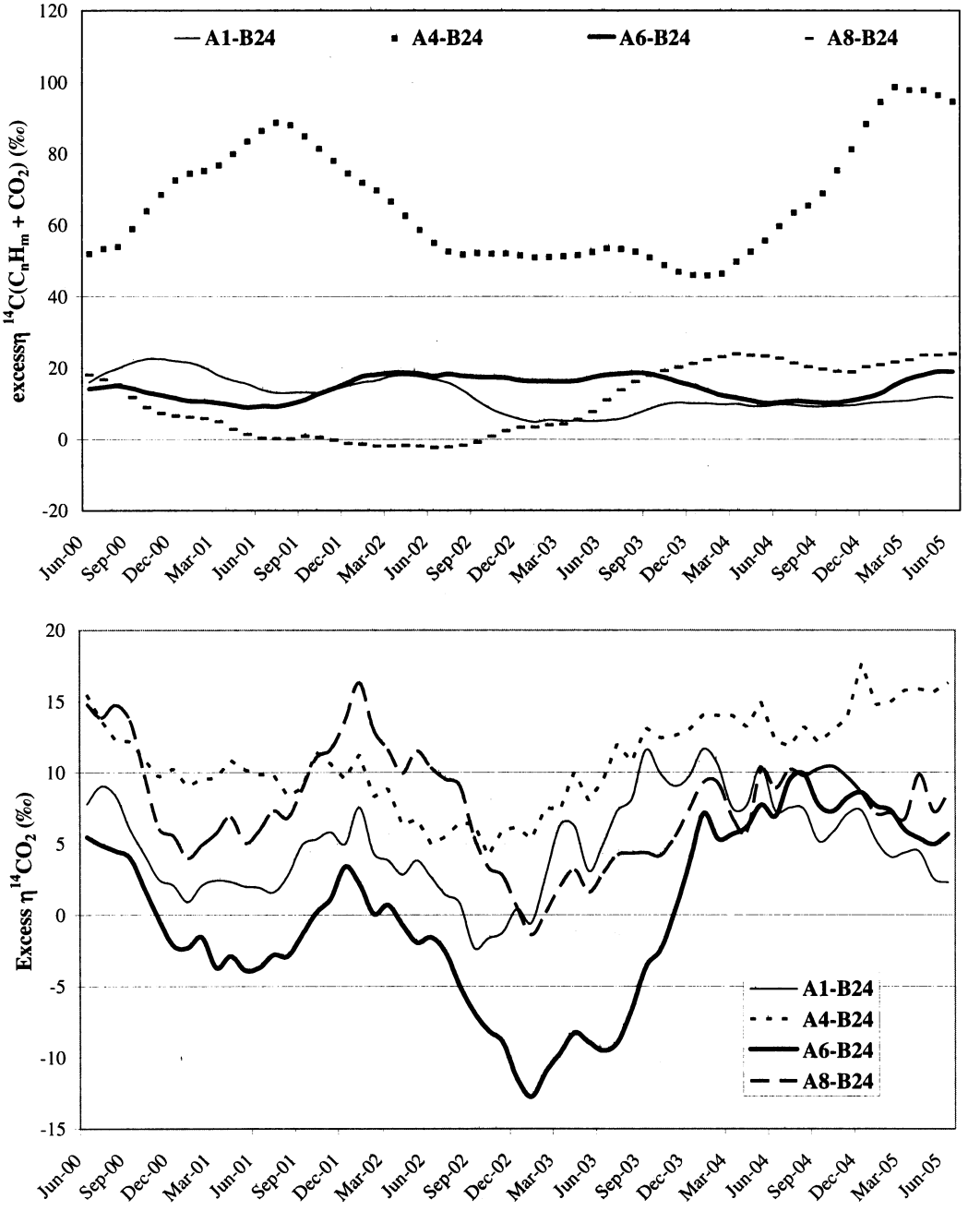

Figure 5 Excess $\Delta^{14}C$ measured on the (a) C_nH_m+CO_2 and (b) CO_2 fractions, smoothing over a year

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For PWR-type NPPs, the dominant chemical forms of ^{14}C emission are hydrocarbons; carbon dioxide plays a minor role (e.g. Veres et al. 1995; Vokal and Kobal 1997; Uchrin et al. 1998). The curves fitted to the sum fractions show that contributions from Paks NPP to the sum fraction are significantly higher than its contribution to the CO_2 fraction. From Table 2, it can be seen that ~95% of the ^{14}C emitted from Paks NPP is in the form of C_nH_m.

Table 2 Total ^{14}C emission (TBq/yr) from Paks NPP in 2000–2005 (Paks NPP 2001–2006).

Year	CO_2	C_nH_m	Total	% C_nH_m
2000	0.05	0.87	0.92	95
2001	0.05	0.76	0.81	94
2002	0.05	0.68	0.73	93
2003	0.05	0.63	0.68	93
2004	0.05	0.66	0.71	93
2005	0.03	0.58	0.61	95

The curves fitted to the excess $\Delta^{14}C$ values measured in the sum fractions (Figure 5a) are rather smooth. The largest intra-annual fluctuations and highest values were observed at the nearest station, A4, situated 870 m from Paks NPP in the path of the prevailing wind, with excess $\Delta^{14}C$ values between 40–100‰ in the sum fraction. At the other stations, the excess $\Delta^{14}C$ values in the sum fraction fluctuate between 0–20‰, and the curves are smooth. Stations A6 and A8 are about the same distance from the NPP (~1400 m), and the farthest one, A1, is about 2 km from the plant. There is no significant difference between the data measured at the A1 and A8 stations.

The curves fitted to the excess $\Delta^{14}C$ values measured in the CO_2 fraction fluctuate strongly in spite of the smoothing (Figure 5b), possibly caused by the changing emission ratio of C_nH_m:CO_2. The highest excess $\Delta^{14}CO_2$ values (5–15‰) were measured at station A4. After the cleaning tank incident in 2003, a 5–10‰ rise in $\Delta^{14}CO_2$(‰) values was observed at all stations. During the remediation activities, the $\Delta^{14}CO_2$(‰) values decreased slowly in 2004 and ceased by the second half of 2005.

Station A6 was installed several meters away from the main road (No.6). In 2000, an excess $\Delta^{14}CO_2$ of 2–5‰ could be observed here. With the growing traffic, the inactive CO_2 emission (Suess effect) exceeded the influence of Paks NPP at this station in 2001–2003. In 2004 and in the first half of 2005, the $\Delta^{14}CO_2$ exceeded the background level by 5–10‰, but in the second half of 2005 it decreased again below the value measured at station B24 (Figure 5b).

Figure 6 shows average $\Delta^{14}C$(‰) values plotted versus distance from Paks NPP, and it is clear that the effect of atmospheric $\Delta^{14}C$ discharges decreases with distance from Paks NPP. Weighted least-square fits were calculated for both series (using the functions $\Delta^{14}C[sum] = 45 \times X^{-3} + 65.6$ and $\Delta^{14}CO_2 = 10 \times X^{-1} + 58.7$, where X is the distance from the plant in km) and show that at surface level the ^{14}C activity of the sum fraction drops more quickly with distance than that of the CO_2 fraction. Reasons for this may be 1) differences in density ($\rho[CH_4]$ 0.7g/L, ρ[air] 1.3g/L, $\rho[CO_2]$ 1.9g/L), 2) dilution/mixing, and 3) oxidation of methane. As a result, the influence of $^{14}C_nH_m$ emissions on the environment decreases rapidly with distance from Paks NPP and is negligible at a distance of 2.5 km under normal operating conditions.

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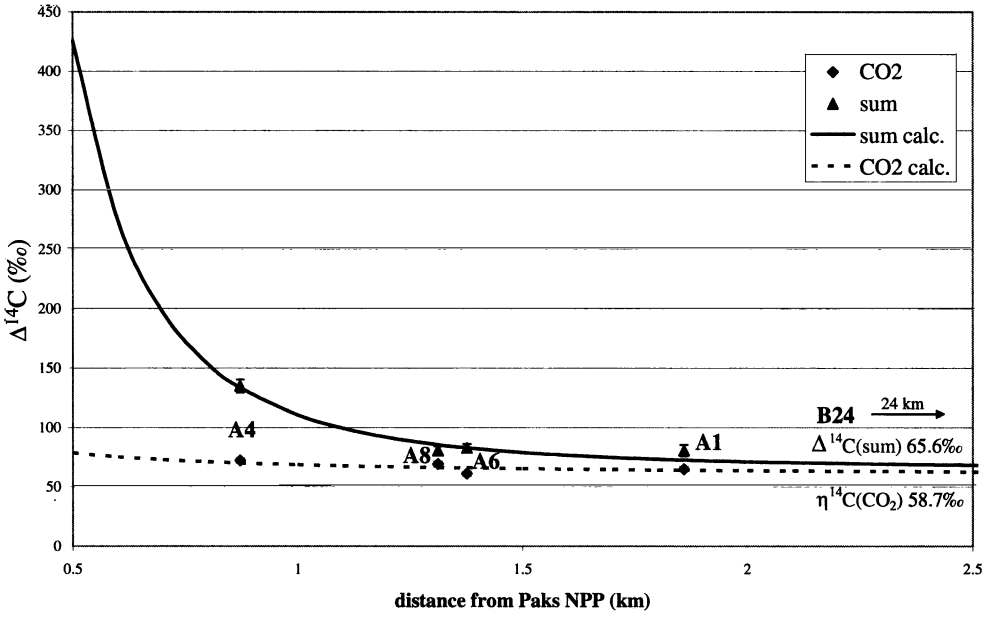

Figure 6 Calculated averages of $\Delta^{14}C$ values versus distance of station from Paks NPP

CONCLUSIONS

The $\Delta^{14}CO_2$ time series for the reference station B24 and Košetice are similar and embody similar trends. The B24 values show relatively high seasonality typical of continental stations and have $\Delta^{14}C$ (‰) values that are generally lower than the Jungfraujoch values, by an average of 13‰ for the CO_2 fraction and 5‰ for the sum fraction. The differences between the Jungfraujoch and B24 data are greater during the autumn/winter period probably due to a combination of the Suess effect, which increases during hot weather (summer), and atmospheric inversion effects, which have greater occurrence during cold weather (winter). The difference between the sum and CO_2 fractions at B24 may show the effect of Paks NPP, but this cannot be firmly stated because the ~7‰ surplus in the sum fraction may be due to global/regional and not local phenomena.

Highest ^{14}C values were measured at site A4, located <1 km from Paks NPP. Here, the excess $\Delta^{14}C$ in the sum fraction fluctuated between 40–100‰, and the $\Delta^{14}C$ CO_2 values fluctuated between 5 and 15‰. At the other stations (1.5–2 km from the plant), the excess $\Delta^{14}C$ was on average 13.0 ± 2.6‰. The influence of the ^{14}C discharge in the environment decreases rapidly with distance from the source, and under normal operating conditions the effect of Paks NPP is negligible at a distance of 2.5 km. After the tank-cleaning incident in 2003, a 5–10‰ rise of $\Delta^{14}CO_2$ was observed at all stations; this decreased slowly in 2004 and ceased by the second half of 2005.

Although the local and regional impact of ^{14}C discharged from NPPs with light-water pressurized reactors during their operating lifetime is low compared with that of naturally produced ^{14}C, regular monitoring of ^{14}C discharges is necessary as significant variations may occur between individual NPPs and in the ratio of the chemical forms in which ^{14}C is released.

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APPENDIX

Appendix $\Delta^{14}C$ (‰) values measured at the atmosphere monitoring stations in the vicinity of Paks NPP, Hungary. Combined uncertainty (Curie 1995) of $\Delta^{14}C$ determination is about 5‰.

	A1		A4		A6		A8		B24	
	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2
Jan-00	85.4	74.5	102.3	88.4	87.4	74.5	99.3	77.5	83.4	68.5
Feb-00	89.4	63.5	—	101.3	90.4	86.4	99.3	79.5	83.4	68.5
Mar-00	95.4	87.4	140.1	69.5	96.3	63.5	154.0	93.4	91.4	64.5
Apr-May-00	95.4	80.4	111.3	83.4	128.2	77.5	120.2	105.3	91.4	66.5
Jun-Jul-00	110.3	77.5	195.7	91.4	91.4	84.4	93.4	89.4	88.4	80.4
Aug-Sep-00	95.4	81.4	182.8	91.4	100.3	86.4	101.3	89.4	89.4	79.5
Oct-00	117.2	84.4	77.5	76.5	103.3	59.6	100.3	67.5	65.5	66.5
Nov-00	130.1	74.5	109.3	76.5	86.4	67.5	87.4	77.5	79.5	63.5
Dec-00	89.4	74.5	86.4	85.4	104.3	61.6	124.2	58.6	80.4	59.6
Jan-01	93.2	82.3	93.2	59.4	74.4	60.4	40.6	58.4	64.4	61.4
Feb-01	104.2	53.5	133.0	82.3	53.5	79.3	40.6	88.3	68.4	66.4
Mar-01	102.2	70.4	123.1	80.3	95.2	69.4	73.4	92.2	76.3	76.3
Apr-May-01	85.3	81.3	221.4	90.3	86.3	73.4	70.4	81.3	66.4	88.3
Jun-01	100.2	67.4	227.4	93.2	78.3	57.5	93.2	78.3	89.3	75.3
Jul-01	82.3	66.4	186.2	79.3	92.2	85.3	126.0	73.4	91.2	83.3
Aug-01	80.3	78.3	144.9	80.3	107.1	78.3	84.3	83.3	87.3	62.4
Sep-01	105.2	78.3	174.7	85.3	74.4	53.5	90.3	92.2	78.3	72.4
Oct-01	76.3	67.4	143.9	75.3	127.0	53.5	73.4	65.4	72.4	50.5
Nov-01	87.3	71.4	209.5	69.4	66.4	56.5	67.4	56.5	97.2	64.4
Dec-01	83.3	63.4	142.9	71.4	73.4	53.5	58.4	58.4	58.4	49.5
Jan-02	101.1	85.2	72.2	66.3	77.2	78.2	64.3	82.2	57.3	68.3
Feb-02	86.1	69.3	108.0	66.3	95.1	80.2	64.3	84.2	71.2	68.3
Mar-02	85.2	77.2	124.9	67.3	71.2	68.3	80.2	97.1	61.3	57.3
Apr-02	81.2	45.4	132.9	80.2	101.1	51.4	42.4	67.3	69.3	47.4
May-02	99.1	67.3	140.8	60.3	98.1	—	64.3	68.3	69.3	68.3
Jun-02	92.1	53.4	185.5	76.2	110.0	78.2	49.4	98.1	77.2	71.2
Jul-02	92.1	70.2	133.8	73.2	84.2	47.4	90.1	79.2	63.3	56.3
Aug-02	86.1	59.3	141.8	64.3	104.0	73.2	91.1	63.3	81.2	82.2
Sep-02	69.3	58.3	130.9	78.2	82.2	47.4	91.1	61.3	64.3	58.3
Oct-02	85.2	72.2	108.0	61.3	77.2	53.4	62.3	62.3	83.2	67.3
Nov-02	102.0	66.3	84.2	57.3	79.2	27.5	41.4	58.3	63.3	47.4
Dec-02	66.3	48.4	68.3	49.4	67.3	56.3	52.4	43.4	61.3	48.4
Jan-03	60.2	42.3	66.1	44.3	42.3	41.3	46.3	44.3	41.3	40.3
Feb-03	24.4	47.3	89.0	63.2	87.0	40.3	55.2	64.2	62.2	53.2
Mar-03	66.1	34.3	136.7	61.2	79.1	43.3	85.0	48.3	52.2	52.2
Apr-03	68.1	73.1	146.6	73.1	89.0	55.2	76.1	61.2	78.1	67.1
May-03	28.4	76.1	134.7	86.0	97.9	45.3	72.1	65.1	63.2	72.1
Jun-Jul-03	90.0	83.0	147.6	88.0	89.0	57.2	86.0	81.0	67.1	81.0
Aug-03	107.9	90.0	129.7	82.0	98.9	85.0	83.0	73.1	92.0	73.1
Sep-03	84.0	103.9	145.6	79.1	93.0	66.1	67.1	86.0	71.1	61.2
Oct-03	75.1	65.1	122.8	87.0	71.1	64.2	82.0	70.1	58.2	61.2
Nov-03	62.2	54.2	69.1	59.2	76.1	45.3	—	—	71.1	73.1

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Appendix $\Delta^{14}C$ (‰) values measured at the atmosphere monitoring stations in the vicinity of Paks NPP, Hungary. Combined uncertainty (Curie 1995) of $\Delta^{14}C$ determination is about 5‰. *(Continued)*

	A1		A4		A6		A8		B24	
	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2	CO_2+ C_nH_m	CO_2
Dec-03	75.1	68.1	76.1	63.2	79.1	46.3	95.0	55.2	53.2	45.3
Jan-04	84.6	71.0	105.6	—	89.8	48.8	101.1	58.9	65.8	41.0
Feb-04	63.9	52.1	106.9	45.1	85.9	63.3	88.6	60.9	68.4	48.2
Mar-04	84.9	68.0	101.1	80.0	65.2	71.3	96.3	41.5	47.0	44.3
Apr-04	67.9	52.4	85.6	64.8	79.5	68.8	88.5	58.5	75.3	66.6
May-04	60.5	50.3	103.8	73.7	76.5	62.4	85.4	65.7	64.3	56.8
Jun-04	74.0	68.8	125.4	—	—	74.8	96.3	81.7	68.8	58.8
Jul-04	63.8	57.8	165.5	58.3	55.8	51.2	80.3	57.8	51.4	32.6
Aug-04	68.2	58.2	136.5	66.1	81.0	45.0	73.9	55.6	71.8	54.6
Sep-Oct-04	102.9	61.0	139.9	73.8	69.5	63.8	93.3	53.3	73.5	54.8
Nov-04	69.3	64.8	210.4	57.7	91.4	41.6	85.2	88.0	—	—
Dec-04	43.1	37.0	102.8	41.6	78.9	43.3	61.0	46.5	—	—
Jan-05	57.6	54.7	107.8	41.6	50.5	60.5	76.0	55.5	41.0	25.1
Feb-05	59.4	39.6	105.7	50.1	71.1	56.9	53.1	44.7	42.6	39.0
Mar-05	77.4	45.5	104.3	71.9	69.3	49.8	63.7	51.7	65.9	47.9
Apr-05	66.0	50.2	123.2	65.0	69.8	53.7	102.8	50.8	59.4	58.5
May-05	47.0	52.5	213.9	71.1	61.8	60.2	69.7	43.2	57.9	45.5
Jun-05	59.5	48.4	308.1	84.1	64.7	56.3	77.2	45.7	44.2	39.8
Jul-05	70.4	71.7	150.4	62.6	79.9	79.0	84.8	79.1	42.1	65.2
Aug-05	54.2	42.5	173.7	66.6	64.3	39.3	102.7	54.6	55.9	53.0
Sep-05	73.9	59.3	114.4	78.3	70.5	41.9	47.0	42.8	47.4	50.4
Oct-05	50.2	53.6	94.4	66.4	91.5	48.5	56.1	82.3	32.0	47.8
Nov-05	54.4	39.8	84.8	53.0	44.0	34.4	51.9	—	40.6	45.2
Dec-05	—	29.6	122.7	45.2	47.2	47.4	92.9	56.9	51.5	42.7

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