^{14}C ANALYSIS OF ANNUAL TREE RINGS FROM THE VICINITY OF THE CHERNOBYL NPP

MICHAEL BUZINNY, ILJA LIKHTAREV, IVAN LOS', NIKOLAY TALERKO and NIKOLAY TSIGANKOV

Research Center for Radiation Medicine, Academy of Medical Sciences of Ukraine, Melnikova 53, Kiev 254050 Ukraine

ABSTRACT. Samples of >40 pine trees were collected from around the Chernobyl NPP for radiocarbon measurement, to determine the spatial distribution of excessive ^{14}C in tree rings from 1986 consequent upon accidental radiocarbon release. Tree samples were collected during 1995–1996 from sites situated at distances >2.5 km from the NPP and covering a variety of directions in relation to the NPP. To evaluate $\Delta^{14}C$ for 1986 annual rings, we compared ^{14}C levels for separate 1985–1987 annual rings, taking into account the trend of operational releases. Early and late wood samples for 1986 annual tree rings were measured separately to increase the sensitivity and precision of measurements. The maximum value observed for excessive accidental radiocarbon levels ($\Delta^{14}C$) was found to be 124 pMC (281.6 Bq kg^{-1} C). We present $\Delta^{14}C$ values for examined sites; their spatial distribution shows a high irregularity of atmospheric ^{14}C depending on direction from the NPP. Using obtained data, we reconstruct the temporal behavior of ^{14}C release during the Chernobyl accident with the aid of atmospheric transport modeling. The total amount of ^{14}C released from 26 April to 5 May 1986 has been estimated as 44 TBq.

INTRODUCTION

Radiocarbon is one of the long-lived radionuclides ($T_{1/2}$ = 5730 yr) of the Chernobyl NPP accidental release. It can be used as a relative tracer for reconstruction of radioactive release and spatial distribution of ^{14}C as well as other radionuclides around the Chernobyl NPP.

The main forms of ^{14}C in the Chernobyl accidental release are as gases (CO_2) and aerosols, mostly graphite dust. We have paid most of our attention until now to the issue of gaseous releases (Buzinny *et al.* 1993a, 1993b, 1995). ^{14}C in tree rings is a prospective tracer useful for a long period after the accident. We assume that spatial distribution data for excessive ^{14}C can be used for retrospective reconstruction of level and time variation of accidental gaseous releases. These data are most significant for zones where ordinary modeling could not be used due to local heterogeneity of releases and fallout.

Radiocarbon accumulation during NPP operation causes significant activity levels in graphite of up to 2.5×10^5 Bq g^{-1}, which corresponds to 128 TBq in all graphite, *ca.* 1800 metric tons (Buzinny *et al.* 1992). ^{14}C-containing CO_2 diffuses into the air, assimilates with plants and forms the trace corresponding to the cumulative impact of gaseous release. Only tree rings were usable for reconstructing excess levels and behavior of $^{14}CO_2$; other samples could not be used due to surface contamination with aerosols and the lack of acceptable sample weight at a variety of sites (Buzinny *et al.* 1995).

Other samples with potential utility for ^{14}C estimation and aerosol transport modeling are graphite aerosols that are transferred and deposited on the land surface (*e.g.,* forest litter may serve as an accumulation system). However, the analysis and interpretation of forest litter is complicated owing to ^{14}C intake from different sources: pine bark and needles from many different years (including 1986), as well as graphite.

The data obtained for ^{14}C levels may be used for the reconstruction of the temporal behavior of the radionuclide releases during the initial stage of the Chernobyl accident. Solving the inverse problem of ^{14}C atmospheric transport leads to a more precise estimation of the radioactivity source characteristics, including the total amount of ^{14}C released as a result of the accident.

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. 373–379

METHODS

Tree samples from 44 trees were collected during 1995–1996 from sites situated at distances >2.5 km from the NPP and covering a variety of directions in relation to the NPP. (At that time, all the trees at distances of <2.5 km had been cut due to their high level of radioactive contamination.) Our sampling network covered almost all the territory of the exclusion zone (*i.e.*, the uninhabited region) surrounding the Chernobyl NPP. Because highly exposed trees have barely defined annual rings for 1986–1989, they could not be dated. (This confirmed our earlier conclusion that it is necessary to use only trees with well identified rings and to have a sufficient mass of sampling material.)

We provide ^{14}C determinations for tree rings using a modern liquid scintillation counting (LSC) based method. To maximize sensitivity and precision, we used separate samples of early and late wood for 1986 annual ring for some of the trees examined (Buzinny *et al.* 1995) following Olsson *and Possnert* (1992). We found earlier that excessive ^{14}C caused by the accident was recorded in early wood of the 1986 ring (Buzinny *et al.* 1995). In that case, ^{14}C concentration for the total annual ring is defined according to the formula

$$^{14}C = \frac{^{14}C_{early} m_{early} + {}^{14}C_{late} m_{late}}{m_{early} + m_{late}}, \quad (1)$$

where

$^{14}C_{early}$, m_{early} = ^{14}C specific activity and mass for the early wood sample;
$^{14}C_{late}$, m_{late} = ^{14}C specific activity and mass for the late wood sample.

This is especially useful for samples with a small observed value of $\Delta^{14}C$.

A Soxhlet apparatus was used for sample treatment with ethanol-benzene mixture following Arslanov (1987). Chemical equipment and related technology developed by V. V. Skripkin was used, combined with LSC measurement based on a Quantulus 1220™ utilizing Teflon® vials with volumes of 7, 3 and 0.8 mL (Buzinny and Skripkin 1995). Sample preparation includes addition of two scintillation materials, 4 g liter^{-1} PPO and 0.1 g liter^{-1} POPOP. Calibration of each LSC vial type includes quench-curve fitting. Additionally we purify benzene to standardize sample preparation. We dated annual tree rings for 1985, 1986 and 1987 for each tree analyzed to determine excessive ^{14}C levels. "Excessive ^{14}C level" was defined as specific activity of the 1986 ring exceeding the average level determined for 1985 and 1987 specific activities.

For calculations of the atmospheric ^{14}C concentration in the 30-km zone around the Chernobyl NPP during the first 10 days after the Chernobyl accident, we used the mesoscale atmospheric transport model LEDI (Buikov, Garger and Talerko 1992). Standard meteorological data obtained from radiosounding measurements were used for the atmospheric transport calculations. Meteorological data included wind velocity and direction, temperature and pressure in a surface layer up to 2000 m. These data were updated every 6 h. During the first days after the accident, the characteristics of the radioactive release changed considerably, so we took into account the temporal variations of ^{14}C-release rate and initial height of radioactivity rise along with the temporal changes of meteorological conditions. (The Gaussian plume models usually used for short-range transport modeling are inapplicable in this case because of the large values for the initial radioactive plume-rise height (up to 1500 m)).

To assess the ^{14}C release intensity in the period from 26 April to 5 May 1986, the inverse problem of reconstructing the time-dependent source has been solved. For this purpose the 10-day period after

https://doi.org/10.1017/S0033822200018257 Published online by Cambridge University Press

the accident was divided into short time intervals (usually 1 h) and the nonstationary continuous release during this period was approximated by a set of instantaneous emissions every hour. Additionally, the time-variability of a height distribution of radioactivity in each of these emissions was taken into consideration. We assumed in the calculations that each emission consists of a set of "elementary" sources with initial heights of 200, 500, 800, 1000 and 1200 m. Using the atmospheric transport model, we calculated the field of a time-integrated near-ground ^{14}C volume concentration (Bq sec m^{-3}) within the 30-km zone near Chernobyl NPP formed by each elementary source, and estimated the additional ^{14}C concentration in vegetation caused by the 1986 accident. The resulting contamination field was determined as a linear combination of contributions of all elementary sources distributed in time and in height. The contribution of each elementary source can be determined by minimizing the differences between observed and calculated ^{14}C concentration fields using a conjugate gradient method (Vakulovski, Shershakov and Golubenkov 1993). This makes it possible to reconstruct both the intensity of the ^{14}C release as a function of time during period of 26 April to 5 May 1986 and the time-variability of initial height of the accidental release from the reactor.

Results

The use of geographical coordinates allows us to display the sampling network (Table 1, Fig. 1) as well as the spatial distribution of excessive ^{14}C (Fig. 2). The maximum level determined for specific activity of a 1986 annual ring for referenced trees was 583 Bq kg^{-1} C, and the maximum level for excessive ^{14}C reached 281.6 Bq kg^{-1} C. It is established that the highest levels of excessive ^{14}C are observed for directions west-northwest and northeast from the Chernobyl NPP (Fig. 2).

TABLE 1. ^{14}C Accidental Excess in Pine Trees in the Vicinity of the Chernobyl NPP

Sample	Longitude, N			Latitude, E			$\Delta^{14}C$ (Bq kg^{-1} C)	Sample	Longitude, N			Latitude, E			$\Delta^{14}C$ (Bq kg^{-1} C)
	°	′	″	°	′	″			°	′	″	°	′	″	
93_80	30	5	11	51	17	40	73.8	96_15	29	55	40	51	25	15	20.7
95_04	30	1	19	51	22	33	170.8	96_16	30	0	30	51	25	30	41.9
95_05	29	59	38	51	22	39	142.4	96_17	30	7	25	51	21	50	203.0
95_06	29	55	23	51	22	20	51.0	96_18	30	9	40	51	19	12	55.2
95_07	29	50	50	51	23	2	37.3	96_19	30	6	40	51	16	24	37.6
95_08	29	45	59	51	23	6	30.1	96_20	30	0	0	51	18	0	10.3
95_09	29	42	16	51	23	13	42.5	96_21	29	53	27	51	17	24	13.5
95_10	30	8	26	51	12	0	9.4	96_22	29	47	54	51	17	24	8.6
95_11	30	10	10	51	14	50	30.7	96_23	29	47	15	51	19	55	23.8
95_12	30	7	40	51	6	55	9.0	96_24	29	51	20	51	21	16	20.0
96_01	30	1	20	51	24	10	18.7	96_25	29	51	20	51	25	30	63.8
96_02	30	1	20	51	22	25	142.2	96_26	29	47	15	51	25	40	31.6
96_03	30	0	30	51	20	40	55.5	96_27	29	50	15	51	24	30	32.1
96_04	30	5	15	51	22	40	144.5	96_28	29	54	18	51	23	3	21.2
96_05	30	8	20	51	29	30	66.8	96_30	30	26	10	51	15	30	5.2
96_08	30	8	30	51	26	50	89.8	96_32	30	17	43	51	18	0	26.0
96_09	30	10	0	51	25	15	103.9	96_33	30	3	38	51	23	25	85.4
96_10	30	12	15	51	23	50	41.1	96_34	30	4	30	51	23	26	222.8
96_11	30	12	50	51	21	40	22.1	96_35	29	38	40	51	17	40	5.9
96_12	30	14	20	51	19	25	18.4	96_36	30	4	30	51	24	0	84.9
96_13	30	4	0	51	23	25	281.6	96_37	30	2	3	51	24	7	41.2
96_14	29	57	50	51	27	10	11.1	96_38	30	8	22	51	20	10	83.6

https://doi.org/10.1017/S0033822200018257 Published online by Cambridge University Press

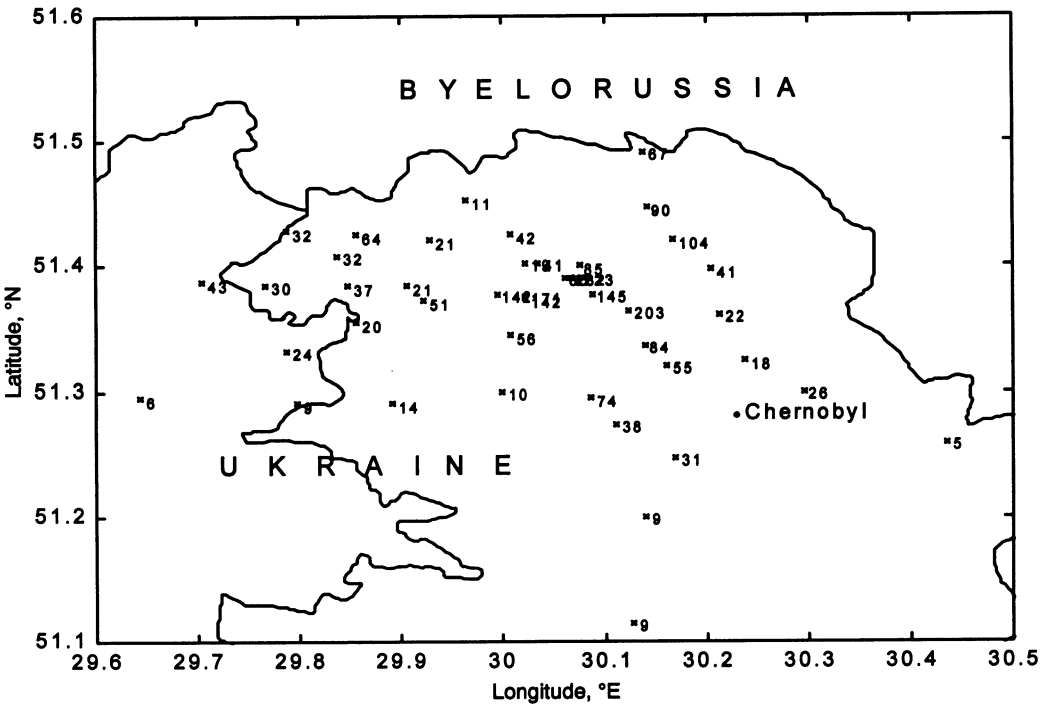

Fig. 1. Tree sampling scheme (60 x 60 km) with $\Delta^{14}C$ data (Bq kg^{-1} C)

The spatial ^{14}C distribution in tree rings for a 30-km zone around the Chernobyl NPP caused by the radioactive accident, reconstructed using atmospheric transport modeling, is shown in Figure 3. There are three main traces in the deposition field: the west and northwest trace formed during 26 April, the east-north one was caused by the intensive radioactive release during 27–28 April, and the south one was caused by the radioactive release during the period from 30 April to 5 May.

Salonen (1987) reports the release rate of ^{14}C for graphite-moderated reactors (RBMK-1000) available at the Chernobyl NPP to be as high as 33000 GBq/GW(e)y. Because the value of average combustion equals 10.9 MW(e) day × kg^{-1} (Buzulukov and Dobrynin 1993) and the period of reactor operation 570 days, the Chernobyl reactor core inventory for ^{14}C may be estimated as 187 TBq.

The ^{14}C daily release amounts during the first 10 days after the Chernobyl accident are reconstructed using atmospheric transport modeling (Fig. 4). The temporal variations of ^{14}C release are similar to the estimates for other volatile radionuclides (Borzilov and Klepikova 1993). The maximum release intensity was during the first 3 days, then the intensity decreased and the second maximum took place on 5 May. Despite a decrease in release intensity after 29 April, the calculations of deposition fields result in the formation of a spot to the south of Chernobyl NPP due to considerable lowering of effective release height. According to these results, the total ^{14}C release during the Chernobyl accident is estimated at 44 TBq, *i.e.,* about 24% of the value obtained for the inventory of the reactor core involved in the accident.

Comparison of the spatial $\Delta^{14}C$ distributions presented in Figures 2 and 3 shows that the calculation results are in rather good agreement with experimental data, especially for the territory to the south of the Chernobyl NPP. Results of some new measurements in the north and east-north parts of the

https://doi.org/10.1017/S0033822200018257 Published online by Cambridge University Press

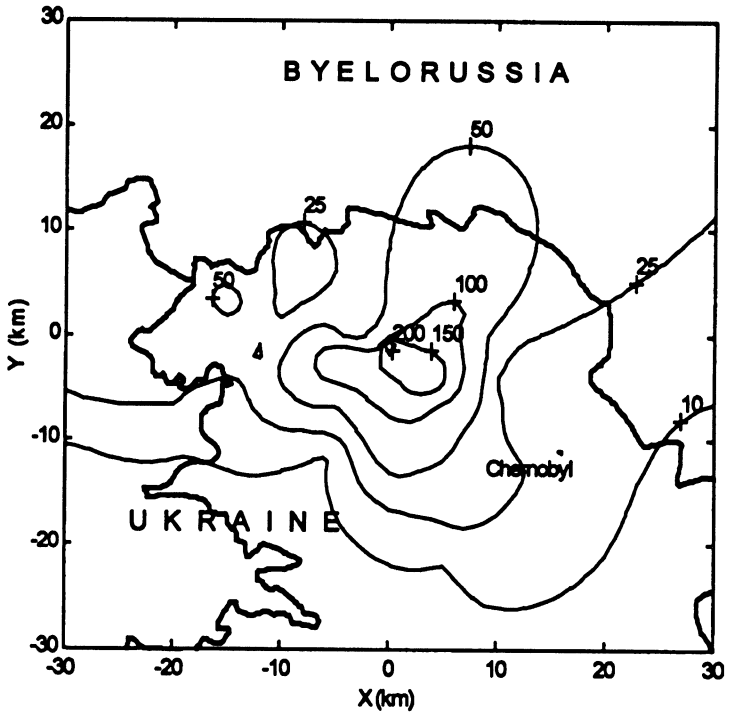

Fig. 2. Spatial $\Delta^{14}C$ distribution (Bq kg^{-1}) for tree rings in the vicinity of the Chernobyl NPP

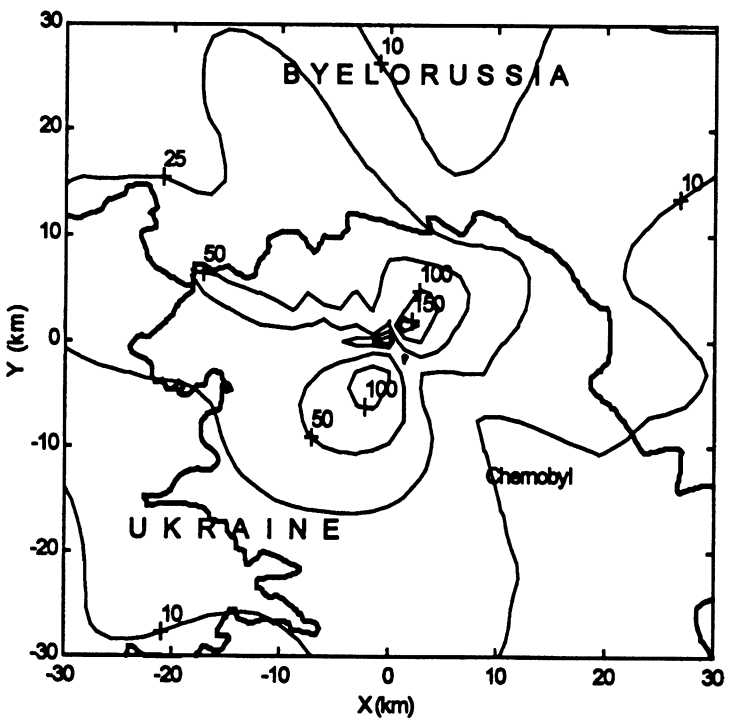

Fig. 3. Isopleths of spatial accidental ^{14}C distribution in 30-km zone around the Chernobyl NPP calculated using atmospheric transport modeling. Contour values are Bq kg^{-1} C.

https://doi.org/10.1017/S0033822200018257 Published online by Cambridge University Press

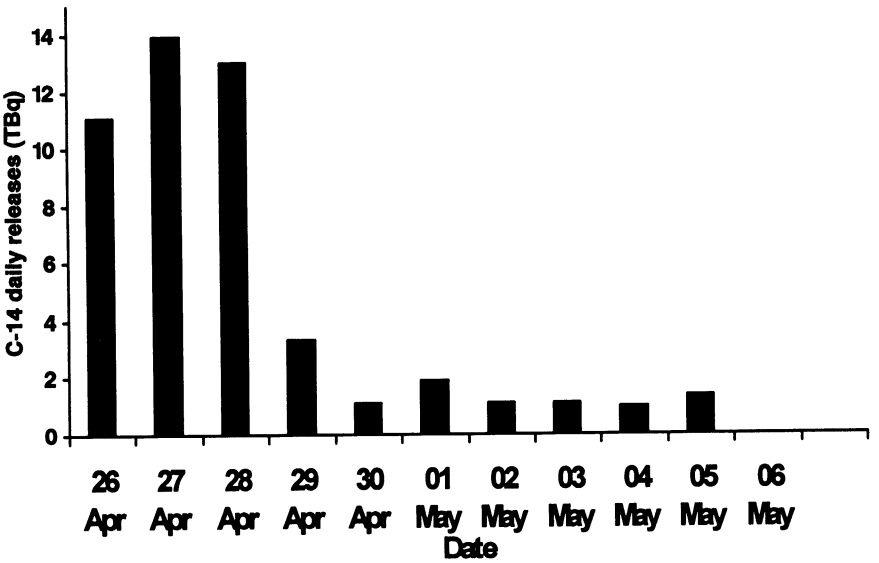

Fig. 4. Estimates of ^{14}C (CO_2) daily release (TBq) during the active stage of the Chernobyl accident

region within 30 km of the NPP will improve the results for the north part. This area was contaminated during the time of maximum release rate (26–28 April) and more precise release estimation for this period is especially important to reconstruct the emission scenario for volatile radionuclides.

Conclusion

- ^{14}C measurement is a practical method for retrospective reconstruction of accidental releases and subsequent transport of radioactive gases.
- Use of discriminating sample preparation technology (*e.g.*, splitting of early and late wood samples) maximizes the precision of ^{14}C measurements.
- Our results show that it is possible to determine amounts of excessive ^{14}C of accidental origin at distances of as much as 30 km from the Chernobyl NPP.
- Experimental data for $\Delta^{14}C$ assimilated in tree rings are clearly in good agreement with modeled ones.
- The time-dependent ^{14}C release rate during the first stage of the Chernobyl accident and the general structure of the accidental deposition field may be reconstructed using the experimental data for ^{14}C assimilated in tree rings with the aid of atmospheric transport modeling.

Future complementary measurements would help more accurately define the deposition field map as well as the value of total ^{14}C release in 1986. We currently provide additional detailed ^{14}C sampling and dating for a number of villages in the 30-km exclusion zone around the NPP—south: Terekhiv, Ilovnytsja, Rossokha, Bychky, Zamoshna, Novoselky; south-southeast: Chernobyl, Kupovate, Otashev, Gorodysche, Zeleny Mys; east: Gorodchan, Chapajevka; north: Usiv. These projects facilitate obtaining detailed and precise data for the spatial distribution of ^{14}C due to the accident. Obviously, it will be useful to expand sampling to the territory of Belarus (*i.e.*, in the northwest and northeast directions) to refine our estimates of both spatial distribution and the level and time variation of the ^{14}C release.

https://doi.org/10.1017/S0033822200018257 Published online by Cambridge University Press

REFERENCES

Arslanov, Kh. A. 1987 *Radiocarbon: Geochemistry and Geochronology.* Leningrad, Leningrad University Press: 300 p. (in Russian).

Borzilov, V. A. and Klepikova, N. V. 1993 Effect of meteorological conditions and release composition on radionuclide deposition after the Chernobyl accident. In *The Chernobyl Papers.* Vol. 1. *Doses to the Soviet Population and Early Health Effects Studies.* Washington, REPS: 47–70.

Buikov, M. V., Garger, E. K. and Talerko, N. N. 1992 Research into the formation of spotted pattern of radioactive fallout with the Lagrangian-Eulerian model. *Meteorologiya i Gidrologiya* 12: 33–45 (in Russian).

Buzinny, M., Kovalyukh, N., Likhtarev, I., Los', I., Nesvetajlo, V., Pazdur, M. F., Sobotovich, E. and Skripkin, V. 1995 Ecological chronology of nuclear fuel cycle sites. *In* Cook, G. T., Harkness, D. D., Miller, B. F. and Scott, E. M., eds., Proceedings of the 15th International ^{14}C Conference. *Radiocarbon* 37(2): 469–473.

Buzinny, M. G., Los', I. P., Zelensky, A. V., Kovalyukh, N. N., Skripkin, V. V. 1993b Possible correlation of ^{14}C level in vegetables and ^{131}I in thyroid gland due to Chernobyl accident. *Actual Problems of Liquidation of Medical Consequences of Chernobyl Accident: Abstracts of Ukrainian Scientific Practical Conference, Kiev, 20–22 April 1993.* Part I. Kiev, Health Ministry of Ukraine, Ukrainian Center for Radiation Medicine: 48–49 (in Russian).

Buzinny, M. and Skripkin, V. 1995 Newly designed 0.8-mL Teflon® vial for microvolume radiocarbon dating. *In* Cook, G. T., Harkness, D. D., Miller, B. F. and Scott, E. M., eds., Proceedings of the 15th International ^{14}C Conference. *Radiocarbon* 37(2): 743–747.

Buzinny, M., Zelensky, A., Kovalyukh, N., Skripkin, V. and Sanin, E. 1993a Retrospective reconstruction of ^{14}C levels in emergency explosion after the Chernobyl accident. In *Conference on Actual Questions of Retrospection, Running and Prediction Radiation Dosimetry after Chernobyl Accident. October 27–29, 1992, Kiev.* Kiev, Ukrainian Scientific Center of Radiation Medicine, Enterprise "ROSA" MinChernobyl of Ukraine: 118–124 (in Russian).

Buzulukov, Yu. P. and Dobrynin, Yu. L. 1993 Release of radionuclides during the Chernobyl accident. In *The Chernobyl Papers.* Vol. 1. *Doses to the Soviet Population and Early Health Effects Studies.* Washington, REPS: 3–21.

Gaiko, V. B., Korablev, N. A., Solov'ev, E. N., *et al.* 1985 Discharge of ^{14}C by a nuclear power plant with RBMK-1000. *Atomnaya Energiya* 59(144) (in Russian).

Kovalyukh, N. N. and Skripkin, V. V. 1992 Chemical sample preparation for radioecological determination of radiocarbon. In *Radioisotopes in Ecological Investigations.* Kiev, Naukova Dumka: 93–101 (in Russian).

Kovalyukh, N. N., Skripkin, V. V., Sobotovich, E. V., *et al.* 1994 Radiocarbon of accidental release of Chernobyl NPP in annual tree rings. *Zeszyty Naukowe Politechniki Slaskiej – Matematyka-Fizyka* 71: 217–224 (in Russian).

Olsson, I. U. and Possnert, G. 1992 ^{14}C Activity in different sections and chemical fractions of oak tree rings, AD 1938–1981. *In* Long, A. and Kra, R. S., eds., Proceedings of the 14th International ^{14}C Conference. *Radiocarbon* 34(3): 757–767.

Salonen, L. 1987 Carbon-14 and tritium in air in Finland after the Chernobyl accident. *Radiochimica Acta* 41: 145–148.

Skripkin, V. and Kovalyukh, N. 1994 A universal technology for oxidation of carbon-containing materials for radiocarbon dating. *Conference of Geochronology and Dendrochronology of Ancient Towns and Radiocarbon Dating of Archeological Finds. Vilnius, Lithuania, October 31–November 4, 1994. Abstracts and Papers.* Vilnius, Vilnius University Press: 37–42.

Vakulovski, S. M., Shershakov, V. M., Golubenkov, A. V., *et al.* 1993 Computer-information support of analysis of radiation environment in the territories polluted as a result of the Chernobyl accident. *Radiation & Risk* (Moscow-Obninsk) 3: 40–61 (in Russian).