Radiocarbon, Vol 65, Nr 6, 2023, p 1343–1350 DOI:10.1017/RDC.2023.125

^{14}C IN TREE RINGS IN THE VICINITY OF THE RBMK REACTOR NUCLEAR POWER PLANT

Evgeniy I Nazarov[1*] • Alexander V Kruzhalov[2] • Maxim E Vasyanovich[1] • Alexey A Ekidin[1] • Maria D Pyshkina[1] • Vladimir V Kukarskikh[3] • Ekaterina V Parkhomchuk[4]

[1]Institute of Industrial Ecology UB RAS, 620108, Ekaterinburg, Russia
[2]Ural Federal University, 620002, Ekaterinburg, Russia
[3]Institute of Plant and Animal Ecology UB RAS, 620144, Ekaterinburg, Russia
[4]Institute of Archeology and Ethnography SB RAS, 630090, Novosibirsk, Russia

ABSTRACT. The paper presents the results of radiocarbon (^{14}C) concentration measurements in tree rings in the vicinity of Kursk NPP (Russia) with four operating RBMK reactors. The sampling was carried out from the site with the highest expected accumulation of radiocarbon in vegetation. The site was determined with long-term meteorological data. The measurements of ^{14}C concentration carried out with accelerator-mass spectrometer in Budker Institute of Nuclear Physics, Novosibirsk, Russia. The obtained results demonstrated the influence of exploitation of Kursk NPP to the concentration of ^{14}C in tree rings. Based on the equilibrium between the ^{14}C ratio in the tree rings and the surrounding air, retrospective estimates of the radiocarbon discharge and effective doses were made. Effective doses were calculated with two approaches: IAEA methodology and less conservative approach, considering the real food consumption in the Kursk region. The values of calculated doses by the second method (0.08–2.58 μSv) are more than 2 times less than IAEA approach (0.17–5.30 μSv). The highest difference between measured and background ^{14}C in tree ring is 41.7 ± 5.8 pMC in 2014 during the restoration of graphite stack. The main contribution to ^{14}C exposure in the considering period is caused by background – from 70 to 99%.

KEYWORDS: carbon-14, effective dose, radioactive discharges, RBMK, retrospective assessment.

INTRODUCTION

Carbon-14 (^{14}C) is a low-energy beta-emitting radionuclide that is produced both naturally and artificially. The natural path is based on the interaction of thermal neutrons and nitrogen-14 atoms in the upper layers of the troposphere and stratosphere: ^{14}N(n,p)^{14}C. About $1.4 \cdot 10^6$ GBq ^{14}C is produces annually by this reaction, while the total amount of ^{14}C in the atmosphere is estimated at $1.4 \cdot 10^8$ GBq (IAEA 2004). Nuclear weapons testing led to an almost twofold increase in the concentration of ^{14}C in the atmosphere. According to estimates, $2.2 \cdot 10^8$ GBq ^{14}C entered the atmosphere in the 1950s and 1960s (IAEA 2004). After acceptance of Partial Nuclear Test Ban Treaty ^{14}C concentration in atmosphere began to decline gradually due to the carbon cycle and is now comparable to pre-test levels ~ 100 pMC ≈ 226 Bq/kg C (IAEA 2004).

At present, atmosphere discharges from nuclear fuel cycle enterprises are the main anthropogenic source of ^{14}C entering the atmosphere. During the operation of a nuclear reactor, ^{14}C is formed mainly as a result of the interaction of thermal neutrons and ^{14}N, ^{13}C, ^{17}O atoms present in fuel elements, structural materials, moderator and coolant, as well as due to uranium and plutonium ternary fission reactions in nuclear fuel (IAEA 2004). It is estimated that about $1.1 \cdot 10^5$ GBq/year is released into the atmosphere in gaseous form from all operating nuclear power plants, while about $3.7 \cdot 10^5$ GBq/year of ^{14}C in both gaseous and liquid form released by spent nuclear fuel reprocessing plants.

In Russia, there are three types of nuclear power reactors: VVER (PWR), FBR and RBMK. In an RBMK ^{14}C is mainly produced in the graphite moderator by ^{13}C(n,γ)^{14}C and ^{14}N(n,p)^{14}C reactions, due to large amounts of impurities. Most of the produced ^{14}C, however, remains in the graphite during the operation of the reactor and is not released to the atmosphere.

*Corresponding author. Email: e.nazarov1005@gmail.com

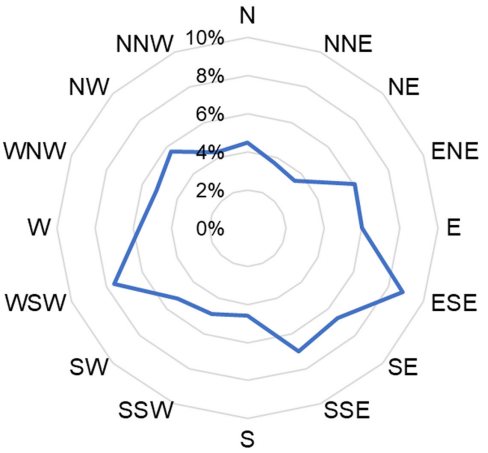

Figure 1 Wind rose in Kurchatov 2006–2022.

Tree rings are a good indicator of the ^{14}C content in the atmosphere due to the process of photosynthesis. The analysis of ^{14}C content in the annual rings of trees located in the vicinity of nuclear facilities allows to perform a retrospective assessment of ^{14}C release and calculate effective doses throughout the entire life cycle of the enterprise. An increased content of ^{14}C was observed after the analysis of tree rings near Paks (Janovics et al. 2013), Ignalina (Ežerinskis et al. 2018), Barseback (Stenstrom et al. 1997), and Beloyarsk (Nazarov et al. 2022) NPPs. The aim of this paper is retrospective annual discharges of ^{14}C by Kursk NPP and effective doses calculation.

SAMPLING AND MEASUREMENTS

Kursk NPP is presented by four RBMK-1000 reactors with an electrical output of 1000 MW. Four reactors were constructed from 1979 to 1986. One of four reactors is permanent shutdown since 2021. The routine release of ^{14}C into the atmosphere is carried out mainly through two ventilation stacks 150 m high and from other organized sources, but with lower concentrations. The sampling site, located 2.7 km west of the geometric center of discharges sources, was defined using long-term meteorological data (Rostekhnadzor 2021). The weather archive since 2006 included more than 50,000 records. The dominant wind direction is east-southeast (Figure 1). Figure 2 presents the location of Kursk NPP and the sampling site.

The core samples of *Pinus sylvestris* L. and *Populus tremula* L., the most representative tree species in the location of Kursk NPP, were sampled using an increment borer at a height of about 130 cm from the soil surface. The age of the trees varied from 50 to 70 years. The annual ring widths on all cores were measured according to standard dendrochronological techniques (Stokes and Smiley 1968; Cook and Kairiukstis 1990; Rinn 1996). We used COFECHA software (Holmes 1983) cross-dating quality control. According to the crossdated series cores were divided into annual rings. Rings of the same age from 10 different individuals were combined into one sample corresponding to a certain year. 14 different samples were selected for analysis. The ^{14}C concentration of a 113-year-old pine tree located in Novosibirsk was used as background (Nazarov et al. 2021) due to the city of Novosibirsk is located at a considerable distance from the operating nuclear facilities.

https://doi.org/10.1017/RDC.2023.125 Published online by Cambridge University Press

Figure 2 The sampling site and Kursk NPP location (numbers of power units).

During the sample preparation, cellulose was extracted from tree rings by the procedure described in details in (Parkhomchuk et al. submitted). In general the procedure consisted in several steps: (1) multistage extraction purification with $C_2H_2Cl_2$, C_2H_5OH and distilled water until a colorless solution was obtained; (2) delignification, which was carried out by the catalytic oxidation by H_2O_2 in an acidic medium with $NaWO_4$ as a catalyst; (3) multistage dehumification with NaOH; and finally (4) acid treatment with HCl for CO_2 removal, distilled water rinsing and drying at 70°C with snow-white cellulose obtaining. Then it was subjected to complete combustion and transformation into graphite-like carbon in the absorption-catalytic setup (Lysikov et al. 2018), with following measurements of ^{14}C concentration. The measurements were carried out on accelerator mass-spectrometer at the AMS Golden Valley laboratory using the facility from Budker Institute of Nuclear Physics (Parkhomchuk and Rastigeev 2011; Nazarov et al. 2021; Petrozhitskiy et al. forthcoming).

METHODS

The used calculation approach assumes that the concentration of ^{14}C in the annual ring of a tree corresponds to the average annual concentration in the ambient air. The sampling site is expected to have the highest concentration of ^{14}C in the air and, as a result, the highest effective dose.

The ^{14}C air concentration A_V (Bq/m^3) was calculated using following formula:

$$A_V = \Delta pMC \cdot S \cdot C_{st}, \quad (1)$$

where ΔpMC – difference between measured and background ^{14}C concentration (Table 1);

S – coefficient between pMC and Bq/kg C. 100 pMC = 226 Bq/kg C;

C_{st} – stable carbon concentration in air. The concentration of stable carbon was determined using carbon dioxide levels in atmosphere (NASA 2023). The carbon dioxide concentration

https://doi.org/10.1017/RDC.2023.125 Published online by Cambridge University Press

Table 1 Concentrations of ^{14}C in tree rings and ^{12}C in air.

Year	^{14}C concentration (pMC)		Stable carbon in air concentration (g C/m^3)
	Kursk NPP	Background	
1976	145.7 ± 1.2	134.8 ± 1.3	0.173
1980	134.8 ± 0.7	126.5 ± 3.2	0.177
1984	128.7 ± 0.7	125.3 ± 2.8	0.180
1986	125.7 ± 0.8	121.9 ± 3.3	0.182
1988	124.0 ± 0.7	115.2 ± 1.8	0.184
1990	121.8 ± 0.7	114.8 ± 2.3	0.185
1994	116.8 ± 1.1	112.0 ± 2.2	0.187
1998	112.7 ± 0.7	109.9 ± 1.5	0.192
2002	112.4 ± 0.8	105.7 ± 2.1	0.195
2006	117.8 ± 0.7	103.2 ± 1.2	0.200
2010	107.7 ± 0.7	102.5 ± 0.9	0.204
2014	143.5 ± 5.8	101.8 ± 0.3	0.208
2018	136.7 ± 2.7	100.7 ± 0.4	0.213
2020	101.3 ± 0.7	100.0 ± 0.4	0.216

Table 2 Dilution factor values for each year.

Year	1976–2002	2006	2010	2014	2018	2020
$G_{n,j,}$ 10^{-8} s/m^3	4.74	5.44	6.06	7.45	5.65	4.95

varied from 332 ppm in 1976 to 414 ppm in 2020. The values of C_{st} used in calculations are presented in Table 1.

The estimate of the annual ^{14}C discharge Q (Bq/a) was calculated by formula:

$$Q = 3.15 \cdot 10^7 A_V / G_{n,j}, \tag{2}$$

where $3.15 \cdot 10^7$ – the conversion factor, s/a;

$G_{n,j}$ – the annual average meteorological dilution factor in the surface layer of the atmosphere at a distance x from the i-th source in the wind direction of the n-th point, s/m^3 (Figure 1). For sampling site, the values were calculated for each year, where it possible, by analyzing the meteorological data (Table 2).

The IAEA approach for estimating annual effective doses from ^{14}C releases assumes that all the food consists only of local products and, as a result, there is an equilibrium in the ^{14}C concentration in food, human tissue, and atmospheric air. Then the estimate of the expected annual dose of internal exposure D (Sv/a) from ^{14}C contained in the atmospheric air in the form of carbon dioxide $^{14}CO_2$ can be obtained by the following formula (IAEA 2001):

$$D = A_V G / C_{st}, \tag{3}$$

https://doi.org/10.1017/RDC.2023.125 Published online by Cambridge University Press

Table 3 Parameters of products consumption in Kursk region.

Product	R_i (kg)[a]	α_i (%)[a]	$f_{p,i}$ (kg C/kg)[b]
Milk	291.7	34.3	0.065
Meat	93.7	45.7	0.2
Bread	89.5	50.0	0.39
Potato	103.4	90.7	0.046
Vegetables	101.9	89.3	0.059
Fruit	81.2	93.6	0.062

[a]Rostekhnadzor (2021).
[b]IAEA (2012).

where G – is the effective dose rate factor that relates the annual dose rate (Sv/a) to the concentration of ^{14}C per gram of carbon in people (Bq/g). The dose rate factor recommended for screening is $5.6{\cdot}10^{-5}$ Sv/a per Bq/g (IAEA 2001).

Since ingestion is the primary mode of expose, and the share of consumption of local products does not always exceed the share of imported, it is reasonable to assume that there may not be an equilibrium of ^{14}C. In this case, the expected annual dose of ^{14}C exposure can be calculated using the formula (Kryshev et al. 2020):

$$D = \varepsilon_{ing} U A_V + \varepsilon_{food} \sum_i \alpha_i R_i A_{food,i}, \quad (4)$$

where $\varepsilon_{ing} = 6.5{\cdot}10^{-12}$ Sv/Bq – dose conversion factor for an adult when inhaling ^{14}C in the form of CO_2 (ICRP 2012);

$U = 8.1{\cdot}10^3$ m^3/a – respiratory rate of an adult (Pleil et al. 2021);

$\varepsilon_{food} = 5.8{\cdot}10^{-10}$ Sv/Bq – dose conversion factor for dietary intake of ^{14}C for an adult (IAEA 2014);

α_i – the share of local products in the diet of the population (Table 3);

R_i – annual consumption of the product by the population, kg (Table 3);

$A_{food,i}$ – ^{14}C concentration in the local food product, Bq/kg.

The ^{14}C concentration in the food product, based on the balance of ^{14}C between atmospheric air and local plant or animal products, was calculated by the formula:

$$A_{food,\,i} = A_V f_{p,i} / C_{st}, \quad (5)$$

where $f_{p,i}$ – the share of stable carbon in plant or animal product, kg C/kg product (Table 3).

RESULTS AND DISCUSSIONS

The ^{14}C concentration in tree rings near Kursk NPP are presented in Figure 3. The launch of four RBMK-type reactors was carried out from 1977 to 1986. Graphite stack restoration terms 2013–2019.

https://doi.org/10.1017/RDC.2023.125 Published online by Cambridge University Press

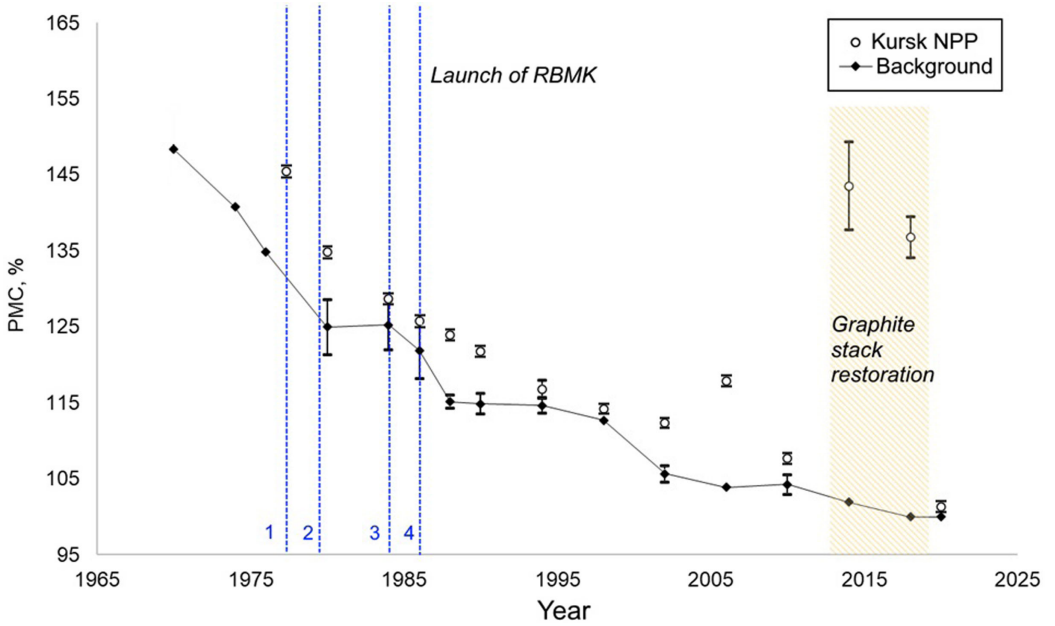

Figure 3 ^{14}C concentration in tree rings.

In all measured samples, the contribution of emissions from the Kursk NPP to the concentration of ^{14}C in the atmospheric air is visible. The highest annual concentrations are observed during the restoration of graphite stack. This confirms that the main part of the produced ^{14}C remains in the graphite. There is no correlation between annual electricity generation and ^{14}C emissions. The annual discharges and dose calculation results are presented in Table 4.

Since it is assumed that a major part of ^{14}C released from RBMKs is in $^{14}CO_2$ form, considering of others chemical forms of ^{14}C is not required. The values of annual discharges vary from 0.4 TBq in 2020 (regular operation) to 9.9 TBq in 2018 during the restoration of graphite stack. The results of effective dose calculation have demonstrated that contribution of inhalation to ^{14}C exposure is about 0.03% while ingestion remains the main way of ^{14}C influence. However, the contribution of background ^{14}C to the annual effective dose is more than 90%, except for the period of restoration of the graphite stack. If we use an approach that considers the actual consumption of products in the Kursk region, the values of annual effective doses are more than 2 times lower than with the conservative approach of the IAEA.

The selection of tree rings from specific years was made to cover both the period of normal operation and the period of restoration of the graphite stack. During normal operation, in our opinion, it is not necessary to consider the tree ring of each year since ΔpMC changes insignificantly. Probably the period of restoration of the graphite masonry requires more detailed consideration. However, in our paper, we demonstrated the scale of ^{14}C release during such process.

https://doi.org/10.1017/RDC.2023.125 Published online by Cambridge University Press

Table 4 The values of average annual discharge and effective dose.

Year	Annual discharge (10^{12} Bq)	Annual effective dose (μSv)			Background ^{14}C contribution to effective dose (%)
		IAEA approach	New approach		
			Ingestion	Inhalation	
1976	2.8	1.39	0.67	$2.24 \cdot 10^{-4}$	92.5
1980	2.6	1.05	0.51	$1.67 \cdot 10^{-4}$	93.9
1984	0.9	0.43	0.21	$7.00 \cdot 10^{-5}$	97.4
1986	1.1	0.48	0.23	$7.90 \cdot 10^{-5}$	97.0
1988	2.4	1.13	0.55	$1.86 \cdot 10^{-4}$	92.8
1990	1.9	0.88	0.43	$1.46 \cdot 10^{-4}$	94.3
1994	0.6	0.57	0.28	$9.59 \cdot 10^{-5}$	96.2
1998	0.4	0.35	0.17	$6.10 \cdot 10^{-5}$	97.6
2002	2.0	0.40	0.19	$6.96 \cdot 10^{-5}$	97.1
2006	3.7	1.86	0.90	$3.34 \cdot 10^{-4}$	87.6
2010	0.8	0.66	0.32	$1.21 \cdot 10^{-4}$	95.2
2014	8.3	5.30	2.58	$9.89 \cdot 10^{-4}$	70.9
2018	9.9	4.58	2.23	$8.75 \cdot 10^{-4}$	73.7
2020	0.4	0.17	0.08	$3.20 \cdot 10^{-5}$	98.7

CONCLUSION

In this paper, we analyzed the concentration of ^{14}C in tree rings in the vicinity of the Kursk NPP. Accelerator mass spectrometry measurements demonstrated $\Delta^{14}C$ excesses in each of the samples, which are due to the operation of four RBMK reactors: from 1.3 pMC in 2020 (normal operation) to 41.6 pMC in 2014 during the restoration of the graphite stack. The assumption that the main part of ^{14}C is emitted as $^{14}CO_2$ made it possible to perform a retrospective assessment of annual releases and effective doses. The release value ranged from 0.4 to 9.9 TBq. Accounting for real consumed products in the Kursk region reduced the values of annual effective doses by more than 2 times compared to the IAEA approach: 0.17–5.30 μSv (IAEA) and 0.08–2.58 μSv (new approach). Background contribution is the main part of ^{14}C exposure—from 70 to 99%

REFERENCES

Cook ER, Kairiukstis LA, editors. 1990. Methods of dendrochronology: applications in the environmental sciences. Dordrecht, Netherlands: Kluwer Academic Publishers.

Ežerinskis Ž, Šapolaitė J, Pabedinskas A, Juodis L, Garbaras A, Maceika E, Remeikis V. 2018. Annual variations of ^{14}C concentration in the tree rings in the vicinity of Ignalina Nuclear Power Plant. Radiocarbon 60(4):1227–1236.

Holmes RL. 1983. Computer-assisted quality control in tree-ring dating and measurement. Tree-Ring Bulletin 43:69–78.

IAEA. 2001. Generic models for use in assessing the impact of discharges of radioactive substances to the environment. Safety Reports Series 19: 143–144.

IAEA. 2004. Management of waste containing tritium and carbon-14. Technical Reports 421: 11–28.

IAEA. 2012. Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environments. Technical Reports 472:139–141.

IAEA. 2014. Safety standards. Radiation protection and safety of radiation sources: international basic safety standards. General Safety Requirements Part 3:137.

ICRP. 2012. Compendium of dose coefficients based on ICRP Publication 60. Publication 119. 59.

Janovics R, Kern Z, Güttler D, Wacker L, Barnabás I, Molnár M. 2013. radiocarbon impact on a nearby tree of a light-water VVER-type nuclear power

https://doi.org/10.1017/RDC.2023.125 Published online by Cambridge University Press

plant, Paks, Hungary. Radiocarbon 55(2): 826–832.

Kryshev AI, Kryshev II, Vasyanovich ME, Ekidin AA, Kapustin IA, Murashova EL. 2020. Population irradiation dose assessment for ^{14}C emissions from NPP with RBMK-1000 and EGP-2 reactors. Atomic Energy 128:53–59.

Lysikov AI, Kalinkin PN, Sashkina KA, Okunev AG, Parkhomchuk EV, Rastigeev SA, Parkhomchuk VV, Kuleshov DV, Vorobyeva EE. 2018. Novel simplified absorption-catalytic method of sample preparation for AMS analysis designed at the Laboratory of Radiocarbon Methods of Analysis (LRMA) in Novosibirsk Akademgorodok. International Journal of Mass Spectrometry 433:11–18.

NASA. 2023. Carbon dioxide. URL: <https://climate.nasa.gov/vital-signs/carbon-dioxide/>

Nazarov EI, Kruzhalov AV, Ekidin AA, Vasyanovich ME, Parkhomchuk VV, Rastigeev SA, Kalinkin PN, Parkhomchuk EV. 2021. Instruments and methods for measuring ^{14}C (a review). Instruments and Experiments Technics 64:790–795.

Nazarov EI, Kruzhalov AV, Vasyanovich ME, Ekidin AA, Kukarskikh VV, Parkhomchuk EV, Petrozhitskii AV, Parkhomchuk VV. 2022. ^{14}C in tree rings in the vicinity of the nuclear facility deployment areas. Nuclear Energy and Technology 8(3):173–177.

Parkhomchuk EV, Petrozhitskiy AV, Ignatov MM, Kuleshov DV, Lysikov AI, Okunev AG, Babina KA, Parkhomchuk VV. Submitted. ^{14}C GIRI samples in AMS Golden Valley: graphite preparation using AGE-3 and absorption-catalytic setup. Radiocarbon.

Parkhomchuk VV, Rastigeev SA. 2011. Accelerator mass spectrometer of the center for collective use of the Siberian Branch of the Russian Academy of Sciences. Journal of Surface Investigation 5(6):1068–1072.

Petrozhitskiy AV, Parkhomchuk EV, Ignatov MM, Kuleshov DV, Kutnyakova LA, Konstantinov ES, Parkhomchuk VV. Forthcoming. Comparative features of BINP AMS and MICADAS facilities working at AMS Golden Valley, Russia. Radiocarbon.

Pleil JD, Ariel Geer Wallace M, Davis MD, Matty CM. 2021. The physics of human breathing: flow, timing, volume, and pressure parameters for normal, on-demand, and ventilator respiration. Journal of Breath Research 15(4).

Rinn F. 1996. Brochure over products and applications. Heidelberg: Frank Rinn Distribution.

Rostekhnadzor. 2021. Safety Guidelines for the Use of Atomic Energy. Recommended Methods for Calculating the Parameters Necessary for the Development and Establishment of Standards for Maximum Permissible Emissions of Radioactive Substances into the Atmosphere. RB-106-21.

Stenstrom K, Skog G, Thornberg C, Erlandsson B, Hellborg R, Mattsson S, Persson P. 1997. ^{14}C levels in the vicinity of two Swedish nuclear power plants and at two "clean-air" sites in southernmost Sweden. Radiocarbon 40(1):433–438.

Stokes M, Smiley T. 1968. An introduction to tree-ring dating. University of Chicago Press.

https://doi.org/10.1017/RDC.2023.125 Published online by Cambridge University Press