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*Slutrapport för projekt*
*SSI P 928.95*

# $^{14}$C EMISSION FROM SWEDISH NUCLEAR POWER PLANTS AND ITS EFFECT ON THE $^{14}$C LEVELS IN THE ENVIRONMENT

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# $^{14}C$ emission from Swedish nuclear power plants and its effect on the $^{14}C$ levels in the environment

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**Abstract**

The radionuclide $^{14}C$ is produced in all types of nuclear reactors mainly by neutron-induced reactions in oxygen ($^{17}O$), nitrogen ($^{14}N$) and carbon ($^{13}C$). Part of the $^{14}C$ created is continuously released during normal operation as airborne effluents in various chemical forms (such as $CO_2$, CO and hydrocarbons) to the surroundings. Because of the biological importance of carbon and the long physical half-life of $^{14}C$, it is of interest to measure the releases and their incorporation into living material.

The $^{14}C$ activity concentrations in annual tree rings and air around two Swedish nuclear power plants (Barsebäck and Forsmark) as well as the background $^{14}C$ activity levels from two reference sites in southern Sweden during 1973-1996 are presented in this report.

In order to verify the reliability of the method some investigations have been conducted at two foreign nuclear sites, Sellafield fuel reprocessing plant in England, and Pickering nuclear generating station in Canada, where the releases of $^{14}C$ are known to be substantial. Furthermore, results from some measurements in the vicinity of Paldiski submarine training centre in Estonia are presented.

The results of the $^{14}C$ measurements of air, vegetation and annual tree rings around the two Swedish nuclear power plants show very low enhancements of $^{14}C$, if at all above the uncertainty of the measurements. Even if the accuracy of the measurements of the annual tree rings is rather good (1-2%) the contribution of $^{14}C$ from the reactors to the environment is so small that it is difficult to separate it from the prevailing background levels of $^{14}C$. This is the case for all sampling procedures: in air and vegetation as well as in annual tree rings. Only on a few occasions an actual increase is observed. However, although the calculations suffer from rather large uncertainties, the calculated release rate from Barsebäck is in fair agreement with reported release data.

The results of this investigation show that the effective doses to man related to the releases of $^{14}C$ from the Swedish light-water reactors at Barsebäck and Forsmark are very low, especially compared to the situation at other nuclear installations, such as the fuel reprocessing plant at Sellafield, England, and the heavy-water reactors at Pickering nuclear generating station, Canada.

Lund/Malmö February 2000

# Contents

**1. Introduction** ..... **1**

**2. Environmental $^{14}C$: Natural and anthropogenic sources** ..... **3**

**2.1 Natural $^{14}C$** ..... **3**

**2.2 Anthropogenic $^{14}C$** ..... **4**

**3. Materials and methods** ..... **7**

**3.1 The AMS technique** ..... **7**

*3.1.1 Sample preparation* ..... 7

*3.1.2 Measurement* ..... 8

**3.2 Decay counting** ..... **8**

**3.3 Radiation dosimetry** ..... **8**

**4. Background data and sampling sites** ..... **9**

**4.1 Background data** ..... **9**

**4.2 Swedish nuclear facility sampling sites** ..... **10**

*4.2.1 Barsebäck* ..... 10

*4.2.2 Forsmark* ..... 10

**5. Results** ..... **11**

**5.1 Background values** ..... **11**

**5.2 $^{14}C$ levels at the Barsebäck nuclear power facility** ..... **14**

*5.2.1 Tree ring samples at Barsebäck* ..... 14

*5.2.2 Vegetation samples at Barsebäck* ..... 18

*5.2.3 Air samples at Barsebäck* ..... 19

**5.3 $^{14}C$ levels at the Forsmark nuclear power facility** ..... **21**

*5.3.1 Tree ring and vegatation samples at Forsmark* ..... 21

*5.3.2 Air samples at Forsmark* ..... 21

**6. Comparison of data with the Gaussian plume model** ..... **23**

**7. $^{14}C$ levels at three foreign sites** ..... **27**

**7.1 Sellafield, England** ..... **27**

**7.2 Pickering, Canada** ..... **28**

**7.3 Paldiski, Estonia** ..... **29**

**8. Absorbed dose estimates** ..... **31**

**9. Concluding remarks** ..... **33**

**References** ..... **35**

**APPENDIX A: Brief literature survey** ..... **39**

**APPENDIX B: Instruction for the preparation of tree ring samples (in Swedish)** ..... **43**

## 1. Introduction

$^{14}C$ is produced by neutron-induced reactions in all types of nuclear reactors. Part of the $^{14}C$ created in reactors is continuously released as airborne effluents mainly through the ventilation system of the power plant. The chemical form of the effluents depends on the type of reactor. In pressurised water reactors (PWRs), the radioactive carbon isotope produced is expected to form mainly lighter hydrocarbons, and to a lesser degree $CO_2$ and CO. In boiling water reactors (BWRs) the dominating form of release is $^{14}CO_2$.

Because of the biological importance of carbon and the long physical half-life of $^{14}C$ (5730 years) it is of interest to study the amount of activity released from nuclear facilities. It has been estimated that $^{14}C$ will produce the highest collective effective dose of all radionuclides discharged in routine operation by the nuclear industry (UNSCEAR, 1993).

It has to be stressed that the local collective doses due to $^{14}C$-releases from reactors only represent a small fraction of the total collective dose commitment. The main significance of $^{14}C$ originates from its entry into the carbon cycle and the resulting global dispersion, leading to long-term irradiation.

Several investigations have been made in the vicinity of BWR and PWR reactors (*e.g.* Levin *et al.*, 1988; Loosi and Oeschger, 1989; Obelic *et al.*, 1986; Uchrin *et al.*, 1998) but only rather small excess activities have been found. In the surroundings of heavy-water reactors and fuel reprocessing plants, the excess activities are much higher (Milton *et al.*, 1995; Otlet *et al.*, 1990; McCartney *et al.*, 1986). A brief summary of some reports on measurements of $^{14}C$ in the vicinity of various nuclear installations are given in Appendix A.

The aim of this investigation was to study the activity concentrations of $^{14}C$ in air and vegetation around Swedish nuclear power plants. Two plants were included, Barsebäck and Forsmark, both with reactors of the BWR type and thus with $^{14}CO_2$ as dominating form of the airborne $^{14}C$ release. The Barsebäck plant was most thoroughly studied with analysis of air, vegetation and annual rings from trees. At the Forsmark plant atmospheric $CO_2$ was analysed as well as a few vegetation samples and one sample of annual tree rings.

The measurements presented in this report are part of an extensive investigation of the $^{14}C$ releases from some Swedish nuclear power plants (Stenström *et al.*, 1993a; 1993b, 1995a; 1995b; 1995c; 1996a). In the previous studies the release rates through the stacks and the chemical form of the releases have been established by use of the accelerator mass spectrometry (AMS) technique.

In order to calculate the excess of $^{14}C$ in the environment originating from the nuclear power plants, $^{14}C$ background values from two reference sites in southern Sweden, from 1973 to 1997, are also presented.

Some measurements of the $^{14}C$ activity in vegetation growing in the vicinity of Sellafield nuclear facility (England), Paldiski submarine training centre (Estonia) and Pickering nuclear generating station (Canada) are also reported for comparison.

Parts of the results presented in this report have been published elsewhere (Stenström *et al.*, 1996b, 1998; Erlandsson *et al.*, 1999).

## 2. Environmental $^{14}C$: Natural and anthropogenic sources

$^{14}C$ is produced both by natural and anthropogenic processes. The following sections give a brief summary of the production mechanisms involved and estimations of the quantity produced by the different sources.

### *2.1 Natural $^{14}C$*

$^{14}C$ is one of many radionuclides that are produced naturally in the atmosphere by nuclear reactions induced by cosmic rays (Lal and Peters, 1967). In the case of $^{14}C$, reactions between secondary moderated neutrons and stable $^{14}N$ nuclei are responsible for the most significant production process, $^{14}N(n,p)^{14}C$ (cross section $\sigma$=1.8 b), which takes place in the upper atmosphere. The $^{14}C$ produced (about $10^{15}$ Bq per year (UNSCEAR, 1993)) is quickly incorporated mainly into molecules of carbon dioxide, which are mixed throughout the atmosphere and the hydrosphere, the latter being the main long-term reservoir. A significant reservoir is provided by the biosphere because of the assimilation of carbon dioxide into vegetation, and the consumption of these plants by animals. The natural production builds up to an equilibrium content of about $8 \cdot 10^{18}$ Bq (Fairhall and Young, 1970; Hayes and MacMurdo, 1977) in the atmosphere, hydrosphere and biosphere.

The long-term mean $^{14}C$ specific activity of natural carbon has been determined, by measurements of tree rings of known age, to be 226 Bq/kg C (Karlén, 1964; Libby, 1965). This value equals the "zero point" in the generally adopted $\Delta^{14}C$ scale[1] (Stuiver and Polach, 1977). This natural $^{14}C$ specific activity leads to an annual effective dose to man of about 12 μSv (UNSCEAR, 1977).

---

1 $^{14}C$ specific activity as well as $\Delta^{14}C$ values are used for the results in this report. $\Delta^{14}C$ values are useful when comparing activities of different sample materials because $\Delta^{14}C$ is corrected for so called isotope fractionation. The fractionation of the isotopes of carbon occurs in nature due to various physio-chemical reactions. The result is that the abundances of the isotopes of the element, which become incorporated into plants during photosyntesis, are different from those in the atmosphere. Thus the specific activity of a plant differ somewhat from the specific activity of the carbon in the $CO_2$ in the contemporary atmosphere. However, $\Delta^{14}C$ is corrected for the fractionation and is thus the same for the plant and the $CO_2$. $\Delta^{14}C$ is defined as:

$$\Delta^{14}C = \left[ \frac{A_{SN}}{A_{ABS}} - 1 \right] \cdot 1000‰$$

where $A_{SN}$ is the sample net activity corrected for isotope fractionation and $A_{ABS}$ is the absolute international standard activity (95% of the activity of the oxalic acid standard normalised for fractionation and corrected for decay since 1950).

### *2.2 Anthropogenic $^{14}C$*

Over the past two centuries anthropogenic activities have influenced the environmental $^{14}C$ balance tremendously. The first major change started with the Industrial Revolution. The large-scale combustion of fossil fuels (such as coal, oil and gas, which originate from very old geological reservoirs and in which essentially all radioactive carbon has decayed) introduced non-radioactive $CO_2$ into the atmosphere. This increased the amount of the stable carbon isotopes in the atmosphere, which resulted in a decrease in ambient $^{14}C$ specific activity. The phenomenon is called the Suess effect and has been traced back to about 1850 AD by analysis of tree-rings (Suess, 1955). Between 1850 and 1950 AD, the Suess effect increased in magnitude and the decrease of atmospheric, $^{14}C$-specific activity during this period amounted to about 3% (Oeschger *et al.*, 1975).

The second major effect on the $^{14}C$ balance is due to the atmospheric testing of nuclear weapons in the past and is called the Bomb effect. The tests, beginning in the 1950's, have, in contrast to the Suess effect, caused a significant increase of the terrestrial, $^{14}C$-specific activity. This $^{14}C$ is produced by neutrons, generated in the explosion, that interact with nitrogen atoms of the atmosphere in the same way as the neutrons resulting from the cosmic rays. In the early 1960's the number of detonations of nuclear devices reached a maximum. This is reflected in a maximum of atmospheric $^{14}C$ activity in 1963 of about twice the natural in the northern hemisphere (Levin *et al.*, 1985). It has been estimated that about $0.22 \cdot 10^{18}$ Bq $^{14}C$ has been produced and released into the atmosphere by weapon tests (UNSCEAR, 1993). In Fig. 1 the atmospheric $^{14}C$ activity during the 20th century is shown.

The $^{14}C$ activity concentration in the atmosphere has steadily decreased since the peak in 1963, due to mixing with the biosphere and the oceans, as can be seen in Fig. 1. However, also an increase in the anthropogenic fossil $CO_2$ emission contribute to the declining curve. The decline is slightly counteracted by airborne $^{14}C$ emissions from the nuclear industry (according to Levin *et al.* (1997) by less than 1.5‰ per year). These $^{14}C$ emissions mainly originate from the nuclear power industry, *i.e.* from nuclear power plants as well as from associated facilities such as fuel reprocessing plants. In nuclear reactors, $^{14}C$ is produced by the neutron activation of stable isotopes of carbon, nitrogen and oxygen in the fuel, cladding, coolant, moderator and structural materials of the reactor. According to UNSCEAR (1993), approximately $1.44 \cdot 10^{15}$ Bq of $^{14}C$ has been released from nuclear sites up to the end of 1989. A smaller part of the $^{14}C$ released and contributing to the global $^{14}C$ budget originates from the use of $^{14}C$ in disciplines such as medicine, biology and chemistry. The amount of $^{14}C$ produced for use in research and industry are not well documented (UNSCEAR, 1993), and thus the

contribution to the atmospheric $^{14}C$ activity is not well-known. However, UNSCEAR (1993) estimates the probable production to be about $5 \cdot 10^{13}$ Bq annually.

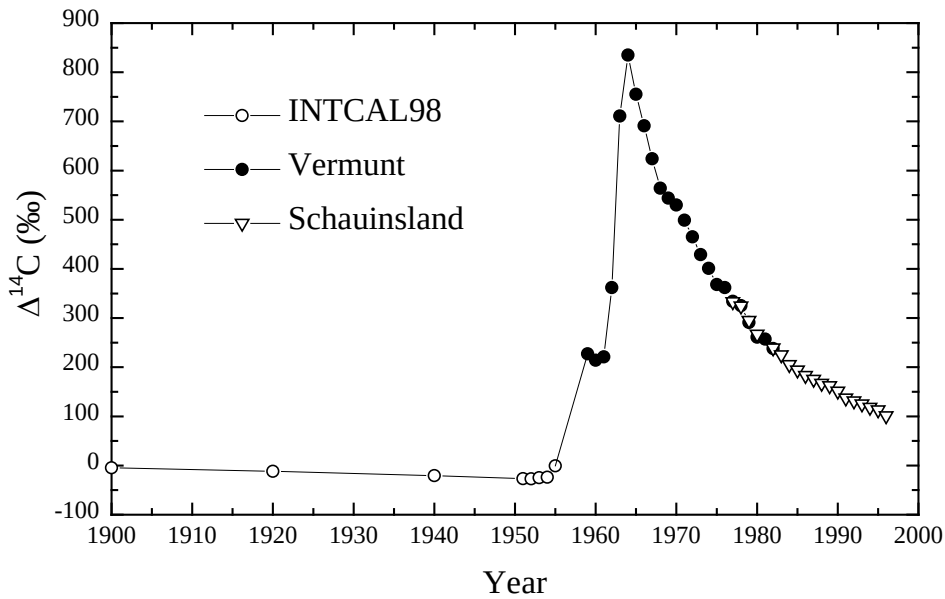


**Fig. 1** The atmospheric $\Delta^{14}C$ during the 20th century. INTCAL98[2] are tree ring data, Vermunt (Levin *et al.*, 1985) and Schauinsland (Levin *et al.*, 1985, Levin *et al.*, 1997) are atmospheric $CO_2$ data.

The $^{14}C$ levels in Fig. 1 represent "clean air" data, *i.e.* with a minimum of local contamination from anthropogenic sources. However, the $^{14}C$ specific activity in various materials (such as air, soil, vegetation and water) may differ considerably in the vicinity of fossil fuel sources (like coal- and oil-heated power plants, traffic roads etc.) as well as of $^{14}C$ emission sources such as nuclear power plants, fuel reprocessing facilities and laboratories.

[2] Researchers using radiocarbon dating rely upon internationally accepted calibration curves to correlate probable calendar date ranges with conventional radiocarbon ages. INTCAL98, that includes graphs and data sets for calibration curves derived from dendrochronology and coral dates, has been published by the journal *Radiocarbon, volume 40, no.3, 1998, p. 1041-1084.*

## 3. Materials and methods

In order to determine the activity concentration and the specific activity of $^{14}C$ in the vicinity of Swedish nuclear power plants, accelerator mass spectrometry (AMS) (Hellborg et al., 1999; Kutschera, 1993; Tuniz, 1998) as well as conventional decay counting technique using gas-filled proportional counters have been used.

### *3.1 The AMS technique*

#### *3.1.1 Sample preparation*

*Pretreatment*

*Annual tree rings* were cut out from bore cores (ϕ=5 mm) giving 50-100 mg of wood per year. The samples were pretreated with a acid-alkali-acid (AAA) procedure followed by cellulose extraction (see *e.g.* Olsson & Possnert, 1992). The AAA pretreatment, which removed humic acids and extracted resin from the wood, was accomplished by boiling the sample in HCl (3M) for 30 min, washing, boiling in NaOH (1M) for 30 minutes, washing and boiling the sample again in HCl (3M) for another 30 minutes prior to final washing. The cellulose extraction was performed 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) were added. After another hour the remaining cellulose was washed, filtered and dried. A detailed description of the sampling and preparation of tree rings is given in Appendix B (in Swedish).

*Vegetation samples* were washed in distilled water, dried and pulverised in a mortar. A few mg of the pulverised material was mixed with a few hundred mg of copper oxide.

*Air samples* (about 40 l per sample) were collected in beach balls, and in the lab each air sample was drawn through a drying agent of $CaCl_2$ prior to absorption of the $CO_2$ in Ascarite® (NaOH on a solid support).

*Graphitization*

Carbon dioxide was formed in a vacuum system by combustion of the organic material (about 10 mg) mixed with CuO (a few hundred mg), or by hydrolysis (using phosphoric acid) of the Ascarite from the air samples. Elemental carbon was then produced by reducing the carbon dioxide, mixed with hydrogen gas, over an iron catalyst at 650°C (Vogel *et al.*, 1984). The reduction of the carbon dioxide to solid carbon took 3-4 hours per sample. The use of several simultaneously operating preparation lines provided an efficient sample preparation. Further information about

the sample preparation system can be found in Stenström *et al.* (1994) and Stenström (1995d).

### *3.1.2 Measurement*

The AMS measurements were performed at the Lund AMS facility, described in detail elsewhere (Wiebert, 1995; Håkansson *et al*., 1996; Hellborg *et al.,* 1999). The samples, each consisting of a few mg of elemental carbon, were placed in the ion source of the AMS system. In this system, the number of $^{14}C$ atoms was counted relative to the amount of $^{13}C$ in the sample. To establish the activity of the samples, carbon samples made from the NBS oxalic acid standard (Stuiver and Polach, 1977), of known activity (Karlén *et al.*, 1964), were also measured and were used as a reference. Samples of $^{14}C$-free anthracite, processed in the sample preparation system, were measured to establish the background of the sample preparation and accelerator systems. Each unknown sample was measured at least three times, each time for about 20 minutes. The $\Delta^{14}C$ values (‰ depletion or enrichment relative to standard normalised for isotope fractionation) and the specific $^{14}C$ activity below were obtained according to Stuiver and Polach (1977) with the modifications for AMS described in Donahue *et al.* (1990).

## ***3.2 Decay counting***

"Clean air" background samples in the form of rush (*Juncus*) from one of the reference sites, Måryd, have been measured at the Radiocarbon Dating Laboratory in Lund according to the standard technique with a gas proportional counter (Håkansson, 1968; 1986).

## ***3.3 Radiation dosimetry***

The common approach to $^{14}C$-dosimetry is to assume that $^{14}C$ in the human body is in equilibrium with that in the environment, so that the specific activity in the body is identical to that in the environment (NCRP, 1985). With this approach the absorbed dose to various organs and tissues has been calculated taking the different carbon content in various organs and tissues into consideration (Killough and Rohwer, 1978; NCRP, 1985). In the normal environmental exposure situation the major pathway of exposure is by ingestion.

## 4. Background data and sampling sites

### *4.1 Background data*

Samples of rush (*Juncus*), grown at the shore of a small pond in Måryd (see Fig. 2), were used as background values of the prevailing $^{14}C$ levels. Måryd (15 km E of Lund, 25 km NE of Malmö and 30 km E of Barsebäck), 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 growth season, *i.e.* the summer months.

Tree rings of pine (*Pinus*) grown at Borrby (east coast of south Scania, see Fig. 2) also served as background data.

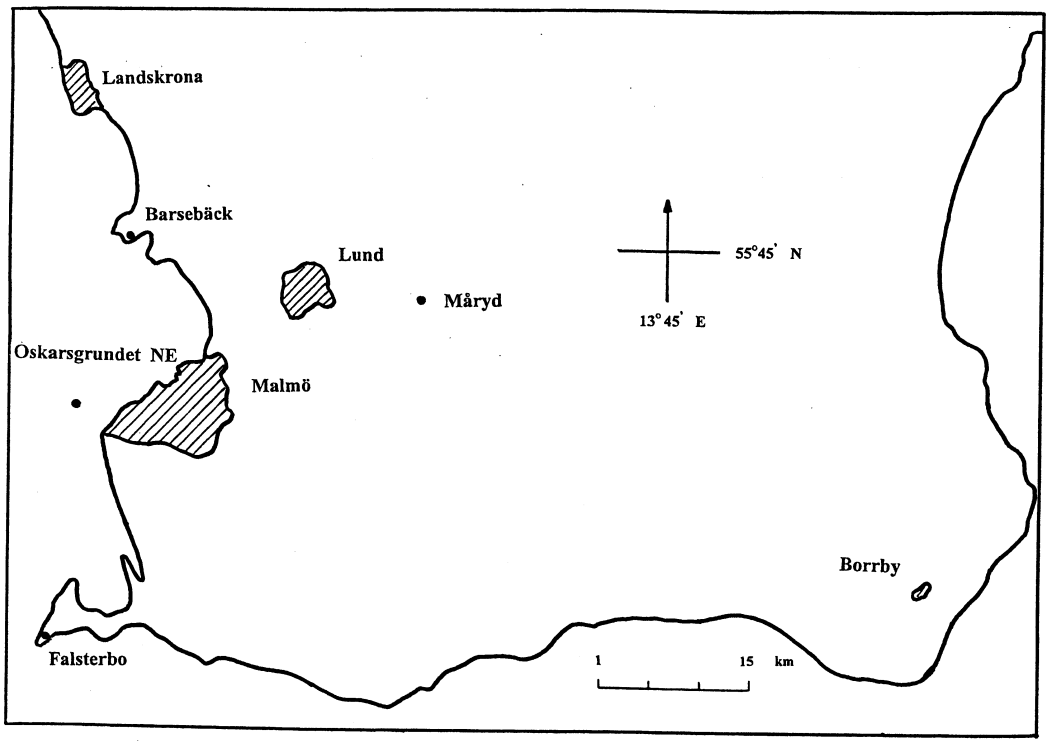


**Fig. 2** Sampling sites in southern Sweden

As additional information, different European background data are also presented. Particularly useful were the atmospheric $CO_2$ data, presented by Levin *et al.* (1985) and Levin and Kromer (1997), which should be representative for Central European background air. These air samples have been continuously collected in biweekly periods at ca 5 m above the ground at the Schauinsland mountain top in southern Germany at 1205 m above sea level. This sampling site should show a minimum of local contamination.

### *4.2 Swedish nuclear facility sampling sites*

#### *4.2.1 Barsebäck*

The Barsebäck (see Fig. 2) nuclear power plant has two BWRs, each with an electrical power of 600 $MW_{el}$.

*Tree ring* samples of pine (*Pinus*) were collected at 1 km and 3 km ENE (dominant down-wind direction) of the reactors, for the years 1981-1997 and 1976-1991, respectively.

*Vegetation* samples in the form of willow (*Salix viminalis*) leaves were collected at various distances from the plant in 1996. The tree leaf samples were collected east of the plant (in the dominant wind direction).

Samples of *atmospheric* $CO_2$ were collected at various distances at one occasion in July 1996 and another in December 1997 in the main downwind direction (ENE).

#### *4.2.2 Forsmark*

The Forsmark nuclear power plant has 3 BWRs: 968 $MW_{el}$, 969 $MW_{el}$ and 1155 $MW_{el}$.

*Tree ring* samples of pine (*Pinus*) were collected at 2.5 km south of the reactors (frequent down-wind direction). The tree ring from the year 1992 was analysed in this project. The year 1992 was chosen since the actual releases of $^{14}C$ from one of the Forsmark reactors, F1, were measured that particular year (Stenström *et al*., 1995c).

*Vegetation* samples, in the form of sallow (*Salix caprea*), were collected on 5 May, 1994, at 1.5 and 3.2 km south of the plant.

*Atmospheric* $CO_2$ was collected in March 1996 in the vicinity of the power plant.

## 5. Results

### *5.1 Background values*

In Table 1 the background values from Måryd (*Juncus*) are compared with other European background data. Some of the Måryd values, which were measured with a gas-filled proportional counter, have been published previously by Håkansson (1977; 1987; 1988). The Måryd data are in good agreement with the summer means (corresponding to the growth period of the Måryd *Juncus*) of the Schauinsland $CO_2$ data.

**TABLE 1.** $\Delta^{14}C$ values from the "clean air" site Måryd in the southernmost Sweden compared with published data of background $^{14}C$ activity from different other places in Europe. Values in () are derived values from data published in graphical form. The Schauinsland data are summer means (May to August), and the values within [] are yearly means. The Måryd samples have been collected in September or October. (St98=Stenström *et al.*, 1998; Ol89=Olsson, 1989; Bu89=Burchuladze *et al.*; 1989; Le85=Levin *et al.*, 1985; Le97=Levin *et al.*, 1997; Ot97=Otlet *et al.*, 1997)

| | $\Delta^{14}C$ (‰) | | | | |
|---|---|---|---|---|---|
| Year | Måryd *Juncus* (St98) | Abisko $CO_2$ (Ol89) | Georgian *Wine* (Bu89) | Schauinsland $CO_2$ (Le85, Le97) | UK *plant material* (Ot97) |
| 1975 | 390 ± 6 | | 368 | | |
| 1976 | | | 332 | | |
| 1977 | | | 298 | 335 ± 3 [333] | |
| 1978 | 342 ± 6 | | 345 | 335 ± 8 [324] | |
| 1979 | 305 ± 6 | | 295 | 300 ± 9 [295] | 341 |
| 1980 | 286 ± 6 | | 279 | 271 ± 5 [267] | 327 |
| 1981 | 269 ± 6 | | 272 | 260 ± 13 | 292 |
| 1982 | 254 ± 6 | | 266 | 244 ± 2 [239] | 279 |
| 1983 | 234 ± 6 | | 223 | 223 ± 4 [225] | |
| 1984 | 223 ± 6 | | 216 | 209 ± 2 [205] | |
| 1985 | 206 ± 6 | (212) | 209 | 200 ± 4 [194] | |
| 1986 | 189 ± 6 | (205) | 190 | 186 ± 4 [183] | |
| 1987 | 176 ± 6 | (193) | 168 | 183 ± 4 [175] | |
| 1988 | 161 ± 8 | | | 169 ± 1 [167] | |
| 1989 | 158 ± 8 | | | 159 ± 5 [162] | |
| 1990 | 155 ± 10 | | | 149 ± 4 [151] | |
| 1991 | 149 ± 10 | | | 138 ± 4 [137] | 146 |
| 1992 | 140 ± 9 | | | 134 ± 4 [131] | 142 |
| 1993 | 126 ± 7 | | | 126 ± 2 [125] | 146 |
| 1994 | 125 ± 6 | | | 120 ± 1 [118] | 128 |
| 1995 | 115 ± 6 | | | 112 ± 2 [113] | |
| 1996 | 109 ± 6 | | | 104 ± 3 [101] | |
| 1997 | 99 ± 5 | | | | |

In Table 2 (see next section), the $\Delta^{14}C$-values from Borrby (*Pinus* tree rings) and Måryd are listed, and the two series of measurements agree within the error limits (one standard deviation) with one exception.

The Schauinsland data are presented in Fig. 3, which also includes the Måryd and Borrby data. As shown in the figure, the background data are in good agreement. The depletion of the Schauinsland data, due to influence of fossil fuel $CO_2$ emission over the European continent, compared to maritime background measurements is up to 5‰ (Levin and Kromer, 1997). The Schauinsland data show seasonal variations of atmospheric $^{14}CO_2$ levels, with minimum values occurring in the winter half year. Part is due to a seasonally varying fossil-fuel $CO_2$ input, and part is due to atmospheric mixing as well as atmosphere-biosphere exchange (Levin and Kromer, 1997). In densely populated areas, like in the vicinity of the nuclear installations reported in this paper, the effects are expected to be stronger due to adjacent fossil fuel sources.

It must be remembered that the background data represent clean air levels, and that the data obtained from trees and vegetation represent the $^{14}C$ levels during the growth season. During wintertime the atmospheric $^{14}CO_2$ levels can decrease significantly in highly populated areas due to the combustion of fossil fuels. During winter in Heidelberg, Levin *et al.* (1989) have observed atmospheric $^{14}C/^{12}C$ ratios of up to 10% lower than the clean air background.

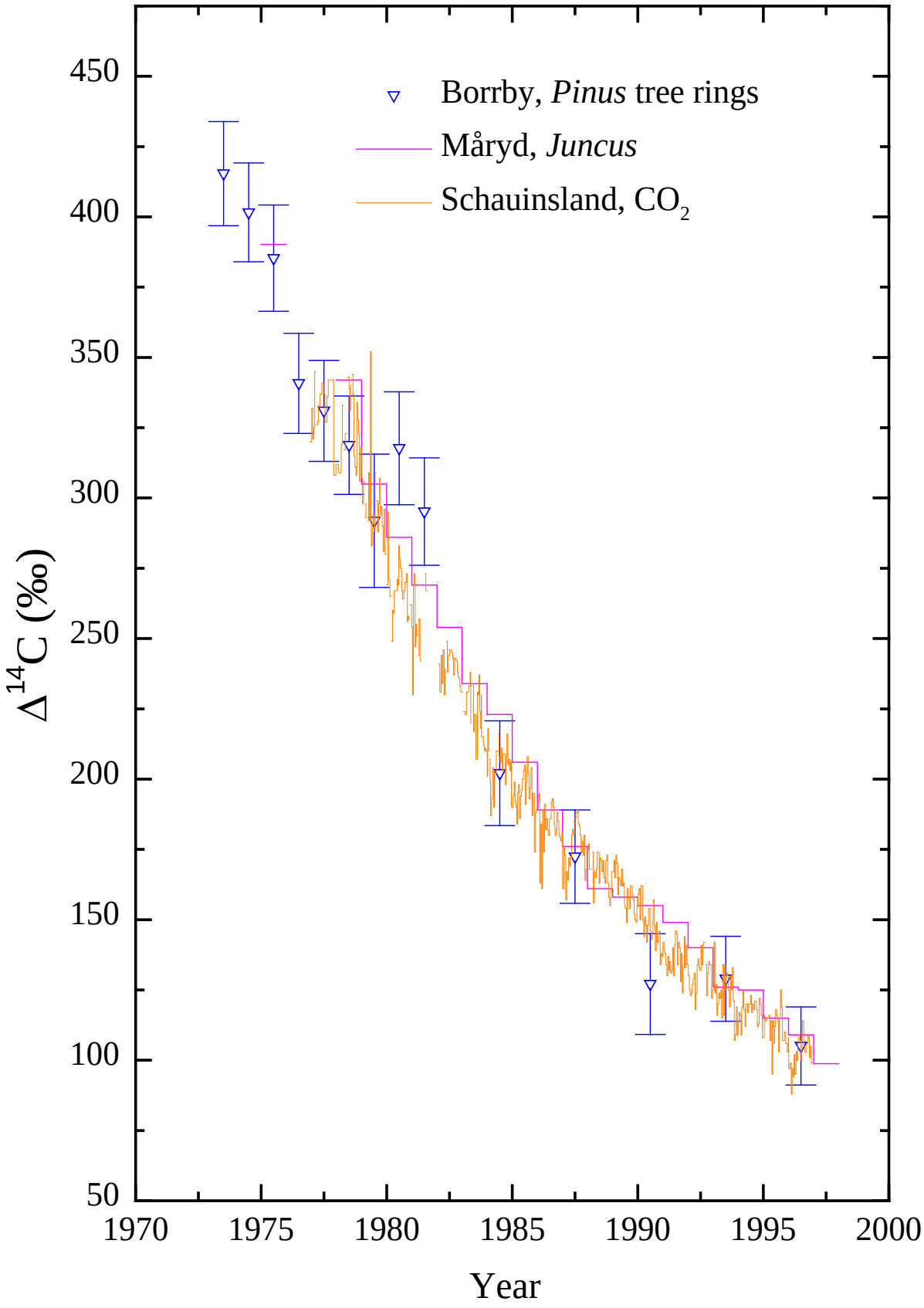


**Fig. 3** Background $\Delta^{14}C$ values of the clean air sites of Måryd (*Juncus*), Borrby (*Pinus*) and Schauinsland (atmospheric $CO_2$) (Levin *et al.*, 1985, Levin *et al.*, 1997). The uncertainty (one standard deviation) in the Måryd and Schauinsland data typically varies between $\Delta^{14}C$ ±5‰ and ±10‰.

### *5.2 $^{14}C$ levels at the Barsebäck nuclear power facility*

#### *5.2.1 Tree ring samples at Barsebäck*

The results of the $^{14}C$ measurements in annual rings from two trees of *Pinus* located 1 km and 3 km ENE (frequent down-wind direction) of the Barsebäck nuclear power plant are shown in Table 2 and Fig. 4. The background data from Måryd, Borrby and Schauinsland are included in the figure.

**TABLE 2.** The $^{14}C$ content in tree rings of *Pinus* located 1 km and 3 km from the Barsebäck nuclear power plant. The background levels are represented by the "clean air" site Måryd (*Juncus*) in the southernmost Sweden and the "clear air" site Borrby (*Pinus*). The values from Borrby and Barsebäck are measured by AMS while the Måryd data have been obtained by decay counting. The Måryd samples have been collected in September or October. Sampling dates for the tree rings: Borrby: 1 January 1996; Barsebäck 3 km: 2 February 1997; Barsebäck 1 km: 26 October 1997.

| | Måryd *Juncus* | | Borrby *Pinus* | | Barsebäck 3 km *Pinus* | | Barsebäck 1 km *Pinus* | |
|---|---|---|---|---|---|---|---|---|
| Year | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) |
| 1973 | | | 415 ± 18 | 320 ± 4 | 419 ± 19 | 321 ± 4 | | |
| 1974 | | | 402 ± 18 | 317 ± 4 | 413 ± 19 | 319 ± 4 | | |
| 1975 | 390 ± 6 | 314 ± 2 | 385 ± 19 | 313 ± 4 | 418 ± 22 | 320 ± 5 | | |
| 1976 | | | 341 ± 18 | 303 ± 4 | 338 ± 17 | 303 ± 4 | | |
| 1977 | | | 331 ± 18 | 301 ± 4 | 312 ± 18 | 296 ± 4 | | |
| 1978 | 342 ± 6 | 303 ± 2 | 319 ± 17 | 298 ± 4 | 331 ± 26 | 301 ± 6 | | |
| 1979 | 305 ± 6 | 295 ± 2 | 292 ± 24 | 292 ± 4 | 323 ± 27 | 299 ± 6 | | |
| 1980 | 286 ± 6 | 291 ± 2 | 318 ± 20 | 298 ± 5 | 283 ± 21 | 290 ± 5 | | |
| 1981 | 269 ± 6 | 287 ± 2 | 295 ± 19 | 293 ± 4 | 311 ± 22 | 296 ± 5 | 297 ± 18 | 293 ± 4 |
| 1982 | 254 ± 6 | 283 ± 2 | | | 241 ± 18 | 280 ± 4 | 246 ± 40 | 282 ± 9 |
| 1983 | 234 ± 6 | 279 ± 2 | | | 235 ± 20 | 279 ± 4 | 199 ± 21 | 271 ± 5 |
| 1984 | 223 ± 6 | 276 ± 2 | 202 ± 19 | 272 ± 4 | 224 ± 18 | 277 ± 4 | 222 ± 22 | 276 ± 5 |
| 1985 | 206 ± 6 | 272 ± 2 | | | 215 ± 20 | 274 ± 5 | 215 ± 14 | 275 ± 3 |
| 1986 | 189 ± 6 | 269 ± 2 | | | 226 ± 19 | 277 ± 4 | 190 ± 21 | 269 ± 5 |
| 1987 | 176 ± 6 | 266 ± 2 | 172 ± 17 | 265 ± 4 | 214 ± 16 | 274 ± 4 | 197 ± 25 | 271 ± 6 |
| 1988 | 161 ± 8 | 262 ± 2 | | | 159 ± 22 | 262 ± 5 | 194 ± 20 | 270 ± 5 |
| 1989 | 158 ± 8 | 262 ± 2 | | | 165 ± 21 | 263 ± 5 | 168 ± 38 | 264 ± 9 |
| 1990 | 155 ± 10 | 261 ± 2 | 127 ± 18 | 255 ± 4 | 172 ± 16 | 265 ± 4 | 173 ± 29 | 265 ± 6 |
| 1991 | 149 ± 10 | 260 ± 2 | | | 163 ± 16 | 263 ± 4 | 135 ± 17 | 256 ± 4 |
| 1992 | 140 ± 9 | 258 ± 2 | | | 162 ± 21 | 263 ± 5 | 160 ± 14 | 262 ± 3 |
| 1993 | 126 ± 7 | 254 ± 2 | 129 ± 15 | 255 ± 4 | 144 ± 17 | 258 ± 4 | 137 ± 15 | 257 ± 3 |
| 1994 | 125 ± 6 | 254 ± 2 | | | 113 ± 15 | 252 ± 4 | 152 ± 16 | 260 ± 4 |
| 1995 | 115 ± 6 | 252 ± 2 | | | 103 ± 18 | 249 ± 3 | 124 ± 16 | 254 ± 3 |
| 1996 | 109 ± 6 | 251 ± 2 | 105 ± 14 | 250 ± 3 | 111 ± 14 | 252 ± 4 | 109 ± 16 | 251 ± 4 |
| 1997 | 99 ± 5 | 248 ± 2 | | | | | 100 ± 15 | 248 ± 3 |

As can be seen in Fig. 4 there might be a small excess of $^{14}C$ in the tree rings for some of the years after the start of the power plant in 1975, but the effect is certainly small. However, it must be observed that the power plant usually is not operating for

several weeks at the yearly outage during the summer (*i.e.* the growth 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 outage periods (Stenström *et al.* 1995a). This leads to lower $^{14}C$ concentrations in the tree rings than if the plant would be operating continuously during the growth period.

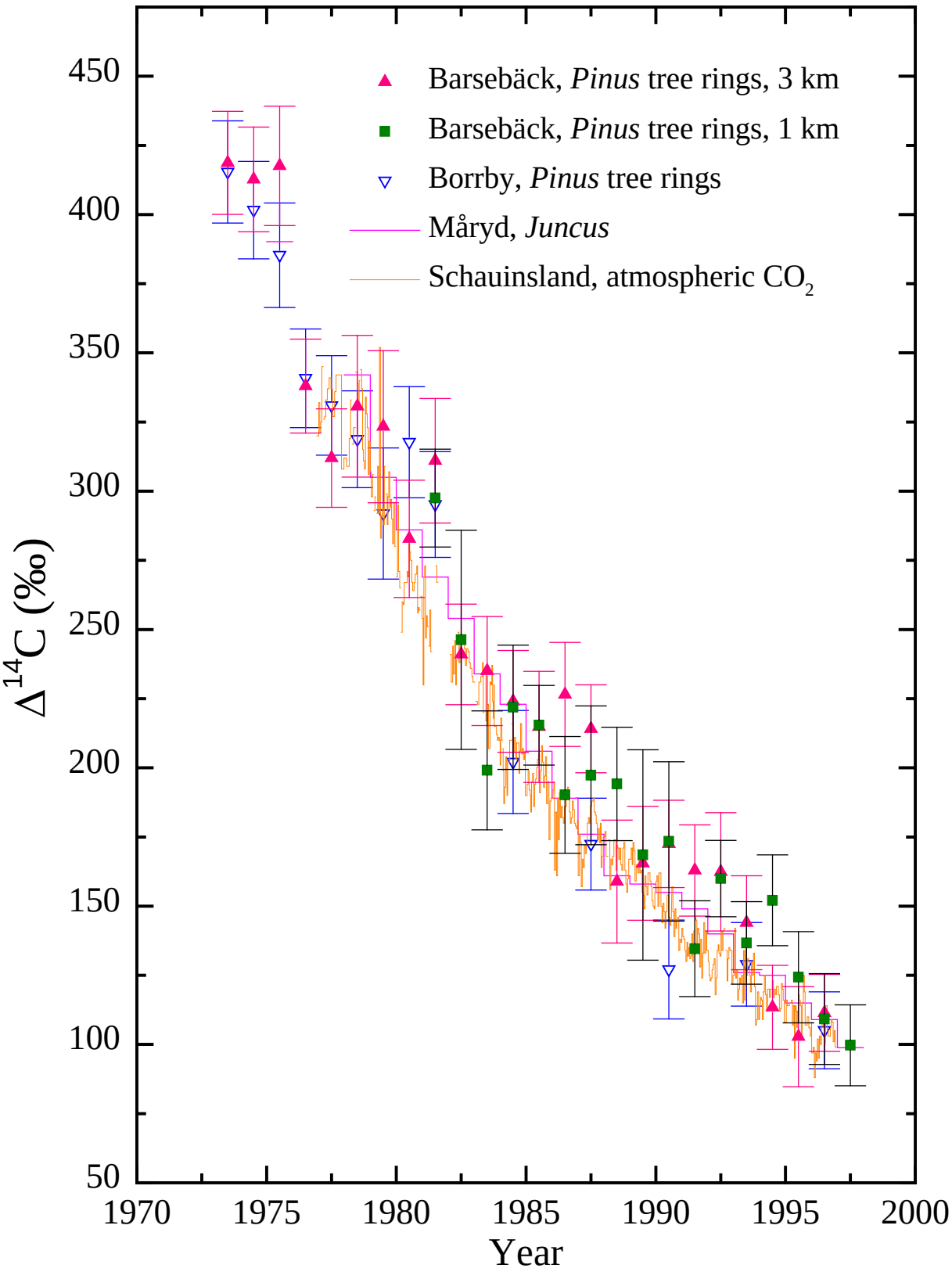


**Fig. 4** $\Delta^{14}C$ values of annual tree rings of *Pinus* at 3 km and 1 km from the nuclear power plant Barsebäck and from the "clean air" site Borrby. Also shown are the $\Delta^{14}C$ values of *Juncus* from Måryd listed in Table 1. The Barsebäck and Borrby values have been obtained by AMS, and the Måryd values by decay counting. As a further reference $\Delta^{14}C$ values of atmospheric $CO_2$ from Schauinsland, Germany, from Levin *et al.* (1985; 1997), are shown. The uncertainty (one standard deviation) in the Måryd and Schauinsland data typically varies between $\Delta^{14}C$ ±5‰ and ±10‰.

In Table 3, the mean $^{14}C$ activity of the two *Pinus* trees at 3 km and 1 km at Barsebäck is compared to the Måryd *Juncus* activity for the years 1981 to 1996. Using the Måryd *Juncus* activity as a background and subtracting it from the mean Barsebäck activity the excess activity at Barsebäck is obtained. The excess activity is certainly small and rarely higher than the uncertainty of the measurements. However, only two of the sixteen values are negative, which indicates a higher $^{14}C$ activity at Barsebäck than at Måryd (at the "clean air" site Borrby 7 of 10 values are negative according to Table 2). The mean excess specific activity at Barsebäck is 2 Bq/kg.

**TABLE 3.** Comparison between the $^{14}C$ content in tree rings of *Pinus* located 1 km and 3 km from the Barsebäck nuclear power plant and the "clean air" site Måryd (*Juncus*). The excess activity at Barsebäck is the mean excess of the activity at Barsebäck compared to Måryd.

| Year | Barsebäck 3 km *Pinus* (Bq/kg C) | Barsebäck 1 km *Pinus* (Bq/kg C) | Barsebäck mean (Bq/kg C) | Måryd *Juncus* (Bq/kg C) | Barsebäck Excess activity (Bq/kg C) |
|---|---|---|---|---|---|
| 1981 | 296 ± 5 | 293 ± 4 | 295 ± 3 | 287 ± 2 | 8 ± 4 |
| 1982 | 280 ± 4 | 282 ± 9 | 281 ± 5 | 283 ± 2 | –2 ± 5 |
| 1983 | 279 ± 4 | 271 ± 5 | 275 ± 3 | 279 ± 2 | –4 ± 4 |
| 1984 | 277 ± 4 | 276 ± 5 | 277 ± 3 | 276 ± 2 | 1 ± 4 |
| 1985 | 274 ± 5 | 275 ± 3 | 275 ± 3 | 272 ± 2 | 3 ± 4 |
| 1986 | 277 ± 4 | 269 ± 5 | 273 ± 3 | 269 ± 2 | 4 ± 4 |
| 1987 | 274 ± 4 | 271 ± 6 | 273 ± 3 | 266 ± 2 | 7 ± 4 |
| 1988 | 262 ± 5 | 270 ± 5 | 266 ± 4 | 262 ± 2 | 4 ± 4 |
| 1989 | 263 ± 5 | 264 ± 9 | 264 ± 5 | 262 ± 2 | 2 ± 5 |
| 1990 | 265 ± 4 | 265 ± 6 | 265 ± 4 | 261 ± 2 | 4 ± 4 |
| 1991 | 263 ± 4 | 256 ± 4 | 260 ± 3 | 260 ± 2 | 0 ± 4 |
| 1992 | 263 ± 5 | 262 ± 3 | 262 ± 3 | 258 ± 2 | 4 ± 4 |
| 1993 | 258 ± 4 | 257 ± 3 | 258 ± 3 | 254 ± 2 | 4 ± 4 |
| 1994 | 252 ± 4 | 260 ± 4 | 256 ± 3 | 254 ± 2 | 2 ± 4 |
| 1995 | 249 ± 3 | 254 ± 3 | 252 ± 2 | 252 ± 2 | 0 ± 3 |
| 1996 | 252 ± 4 | 251 ± 4 | 252 ± 2 | 251 ± 2 | 1 ± 4 |

*5.2.2 Vegetation samples at Barsebäck*

The measured activity concentrations of $^{14}C$ in the collected willow (*Salix viminalis*) leaves are listed in Table 4 and graphically shown in Fig. 5. The error of the results presented corresponds to one standard deviation, and within one standard deviation only one value (at 0.85 km) is above the Måryd value, while two values (at 0.20 and 0.75 km) are below. The rest of the values are generally very close to the Måryd background data.

**TABLE 4.** The $^{14}C$ content in willow (*Salix viminalis*) leaves collected at various distances and in the dominant down-wind direction from the Barsebäck nuclear power plant. Sampling date: 8 October 1996.

| Distance (km) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) |
|---|---|---|
| 0.20 | 68 ± 15 | 240 ± 3 |
| 0.75 | 74 ± 28 | 242 ± 6 |
| 0.85 | 138 ± 23 | 256 ± 5 |
| 1.05 | 98 ± 20 | 248 ± 5 |
| 1.25 | 105 ± 23 | 249 ± 5 |
| 2.00 | 84 ± 24 | 244 ± 5 |
| 2.50 | 95 ± 22 | 247 ± 5 |
| 3.10 | 101 ± 17 | 248 ± 4 |
| 6.10 | 102 ± 26 | 248 ± 6 |
| 8.00 | 98 ± 21 | 248 ± 5 |
| 8.75 | 90 ± 22 | 246 ± 5 |

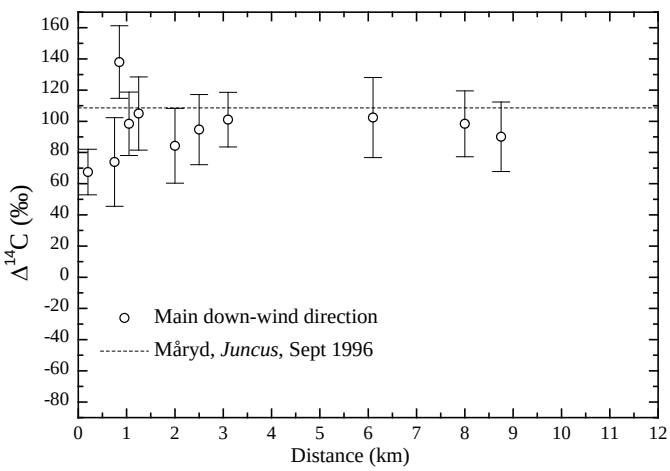


**Fig. 5** $\Delta^{14}C$ values of willow (*Salix viminalis*) leaves collected at various distances from the Barsebäck nuclear power plant, October 1996.

### *5.2.3 Air samples at Barsebäck*

The activity concentration of $^{14}C$ in air samples collected in the down-wind direction at various distances from the Barsebäck nuclear power plant are shown in Fig. 6 (listed in Table 5 and Table 6). For comparison, the measured $\Delta^{14}C$ value of an air sample from a cross-wind site is shown as well as the $\Delta^{14}C$ value of *Juncus* in Måryd (see Table 1). Mass spectrometry of the extracted $CO_2$ gave $\delta^{13}C$ =–16.3‰[3]. Thus, an isotopic fractionation occurs in the sample preparation procedure (in the absorption in the Ascarite) since for atmospheric $CO_2$ the $\delta^{13}C$ value is around –8‰. The actual level of $^{14}C$ in Bq/kg is normalised to this $\delta^{13}C$-value.

**TABLE 5.** The $^{14}C$ content in atmospheric $CO_2$ collected at various distances and in the down-wind direction from the Barsebäck nuclear power plant.
Sampling date: 10 July 1996.
Wind speed: 7 m/s.

| Distance (km) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) |
|---|---|---|
| 0.25 | 124 ± 16 | 263 ± 4 |
| 0.70 | 108 ± 25 | 259 ± 6 |
| 3.1 | 145 ± 16 | 268 ± 4 |
| 4.0 | 143 ± 21 | 267 ± 4 |
| 6.1 | 119 ± 16 | 262 ± 4 |
| 9.5 | 102 ± 16 | 258 ± 4 |
| 6.0* | 112 ± 18 | 260 ± 4 |

* Sample collected in a cross-wind direction.

**TABLE 6.** The $^{14}C$ content in atmospheric $CO_2$ from air samples collected at various distances and in the down-wind direction from the Barsebäck nuclear power plant.
Sampling date: 5 December 1997.
Wind speed: 2-5 m/s.

| Distance (km) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) |
|---|---|---|
| 0.30 | 61 ± 17 | 248 ± 4 |
| 0.65 | 97 ± 18 | 257 ± 4 |
| 1.3 | 68 ± 15 | 250 ± 4 |
| 2.8 | -23 ± 23 | 229 ± 4 |

[3] Isotopic fractionation is the enrichment or depletion of one isotope relative to another during the formation of a particular compound in any process, due to different rates of reaction of each isotope. The stable carbon isotope ratio used to study these variations is expressed as parts per thousand deviation from a standard, according to the equation:

$$\delta^{13}C = \left[\frac{R}{R_0} - 1\right] \cdot 1000‰$$

where R is the measured ratio of $^{13}C$ to $^{12}C$ of the sample and $R_0$ is the same ratio for the standard.

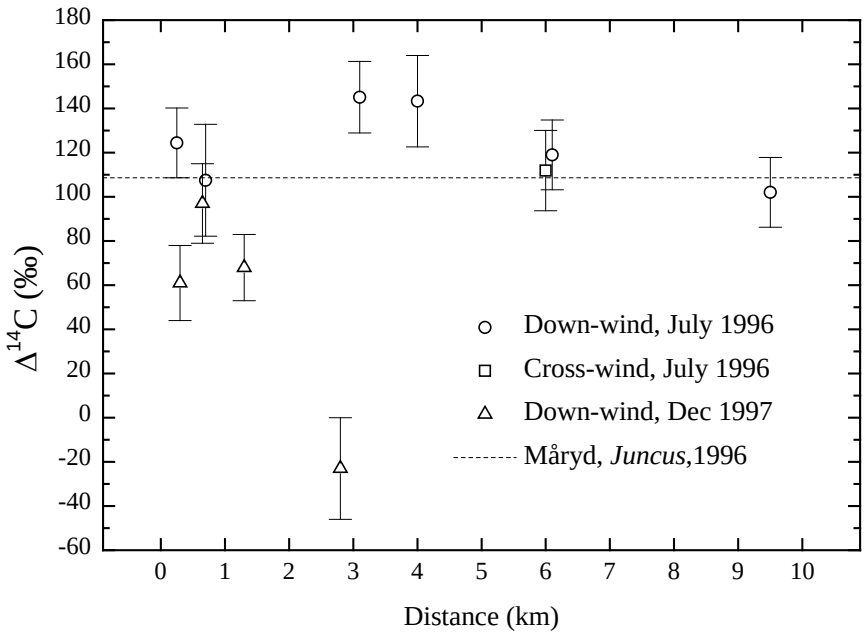


**Fig. 6** $\Delta^{14}C$ values of atmospheric $CO_2$ at various down-wind distances and at one cross-wind site from the Barsebäck nuclear power plant. Sampling: 10 July 1996, wind speed 7 m/s, temperature +16°C; 5 December 1997, wind speed 2-5 m/s, temperature +2°C.

The air samples collected in July 1996 at Barsebäck show elevated $^{14}C$-concentrations at 3 and 4 km from the power plant (Fig. 6). The corresponding excess $^{14}C$ is 36±17‰ and 34±22‰, respectively. The other values in the down-wind direction, as well as the one collected in the cross-wind direction, agree within one standard deviation with the Måryd background data.

The air samples collected at Barsebäck in December 1997 show significantly lower specific activities than Måryd *Juncus*. This is probably due to influence combustion of fossil fuels, as previously discussed.

### *5.3 $^{14}C$ levels at the Forsmark nuclear power facility*

#### *5.3.1 Tree ring and vegatation samples at Forsmark*

The measured activity concentrations of $^{14}C$ in the tree ring and the sallow collected at Forsmark are listed in Table 7. The tree ring sample from 1992 show an excess $^{14}C$ specific activity above the background (Måryd *Juncus*) of 25±5 Bq/kg C, *i.e.* an elevation of 11±2% above the background. The sallow collected at 1.5 and 3.2 km showed excess activities of 28±4 Bq/kg C (11±2% above background) and 6±4 Bq/kg C (2±2% above background), respectively.

**TABLE 7.** The $^{14}C$ content in 1992 year's tree ring of pine (*Pinus*), at 2.5 km south (frequent down-wind direction) of the Forsmark reactors, and in sallow (*Salix caprea*), at 1.5 and 3.2 km south of the plant, collected 5 May in 1994.

| Sample | Year | Distance (km) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) | Måryd *Juncus* (Bq/kg C) | Excess activity (Bq/kg C) |
|---|---|---|---|---|---|---|
| Pine tree ring | 1992 | 2.5 | 253 ± 23 | 283 ± 5 | 258 ± 2 | 25 ± 5 |
| Sallow | 1994 | 1.5 | 247 ± 17 | 282 ± 4 | 254 ± 2 | 28 ± 4 |
| Sallow | 1994 | 3.2 | 149 ± 16 | 260 ± 3 | 254 ± 2 | 6 ± 4 |

#### *5.3.2 Air samples at Forsmark*

The $^{14}C$ content of air samples collected in the down-wind direction at various distances from the Forsmark plant is presented in Table 8 and Fig. 7. The data show no peak at all which might be explained by the weather conditions with almost no wind. The samples, which were collected during the winter season, also show lower $^{14}C$-concentrations than the Måryd clean air, as did the Barsebäck samples collected in the winter.

**TABLE 8.** The $^{14}C$ content in atmospheric $CO_2$ from air samples collected at various distances and in the down-wind direction from the Forsmark nuclear power plant.
Sampling date: 17 March 1996. Wind speed: 2 m/s. The samples were collected in the down-wind direction, *i.e.* south of the two eastly reactors and south of the single westly reactor. The distance between the two easterly reactors and the single westerly reactors about 0.7 km.

| | Distance (km) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) |
|---|---|---|---|
| South of the | 0.5 | 85 ± 14 | 254 ± 3 |
| two easterly | 1.0 | 61 ± 16 | 248 ± 4 |
| reactors | 1.8 | 49 ± 15 | 245 ± 4 |
| South of the | 0.5 | 57 ± 15 | 247 ± 4 |
| single westerly | 1.0 | 93 ± 19 | 256 ± 4 |
| reactor | 1.5 | 83 ± 15 | 253 ± 4 |
| | 2.0 | 80 ± 18 | 253 ± 4 |
| | 3.0 | 89 ± 19 | 254 ± 4 |
| | 4.0* | 84 ± 18 | 254 ± 4 |

* Sample collected in a cross-wind direction.

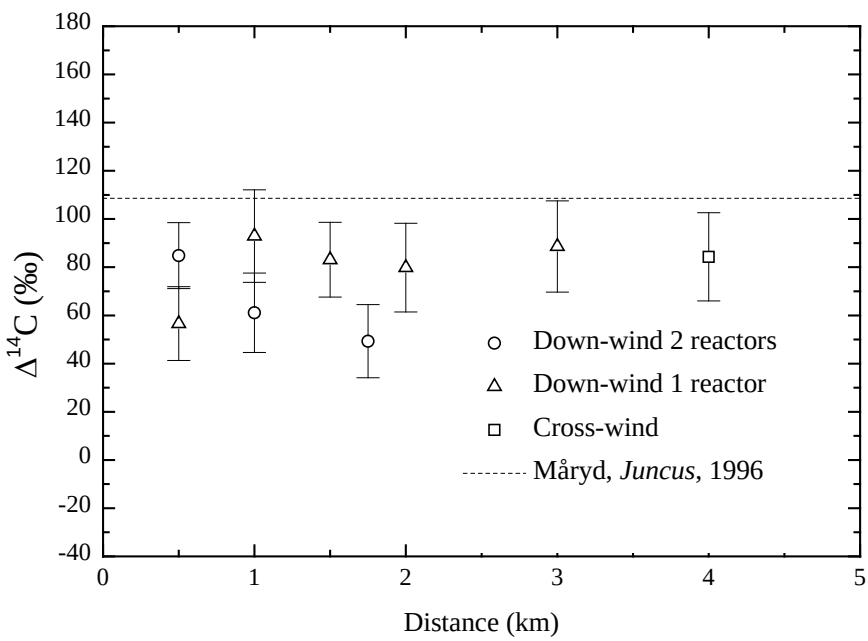


**Fig. 7** $\Delta^{14}C$ values of atmospheric $CO_2$ at various downwind distances from the Forsmark nuclear reactors. Sampling date 3 March 1996; wind speed 2 m/s; temperature 0°C. The 1996 year value of *Juncus* from the "clean air" site Måryd is indicated.

## 6. Comparison of data with the Gaussian plume model

A transfer factor connecting the activity flow rate (Bq/s) of $^{14}C$ in the stack air of the F1 reactor at Forsmark nuclear power station with the activity concentration in the air at the place where the tree rings were collected was calculated using the continuous point source Gaussian plume model (Slade, 1968). The model was also adapted for measurements performed over a longer time period. The activity concentration was integrated over the different wind directions which prevailed during the measurements. A homogeneous distribution of wind directions within the individual wind sector was also assumed. The long-term average concentration of $^{14}C$ is then given by

$$\chi = \sqrt{\frac{2}{\pi}} \frac{fnQ}{2\pi x \sigma_z u} e^{-\frac{h^2}{2\sigma_z^2}}$$

where

- $\chi$ long -term average concentration (Bq/m$^3$)
- $f$ frequency of wind directions into the wind sector
- $\sigma_z$ vertical standard deviation of the concentration distribution (m)
- $n$ number of wind sectors
- $Q$ release rate (Bq/s)
- $x$ distance from source (m)
- $h$ release height (m)
- $u$ mean horizontal air velocity (m/s)

The wind direction data were taken from the weather station Örskär (60$^o$ 32’ N; 18$^o$ 23’ E) 15 km NE of Forsmark and were supplied by the Swedish Meteorological and Hydrological Institute as it was not possible to obtain data from the meteorological mast at Forsmark for the time period in question. The frequency of the wind blowing towards a 60$^o$ sector centred on the tree during the growth season (April to September) was calculated.

In 1992 we investigated the activity concentration of $^{14}C$ in the stack air at F1 (Stenström *et al*., 1995). The results are given in column 2 in Table 9, in column 3 the release rate (Bq/s) for the different time periods, in column 4 the wind frequency *f* into the sector NW-NNW-N-NNE-NE, in column 5 the release rate into the sector (Bq/s), in column 6 the mean wind speed and in column 7 the calculated activity concentration (Bq/m$^3$) in the air 2500 m from Forsmark, assuming category D weather (Slade, 1968). We also assumed that the output from all 3 reactors was the same during the actual time period. A long-term average activity concentration of $18 \cdot 10^{-4}$ Bq/m$^3$ was then obtained.

**TABLE 9.** Parameters related to the $^{14}C$ releases from the F1 reactor in Forsmark in 1992.

| Period | Stack air activity concentration (Bq/m$^3$) | Release rate (Bq/s) | Wind frequency | Release rate into sector ($10^3$ Bq/s) | Wind speed (m/s) | Ground level activity concentration at 2500 m ($10^{-4}$ Bq/m$^3$) |
|---|---|---|---|---|---|---|
| 6/4-21/4 | 123 | 15.7 $10^3$ | 0.53 | 8.3 | 7.9 | 6.27 |
| 21/4-4/5 | 87 | 11.1 | 0.29 | 3.2 | 4.2 | 4.57 |
| 4/5-18/5 | 63 | 8.0 | 0.25 | 2.0 | 6.3 | 1.90 |
| 18/5-1/6 | 85 | 10.8 | 0.21 | 2.3 | 6.3 | 2.18 |
| 1/6-15/6 | 72 | 9.2 | 0.29 | 2.7 | 6.3 | 2.56 |
| 15/6-29/6 | 123 | 15.7 | 0.35 | 5.5 | 6.3 | 5.07 |
| 29/6-13/7 | 85 | 18.8 | 0.34 | 6.4 | 7.5 | 5.89 |
| 13/7-27/7 | 4 | 0.5 | 0.13 | 0.07 | 7.5 | 0.05 |
| 27/-10/8 | 54 | 6.9 | 0.19 | 1.3 | 4.0 | 1.97 |
| 10/8-24/8 | 58 | 7.4 | 0.23 | 1.7 | 7.8 | 1.30 |
| 24/8-7/9 | 58 | 7.4 | 0.28 | 2.1 | 7.8 | 1.58 |
| Mean value | | | | 3.23 | 6.5 | 3.03 |

According to Table 7, the activity concentration of $^{14}C$ was 283±5 Bq/kg C in a tree ring from 1992 in a pine tree (*Pinus*) growing 2500 m from Forsmark power station. The background ("clean air") was supposed to be 258±2 Bq/kg C and was taken from Måryd. Although Måryd is about 600 km from Forsmark it can be used for background calculations because of the small variations over long distances (Stenström *et al.*, 1998).

The transfer factor, $\tau_{F,el}$ connecting the activity concentration of $^{14}C$ in the air, $18.1 \cdot 10^{-4}$ Bq/m$^3$, at the site of the pine tree and the activity concentration of $^{14}C$ in the tree ring, 255 Bq/kg C is 1.38 $10^4$ m$^3$/kg C.

This factor can, in a more general way, be used to calculate the emission of $^{14}C$ from the stacks of a nuclear power plant if the energy production also is taken into account. During the growth season, April to September, of 1992 the reactor F1 produced 2.69 $TWh_{el}$, the reactor F2 produced 2.58 $TWh_{el}$ and the reactor F3 produced 3.13 $TWh_{el}$. The total energy production was 8.40 $TWh_{el}$ and the transfer factor $\tau_F$ per $TWh_{el}$ should be 0.16 $10^4$ m$^3$/kg C per $TWh_{el}$.

As both Forsmark and Barsebäck are equipped with boiling water reactors the Forsmark transfer $\tau_F$ is assumed to be equal to that of Barsebäck $\tau_F = \tau_B$. During the growth season in 1997 (April to September) the reactor B1 produced 1.65 $TWh_{el}$ and the reactor B2 1.78 $TWh_{el}$, together 3.43 $TWh_{el}$, which gives a transfer factor, $\tau_{B,el}$ of $0.56 \cdot 10^4$ m$^3$/kg C.

The activity of $^{14}C$ which is in excess of the normal $^{14}C$ activity and which can be attributed to the Barsebäck plant is very small (Table 3), but for the four years 1987 – 1990 the mean value is 4±4 Bq/kg C. Applying the transfer factor, $\tau_{B,el}$ 0.71 $10^4$ $m^3$/kg C, the activity concentration in the air around the Barsebäck pine trees is 5.6 $10^{-4}$ Bq/$m^3$, which is 1.3% of the normal $^{14}C$ concentration in air and in good agreement with 1.5% which is the excess activity in the pine trees. In order to calculate the release rate from Barsebäck, wind data were taken from the weather station Oskarsgrundet NO which is at a position of 55$^o$ 36$'$ N; 12$^o$ 51$'$. The wind frequency into a 60$^o$ sector centered at the Barsebäck plant was during the growth season 1987 0.33, 1988 0.28, 1989 0.24 and 1990 0.28 with a mean value of 0.28. The mean wind speed was 6 m/s. The release rate for B1+B2 is then 86 kBq/s for the distance 1000 m and 12 kBq/s for 3000 m, with a mean value of (48 ± 30%) kBq/s. According to the monthly report from the Barsebäck nuclear power station the release rate for December 1997 was 23 kBq/s for B1+B2. With regard to the energy production in the two reactors the release rate for B1 ought to be 26 kBq/s and for B2, 22 kBq/s. According to the calculations above, based on activity concentration measurements in the tree rings, the calculated release rate is a factor 2 higher than the reported.

## 7. $^{14}C$ levels at three foreign sites

In order to verify the reliability of the method some investigations have been conducted at two foreign nuclear sites (Sellafield fuel reprocessing plant in England, Pickering nuclear generating station in Canada) where the releases of $^{14}C$ are known to be substantial. Furthermore, results from some measurements in the vicinity of Paldiski submarine training centre in Estonia are presented.

### *7.1 Sellafield, England*

The fuel reprocessing plant Sellafield in the north-west of England is known to release substantial amounts of $^{14}C$. McNamara and McCartney (1998) have estimated that the total release of $^{14}C$ to the atmosphere from this site from the start of operation up to 1995 has been $2{,}4 \cdot 10^{14}$ TBq. Historically, the main part of the $^{14}C$ effluent has been released to the atmosphere. However, since 1994 the $^{14}C$ effluent has been diverted from the aerial to the liquid waste stream, which means that the discharges to the marine environment now exceed the atmospheric discharges.

**TABLE 10.** The $^{14}C$ content in grass samples collected at various distances in the NNE direction from the Sellafield nuclear facility. The distances are relative the Thorp reprocessing facility. Sampling date: 30 September 1996.

| Distance (km) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) |
|---|---|---|
| 1.5 | 991 ± 40 | 447 ± 9 |
| 2.0 | 873 ± 39 | 420 ± 9 |
| 2.5 | 549 ± 39 | 347 ± 9 |
| 3.5 | 403 ± 37 | 315 ± 8 |
| 4.2 | 363 ± 27 | 306 ± 6 |
| 5.3 | 314 ± 19 | 295 ± 4 |
| 5.8 | 237 ± 29 | 278 ± 7 |
| 7.3 | 177 ± 24 | 264 ± 5 |
| 9.2 | 161 ± 21 | 260 ± 5 |
| 11.5 | 202 ± 27 | 270 ± 6 |
| 15 | 76 ± 33 | 241 ± 7 |

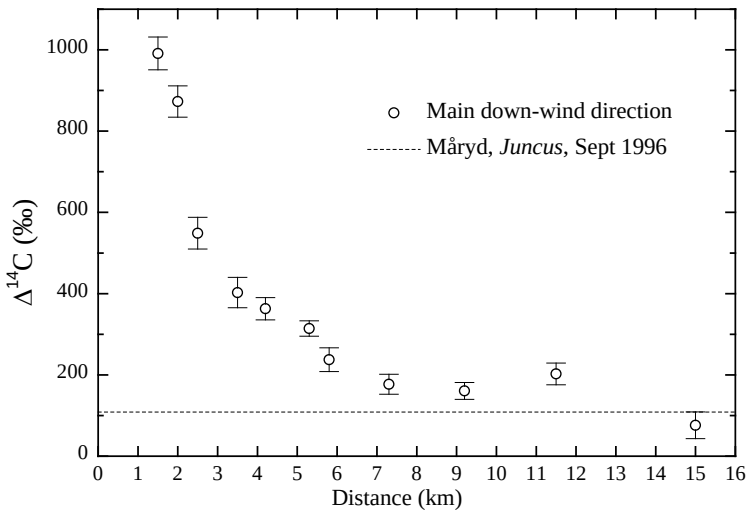


**Fig. 8** $\Delta^{14}C$ values of grass collected at various distances from the Sellafield nuclear site, 30 September 1996.

In Table 10 the $^{14}C$ content in grass samples collected in September 1996 at various distances in the NNE direction from the Sellafield fuel reprocessing plant are presented. The results, obtained by AMS-measurements, are also shown graphically in Fig. 8. The

highest activity, 447±9 Bq/kg carbon, was found at the sample site closest to the facility (at 1,5 km). This is about 80% above the natural level. The results are in the same range as previously found by others (McCartney *et al.*, 1986; Otlet *et al.*, 1990) (see Appendix A).

### *7.2 Pickering, Canada*

Pickering nuclear generating station (PNGS) is one of the world's largest nuclear facilities. It is equipped with 8 units, each 540 $MW_{el}$, of Canadian deuterium uranium (CANDU) pressurized heavy-water reactors (PHWR). Heavy water reactors (HWRs) are known to discharge most $^{14}C$ as $CO_2$ into the atmosphere of all types of nuclear reactors. According to UNSCEAR (1993) the normalised $^{14}CO_2$ release from HWRs during operation is 4800 GBq $(GW_{el}\ a)^{-1}$ (for PWRs the corresponding figure is 120 GBq $(GW_{el}\ a)^{-1}$, and for BWRs 450 GBq $(GW_{el}\ a)^{-1}$).

Four grass samples were collected at various distances from the PNGS in April 1998 and were analysed by AMS. The results are presented in Table 11. The highest activity measured, at about one kilometre north-east of the plant, showed a specific activity of 4820±100 Bq/kg C, *i.e.* about 20 times the natural $^{14}C$ level. Similar results (with even higher activites) have been found by others (Milton *et al.*, 1995) (see Appendix A).

**TABLE 11.** The $^{14}C$ content in grass samples from the vicinity of Pickering nuclear generating station, Canada. Sample date: 980430. Stack height: <20 m (Milton *et al.*, 1995).

| Distance (km) | $\Delta^{14}C$ (‰) | Activity (Bq/kg C) |
|---|---|---|
| 0.7-0.8 E | 1280 ± 30 | 511 ± 7 |
| 0.8-1.0 NE | $(20.5\pm0.5)\cdot10^3$ | 4820 ± 100 |
| 1.5-2.0 NE | 5800 ± 150 | 1530 ± 50 |
| 100 | 191 ± 30 | 268 ± 5 |

### *7.3 Paldiski, Estonia*

At Paldiski submarine training centre in Estonia two PWRs (full power 70 MW and 90 MW) were in operation during 1968-1989 and 1983-1989, respectively. Tree rings in the vicinity of Paldiski were collected in the spring of 1998. The $^{14}C$ content in the samples are presented in Table 12. The data show no significant $^{14}C$ excess activity.

**TABLE 12.** The $^{14}C$ content and excess activity in tree rings in the vicinity of Paldiski Submarine Training Centre, collected in the spring of 1998. Stack height 95 m. Dominant wind direction from west.

| Distance (km) | Year | $\Delta^{14}C$ (‰) | Paldiski tree rings (Bq/kg C) | Måryd/ Barsebäck* "clean air" (Bq/kg C) | Paldiski Excess activity (Bq/kg C) |
|---|---|---|---|---|---|
| 1.5 NE | 1974 | 399 ± 33 | 316 ± 7 | (317 ± 4)* | –1 ± 8 |
| | 1979 | 371 ± 34 | 310 ± 8 | 295 ± 2 | +15 ± 8 |
| 1.4 E | 1974 | 341 ± 46 | 303 ± 10 | (317 ± 4)* | –14 ± 11 |
| | 1979 | 305 ± 40 | 295 ± 9 | 295 ± 2 | 0 ± 10 |
| | 1980 | 260 ± 56 | 285 ± 13 | 291 ± 2 | –6 ± 14 |
| | 1981 | 306 ± 26 | 295 ± 6 | 287 ± 2 | +8 ± 6 |

## 8. Absorbed dose estimates

From information given in this report, about the excess $^{14}C$ specific activity within 3 km from different types of nuclear facilities, the effective dose to man can be calculated as given in Table 13.

**TABLE 13.** Local excess $^{14}C$ specific activity due to releases from various nuclear installations and the related effective dose to the most exposed individual[4]. Values within () according to Milton *et al.*, 1995.

| Site | Excess $^{14}C$ specific activity (Bq/kg C) | Effective dose rate (μSv/year) |
|---|---|---|
| Barsebäck | 2 - 4 | 0.1 – 0.2 |
| Forsmark | 6 - 30 | 0.3 – 1.7 |
| Sellafield | 200 | 11 |
| Pickering | 4600<br>(12 000) | 300<br>(7 000) |
| Natural production | 226 Bq/kg C | 12 μSv/year |

The absorbed dose (mGy) to fat and to bone marrow can be 2-3 times higher than the effective dose (μSv).

[4] The most exposed individual is a person living within 1 to 3 km from the source of the releases. The person is producing all his food locally.

## 9. Concluding remarks

The results of the $^{14}C$ measurements of air, vegetation and annual tree rings around Swedish nuclear power plants show very low enhancements of $^{14}C$, if at all above the uncertainty of the measurements. Even if the accuracy of the measurements of the annual tree rings is rather good (1-2%) the contribution of $^{14}C$ from the reactors to the environment is so small that it is difficult to separate it from the prevailing background levels of $^{14}C$. This is the case for all sampling procedures: in air and vegetation as well as in annual tree rings. Only on a few occasions an actual increase is observed. However, although the calculations in section 6 suffer from rather large uncertainties, the calculated release rate from Barsebäck is in fair agreement with reported release data.

Most of the measurements in this investigation were made on samples from the vicinity of the Barsebäck nuclear power plant. The largest signal of $^{14}C$ is shown in tree ring and vegetation data at the Forsmark plant (because of the higher energy production and thus higher $^{14}C$ releases in Forsmark than in Barsebäck). Thus, by measuring the $^{14}C$ specific activity in annual tree rings in a tree close to the Forsmark plant we believe that the total releases of $^{14}C$ from the Forsmark plant, since the start of operation, can be measured with fair accuracy.

The results of this investigation show that the effective doses to man related to the releases of $^{14}C$ from the Swedish light-water reactors at Barsebäck and Formark are very low, especially compared to the situation at other nuclear installations, such as the fuel reprocessing plant in Sellafield, England, and the heavy-water reactors at Pickering nuclear generating station, Canada.

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## APPENDIX A: Brief literature survey

### 1. LIGHT-WATER REACTORS

**Levin, I., Kromer, B., Barabas, M. and Münnich, K.O. 1988 Environmental distribution and long-term dispersion of reactor $^{14}CO_2$ around two German nuclear power plants. *Health Physics* 54(2): 149-156.**

•Sample material: Atmospheric $CO_2$ and plant material (tree leaves and wheat) from the vicinity of two German BWRs (Philippsburg and Isar/Ohu).

•Atmospheric $CO_2$ collected continuously over one or two weeks (during totally 5 years) 1.75 km from the Philippsburg reactor (900 $MW_e$) show a maximum $^{14}C$ excess concentration of $\Delta^{14}C$(excess)=300±7‰. The long-term average excess amounted to $\Delta^{14}C$(excess)=47±3‰. Plant material at the same site ranged between $\Delta^{14}C$(excess)=0‰ and 120‰. The $^{14}C$ excess concentrations found in tree leaves around the Isar/Ohu reactor (907 $MW_e$) at 1-2 km distance fall into the same range as observed at the Philippsburg reactor.

**Loosli, H.H. and Oeschger, H. 1989 $^{14}C$ in the environment of Swiss nuclear installations. *Radiocarbon* 31(3): 747-753.**

•10 year survey of the $^{14}C$ content in tree leaves (beech) from the vicinity of Swiss nuclear installations.

•In general, maximum concentrations are found between 0.5 and 1.5 in the main wind directions.

•Higher activities around BWRs than around PWRs.

•Excess activities of up to ca 100‰ are observed in the vicinity of nuclear power plants.

•A higher excess of up to 1400‰ is measured in the close vicinity of a small research plant, which releases the activity on top of the roof.

•Additional doses of $^{14}C$ to plants and population from excess $^{14}C$ activities are estimated to be negligible compared to natural doses.

**Obelic, B. Krajcar-Bronic, I., Srdoc, D. and Horvatincic, N. 1986 Environmental $^{14}C$ levels around the 632 $MW_e$ nuclear power plant Krsko in Yugoslavia. *Radiocarbon* 28(2A): 644-648.**

•632 $MW_e$ nuclear power plant Krsko in Slovenia: PWR.

•Sample material: atmospheric $CO_2$ (continuously collected monthly over one year), cereals, vegetables, grass and tree rings (at 1.5 km from the plant). Vegetation samples from 1976-1985.

•Sampling points: 1.5 km, 12 km and 34 km.

•An average excess of (2.2±0.9)% above the reference point activity during normal reactor operation was observed in atmospheric $CO_2$ at 1.5 km from the stack.

•Maximal increase of activity in atmospheric $CO_2$ at 1.5 km of (4.5±2.1)% coincides with the yearly change of 1/3 of the fuel elements in the reactor core.

•Activity measurements of plants show no significant variations and agree, within the error, with the activity of atmospheric $CO_2$ in the northern hemisphere.

•Activity in tree rings show no elevation of the $^{14}C$ activity and follow the general trend in the northern hemisphere.

•Estimated release, based on the average 2.2% excess, is 0.1 TBq/year.

**Uchrin, G., Csaba, E., Hertelendi, E., Ormai P. and Barnabas I. 1992 $^{14}C$ release from a Soviet-designed pressurized water reactor nuclear power plant. *Health Physics* 63(6): 651-655.**

•Paks Nuclear Power Plant in Hungary with four Soviet designed PWR's, each of 440 $MW_e$.

•Yearly discharge releases for the time period 1988-1991 are equal to 0.7 TBq/($GW_e$·year) for hydrocarbon and 0.05 TBq/($GW_e$·year) for $CO_2$.

•Biweekly samples of air (total carbon) showed a average radiocarbon excess of 4% at 1 km from the release point.

•A tree situated about 1.7 km from the 100-m-high stack showed no $^{14}C$ excess higher than the uncertainties of the measurements $\Delta^{14}C=\pm 4‰$ during the measured years 1982-1986. This reflects the low fraction of $^{14}CO_2$ in the releases.

**Uchrin, G., Hertelendi, E., Volent, G., Slavik, O., Morávek, J., Kobal, I. and Vokal B. 1998. Carbon-14 measurements at PWR type nuclear power plants in three middle European Countries. *Radiocarbon* 40:1, 439-446.**

•Environmental air in the vicinity of Paks NPP (Hungary) show a long-term average excess of $^{14}C$ of $D^{14}C$ =50‰ for hydrocarbons.

•Maximum atmospheric excess: 5 mBq/$m^3$ above the natural level of ca 45 mBq/$m^3$.

•Tree ring analysis has shown a slight excess around Krsko NPP (Slovenia): $D^{14}C$=199.9‰ for a tree at 1 km from the NPP compared with a reference tree which has $D^{14}C$=111.6‰ in 1994.

**Veres, M., Hertelendi, E., Uchrin, G., Csaba, E., Barnabás, I., Ormai, P., Volent, G. and Futó, I. 1995. Concentration of radiocarbon and its chemical forms in gaseous effluents, environmental air, nuclear waste and primary water of a pressurized water reactor power plant in Hungay. *Radiocarbon* 37(2): 497-504.**

•Sampling of environmental air ca 2 km from the stack.

•The long-term average excess due to the NPP correspond to $D^{14}C$=3.5‰ for $CO_2$ and $D^{14}C$=20‰ for hydrocarbons.

## HEAVY-WATER REACTORS

**Milton, G.M., Kramer, S.J., Brown, R.M., Repta, C.J.W., King, K.J. and Rao R.R. 1995 Radiocarbon dispersion around Canadian nuclear facilities. Proceedings of the 15th International Radiocarbon Conference, Cook, G. T., Harkness, D.D., Miller, B.F. and Scott, E.M. (eds), *Radiocarbon* 37(2): 485-496.**

•Surveys of atmospheric dispersion of $^{14}CO_2$ at the Chalk River Laboratories (CRL) and Pickering Nuclear Generation Station (PNGS).

•CANDU pressurized heavy-water reactors at PNGS and heavy-water research reactors at CRL (NRX and NRU).

•Main release form $^{14}CO_2$.

•Analysed air vegetation, soil and tree rings.

•PNGS: maximum tree ring activity at 1 km NE of PNGS: 12 Bq/g carbon (1987): Stack height <20 m!

•CRL: stack height 50 m. Grass 1982: maximum twice the natural level. Air 1993: Twice the natural level.

## FUEL REPROCESSING FACILITIES

**Baxter, M.S., McCartney, M., McMay, K. and Scott, E.M. 1984 Artificial carbon-14 in the terrestrial biosphere. Presented at the second meeting of the international atomic agency's co-ordinated research programme on carbon-14 from nuclear facilities - at Bhabha Atomic Research Centre, Trombay, India, 10-14 December 1984.**

•The local effect of $^{14}C$ discharges from Sellafield fuel-reprocessing plant by measurement of plant samples.

•Levels are maximum about four times the natural level.

•Levels are clearly measurable to more than 12 km from the plant.

•The $^{14}C$ activities predicted by the model are in good agreement with the Gaussian plume atmospheric dispersion model.

•Estimated releases from the plume model: 3.7 TBq/year.

**Buzinny, M., Kovalyukh, N., Likhtarjov, I., Los, I., Nesvetajlo, V., Pazdur, M.F., Skripkin, V., Shkvorets, O. and Sobotovich, E. 1995 Ecological chronology of nuclear fuel sites. *Radiocarbon* 37(2): 469-473.**

•Annual tree rings around the Tomsk nuclear fuel reprocessing plant from 8-10 km to 10-15 km southeast and northwest from the plant.

•Levels excceding the “clean air” value by 120 Bq/kg carbon in tree ring 10 km from the plant.

•Grass samples from 1994 collected at 5-6 km southwest (wind direction) yield an excessive $^{14}C$ level of 54.9±4.6 Bq/kg carbon above natural level.

**McCartney, M., Baxter, M. S. and McKay, K. 1986 Global and local effects of $^{14}C$ discharges from the nuclear fuel cycle. *Radiocarbon* 28(2A): 634-643.**

•Sellafield

•Maximum levels of about 350% above the natural level were observed <1 km from the plant, with enhanced activities detectable to at least 12 km.

•The pattern shows that the Gaussian plume model performs successfully.

**Otlet, R.L., Walker, A.J. and Fulker, M.J. 1990 Survey of the dispersion of $^{14}C$ in the vicinity of the UK reprocessing site at Sellafield. *Radiocarbon* 32(1):23-30.**

•Samples: locally grown foodstuffs, tree rings, hawthorn berries.

•Tree 1.6 km NE of the plant, a 100 year survey. Maximum activity about four times the natural level.

## APPENDIX B: Instruction for the preparation of tree ring samples (in Swedish)

### Instruktion för beredning av trädprover

#### 1. Provtagning

Leta upp ett någorlunda stort träd som helst står skilt från andra träd och som har bra växtbetingelser. Tall är ett lämpligt val eftersom årsringarna på tall är relativt lätta att urskilja. Träd såsom pil är inte att föredra eftersom de ofta hamlas och då kan hoppa över en årsring.

När du borrar, se till att du borrar rakt in mot mitten av trädet. När du borrat hela vägen sätt in staven som du tar ut borrkärnan med, vrid borren tillbaka ett halvt varv och dra sedan ut borrkärnan.

Se efter så att det inte är för kort avstånd mellan årsringarna, de kan då vara svåra att separera. Mindre än en mm över lag bör de inte vara. Man behöver då 5-6 borrkärnor från samma träd för att få tillräckligt med material. Är ringarna mycket breda behövs inte lika många borrprov.

Förpacka borrkärnorna väl så att de inte bryts, de kan då vara svåra att identifiera sedan. Skriv upp plats, tidpunkt, trädslag.

#### 2. Räkna årsringar

För att tydligare kunna se årsringarna kan man blötlägga borrkärnan en stund. Se till att provet ligger stadigt mot underlaget och var försiktig så att borrkärnan inte bryts. Det är lättast att urskilja ringarna om borrkärnan placeras så att fibrerna går i vertikal riktning, dvs att borrkärnan läggs på samma håll som den satt i trädet.

Tag en vass kniv eller rakblad och hyvla lite längs med borrkärnan så att en flat yta fås fram. Hyvla endast ytterst lite längs ovansidan! Nu skall man kunna se årsringarna tydligt.

Beroende på årstid kan det se något olika ut längst ut vid barken. De ljusa ringarna är maj-juni ved, medan de mörka vuxit under juli-augusti, de mörka är alltså senare ved. De ljusa partierna är oftast större än de mörka. Om provet tagits under höst-vinter eller vårmånaderna börjar alltså den yttersta ringen med ett mörkt parti. Man ser oftast en skarp gräns mellan mörka och ljusa partier som hör till olika år, de som hör till samma år går mer in i varandra, en mer diffus gräns.

Märk t ex var tionde år med en penna och jämför gärna proverna med varandra, träd inom samma område växer likartat, och årsringarna från samma år skall vara ungefär lika breda.

Tag sedan en liten kniv eller rakblad, och dela upp borrkärnan i år. Skär alltså vid den skarpa gränsen mellan ljusa och mörka partier. Se till att få med allt material som hör till respektive år, så att de inte blandas. Blöt gärna provet lite, så går det lättare att skära. Lägg sedan proverna från respektive år i väl märkta burkar.

***3. Kemisk preparering av proverna***

*Förbehandling (syra-bas-syra):*

1. Finfördela träet till små flisor.

2. Lägg i en bägare, fyll på ca 15 ml 3M saltsyra. Låt det koka i 30 minuter.

3. Häll över i ett filterglas, skölj några gånger med destillerat vatten. Lägg tillbaka träflisorna i bägaren.

4. Fyll på ca 15 ml NaOH lösning, 1M, låt koka i 30 minuter.

5. Filtrera och skölj igen, lägg tillbaka i bägaren och koka i saltsyra som ovan ytterligare 30 minuter. Filtrera, skölj och placera tillbaka i bägaren.

*Blekning till cellulosa:*

1. Fyll ca 16 ml destillerat vatten i bägaren. Värm till 75°C.

2. Tillsätt 0,1 g 80% $NaClO_2$ och därefter 0.14 ml 1M saltsyra. Låt stå i en temperatur mellan 65°C-95°C i 30 minuter. Denna temperatur skall hållas under hela tiden.

3. Tillsätt därefter 0,2 g $NaClO_2$ samt 0,28 ml HCl. Låt stå under 1 timme.

4. Upprepa föregående punkt 3 ggr till, låt därefter stå ytterligare ca 1 timme.

5. Skölj rent provet, lägg det att torka.

*Förbränning och grafitisering:*

Provet förbränns och grafitiseras enligt referens Stenström, 1995d.