^{14}C LEVELS IN THE VICINITY OF TWO SWEDISH NUCLEAR POWER PLANTS AND AT TWO "CLEAN-AIR" SITES IN SOUTHERNMOST SWEDEN

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ABSTRACT. ^{14}C is one of the radionuclides that are produced to different degrees by neutron-induced reactions in all types of nuclear reactors. Part of the ^{14}C created is continuously released into the surrounding environment during normal operation as airborne effluents in various chemical forms (such as CO_2, CO and hydrocarbons) through the ventilation system of the plant. Because of the biological importance of carbon and the long half-life of ^{14}C, it is of interest to measure the releases and their incorporation into living material. We report here on the ^{14}C activity concentrations in annual tree rings and the air around two Swedish nuclear power plants, as well as the background ^{14}C activity levels from two reference sites in southern Sweden from 1973–1996. We used both accelerator mass spectrometry (AMS) and decay counting in the investigation.

INTRODUCTION

Besides the natural cosmic-ray induced production of radiocarbon in the atmosphere, ^{14}C also results from certain anthropogenic activities. The main man-made contributions come from nuclear weapons testing (the bomb effect) and the nuclear power industry. A considerable amount of artificially produced ^{14}C is also used as a tracer in research and industry.

The most common power reactor types are pressurized water reactors (PWRs) and boiling water reactors (BWRs). Both designs are light-water cooled and moderated. The main part of the ^{14}C discharged from these reactors during normal operation is produced through neutron-induced reactions in the cooling water and is released as airborne effluents through the ventilation system of the plant to the surrounding environment (Kunz 1985; Stenström *et al.* 1995). The dominant chemical form of the ^{14}C released is carbon dioxide from BWRs and hydrocarbons from PWRs (Hertelendi, Uchrin and Ormai 1989; Kunz 1985; Snellman 1988; Stenström *et al.* 1996a; Uchrin *et al.* 1992; Wahlen and Kunz 1978a,b; Winkelmann and Vogl 1984). It has been estimated that ^{14}C is the radionuclide that dominates the committed effective doses to humans from the nuclear power industry (UNSCEAR 1988). Therefore, it is important to determine ^{14}C releases from nuclear reactors and their associated fuel-reprocessing and waste-management facilities.

We present here some results of a study on ^{14}C emissions from Swedish nuclear power plants and the resulting excess ^{14}C in air and vegetation in the area surrounding the power plants. The measurements are part of an extensive investigation of ^{14}C releases from some Swedish nuclear power plants (Stenström *et al.* 1995, 1996a,b). In these studies, the release rates through the stacks and the chemical form of the releases were determined by accelerator mass spectrometry (AMS). We also present background ^{14}C activity levels from two reference sites in southern Sweden from 1973–1996.

METHODS

Annual tree rings are cut out from bore cores (yielding 50–100 mg of wood per year). The samples are pretreated with a acid-alkali-acid (AAA) and cellulose extraction (see, *e.g.*, Olsson and Possnert 1992). The AAA pretreatment, which removes humic acids and extracts resin from the wood, is accomplished by boiling the sample in HCl (3M) for 30 min, washing, then boiling in NaOH (1M)

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Proceedings of the 16th International ^{14}C Conference, edited by W. G. Mook and J. van der Plicht
RADIOCARBON, Vol. 40, No. 1, 1998, P. 433–438

for 30 min, washing and boiling the sample again in HCl (3M) for another 30 min prior to a final washing. The cellulose is extracted by heating the pretreated sample in distilled water (16 mL), then adding 100 mg $NaClO_2$ (80%) and 0.15 mL HCl (1M). After 0.5 h, 1.5 h, 2.5 h and 3.5 h, portions of 200 mg $NaClO_2$ and 0.30 mL HCl (1M) are added. After 1 h more, the remaining cellulose is washed, filtered and dried. Air samples (*ca.* 40 liters per sample) are collected in balloons. In the laboratory, the air sample is drawn through a drying agent of $CaCl_2$ prior to absorption of the CO_2 in CO_2-free Ascarite® (NaOH on a solid support).

CO_2 is formed in a vacuum system by combustion of the organic material (*ca.* 10 mg) mixed with CuO (*ca.* 300 mg), or by hydrolysis (using phosphoric acid) of the Ascarite® from the air samples. Finally, elemental carbon is produced by reducing the CO_2, mixed with hydrogen gas, over an iron catalyst at 650°C (Vogel *et al.* 1984; Stenström 1995).

The Lund AMS system, described in detail elsewhere (Wiebert 1995; Stenström *et al.* 1997), is based on a 3 MV Pelletron tandem accelerator. The injector is equipped with a 20-position Cesium-sputtering ion source (Håkansson *et al.* 1996). To eliminate scattered particles, we installed a new dipole magnet during the autumn of 1996. The introduction of the dipole, placed between the Wien filter and the particle detector, has reduced the machine background from *ca.* 2% to <0.3% of modern standard. The ^{14}C yield from a CuO combusted anthracite sample which has been processed as described above corresponds to *ca.* 40,000 BP. The $\Delta^{14}C$ values below were obtained according to Stuiver and Polach (1977) with modifications for AMS described in Donahue, Linick and Jull (1990).

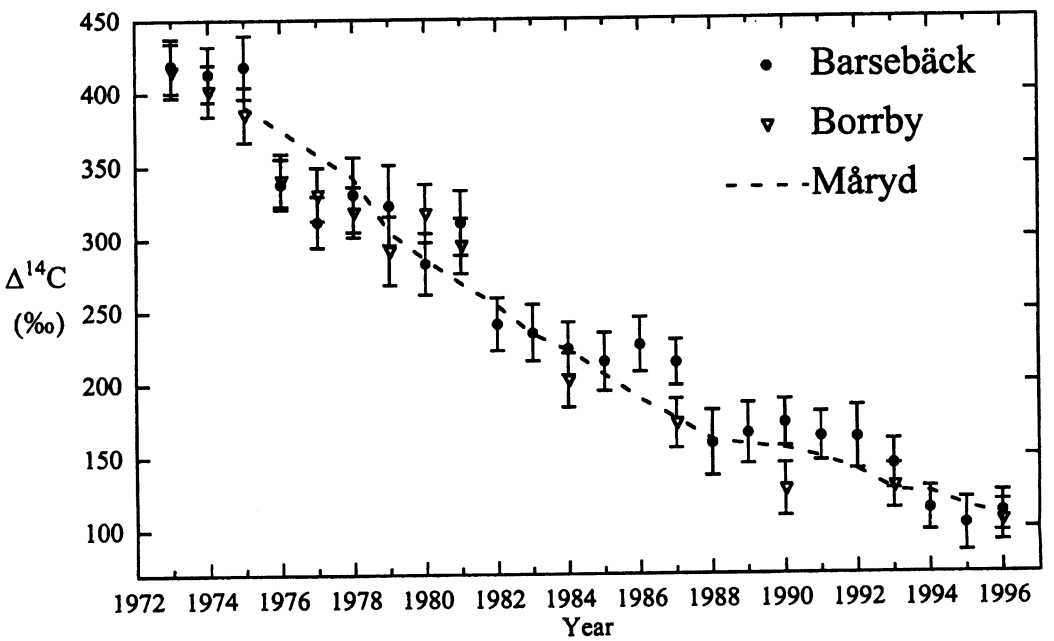

Fig. 1. $\Delta^{14}C$ values of annual tree rings of *Pinus* at 3 km from the Barsebäck nuclear power plant and from the Borrby "clean air" site. The dotted line interpolates the $\Delta^{14}C$ values of *Juncus* from Måryd listed in Table 1. The Barsebäck and Borrby values were obtained by AMS, and the Måryd values by decay counting.

https://doi.org/10.1017/S0033822200018312 Published online by Cambridge University Press

Results and Discussion

We used AMS to measure the ^{14}C content of annual rings from a tree of *Pinus* located 3 km east-northeast (frequent downwind direction) of the Barsebäck nuclear power plant (southwest Scania, 2 BWRs, each 600 MW). The results, covering the period 1973–1996, are shown in Figure 1. To measure the excess ^{14}C caused by the nuclear power plant, we also analyzed annual tree rings from the same species at a distant reference point (Borrby, southeast Scania, 90 km from Barsebäck). No measurements of $\delta^{13}C$ values for the tree rings were made ($\delta^{13}C = -25‰$ was used for all tree-ring samples). For further comparison, we examined $\Delta^{14}C$ values covering the same period from *Juncus* grown at the shore of a small pond in Måryd (south Scania, 30 km east of Barsebäck). The data from Måryd, listed in Table 1, were obtained by decay counting at the Radiocarbon Dating Laboratory in Lund (Håkansson 1968, 1986). Some of those values were published previously (Håkansson 1977, 1987, 1988). The $\Delta^{14}C$ values from Borrby and Måryd agree within the error limits (1 σ), with one exception. The $\delta^{13}C$ corrections for the data from Måryd were all obtained from mass spectrometry measurements. Måryd (15 km east of Lund, 25 km northeast of Malmö), situated fairly far from any densely populated area, should provide a good estimate of the mean atmospheric ^{14}C level for southern Sweden during the growing season. In Table 1, the Måryd values are compared to other European background data (see, *e.g.*, Meijer *et al.* 1994).

Table 1. $\Delta^{14}C$ Values from the Måryd "Clean Air" Site in Southernmost Sweden Compared with Published Data of Background ^{14}C Activity from Different Locations in Europe

			$\Delta^{14}C$ (‰)		
Year	Måryd (*Juncus*)	Abisko (air)*	Georgia (wine)†	Schauinsland (CO_2)‡	U.K. (plant material)§
1975	390 ± 6		368		
1976			332		
1977			298	333	
1978	342 ± 6		345	324	
1979	305 ± 6		295	295	341
1980	286 ± 6		279	267	327
1981	269 ± 6		272		292
1982	254 ± 6		266	239	279
1983	234 ± 6		223	225	
1984	223 ± 6		216	205	
1985	206 ± 6	212#	209	194	
1986	189 ± 6	205#	190	183	
1987	176 ± 6	193#	168	175	
1988	161 ± 8			167	
1989	158 ± 8			162	
1990	155 ± 10			151	
1991	149 ± 10			137	146
1992	140 ± 9			131	142
1993	126 ± 7			125	146
1994	125 ± 6			118	128
1995	115 ± 6			113	
1996	109 ± 6			101	

*Reference: Olsson (1989)
†Reference: Burchuladze *et al.* (1989)
‡References: Levin *et al.* (1985); Levin and Kromer (1997)
§Reference: Otlet *et al.* (1997)
#Derived values from data published in graphic form

https://doi.org/10.1017/S0033822200018312 Published online by Cambridge University Press

Several direct measurements on BWRs have shown that the mean ^{14}C release rate is *ca.* 0.5 TBq GW_{el}^{-1} a^{-1} (Kunz 1985; Stenström *et al.* 1995; Wahlen and Kunz 1978a; Winkelmann and Vogl 1984). This release rate would also be expected from the Barsebäck plant. As Figure 1 shows, there might be a small excess in the tree rings for some of the years after the power plant began operating in 1975, but the effect is certainly small. However, we should note that the power plant usually does not operate for several weeks during the yearly summer outage (*i.e.,* the growing season of trees). For BWRs, the ^{14}C release rate is expected to be proportional to the power level, and hence, the releases of ^{14}C should be very low during the outage periods (Stenström *et al.* 1995). This leads to lower ^{14}C concentrations in the tree rings than if the plant had been operating continuously during the growing period. It is also possible that the ^{14}C activity concentration would be higher in a tree located closer to the source of the releases. We will investigate this in the near future.

Figures 2A and 2B show the ^{14}C enrichment of air samples collected in the downwind direction at various distances from the nuclear power plants of Barsebäck and of Forsmark (120 km north of Stockholm, 3 BWRs: 968 MW, 969 MW, 1155 MW). For comparison, the measured $\Delta^{14}C$ value of an air sample from a crosswind site is shown, as well as the prevailing $\Delta^{14}C$ values of *Juncus* in Måryd (see Table 1). AMS measurements of the extracted CO_2 gave results of $\delta^{13}C = -16.3‰$. Thus, an isotopic fractionation occurs in the sample preparation procedure, since, for atmospheric CO_2, the $\delta^{13}C$ value is *ca.* −8‰. If the actual level of ^{14}C in Bq m^{-3} of air is to be calculated, the data must be normalized to this $\delta^{13}C$ value. The air samples collected at Barsebäck show elevated ^{14}C concentrations a few km from the power plant (Fig. 2A). Data from Forsmark (Fig. 2B) show no peak at all, which might be explained by the weather conditions (almost no wind). The samples from Forsmark, which were collected during the winter season, also show lower ^{14}C concentrations than the Barsebäck samples. This could be because the concentration of ^{14}C is influenced by the combustion of fossil fuels (Suess effect), which decreases the ambient ^{14}C specific activity, especially during winter (Levin *et al.* 1988, 1989). Our samples were taken on the same day but at different times, and each sample was collected during a few minutes.

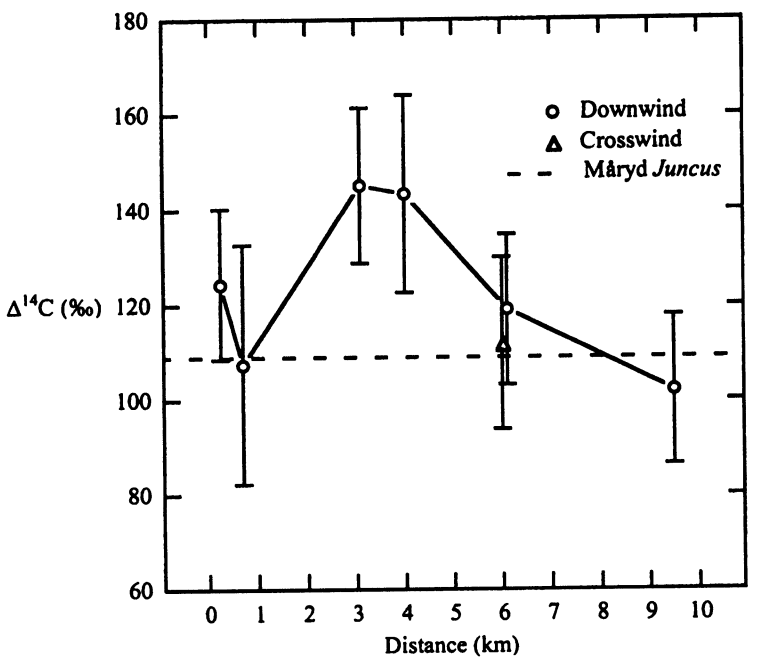

Fig. 2A. $\Delta^{14}C$ values of atmospheric CO_2 at various downwind distances and at one crosswind site from the Barsebäck nuclear power plant. Sampling date was 10 July 1996; wind speed 7 m s^{-1}; temperature 16°C. The 1996 year value of *Juncus* from the "clean air" site Måryd is indicated.

https://doi.org/10.1017/S0033822200018312 Published online by Cambridge University Press

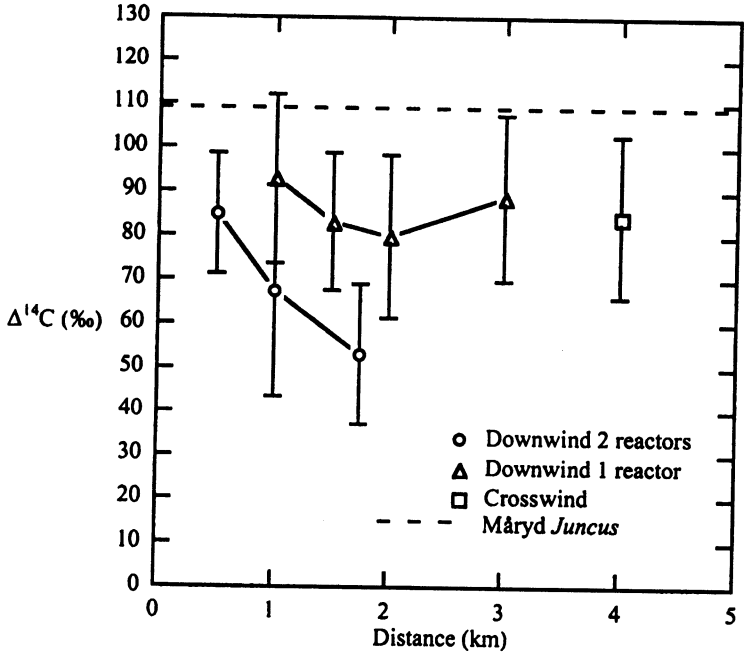

Fig. 2B. $\Delta^{14}C$ values of atmospheric CO_2 at various downwind distances from the Forsmark nuclear reactors. Sampling date 3 March 1996; wind speed 2 m s^{-1}; temperature 0°C. The 1996 year value of *Juncus* from the "clean air" site Måryd is indicated.

Conclusion

A slight excess of ^{14}C has been found in tree rings and air samples around the Barsebäck nuclear power plant. To be able to draw any conclusions about the dispersion of ^{14}C released from the nuclear power plants, we intend to take simultaneous measurements of the release rate (by collecting air from the stack) and at various distances from the plant.

Acknowledgment

This investigation was financially supported by the Swedish Radiation Protection Institute (project SSI P 928.95).

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