

RADIOACTIVE GRAPHITE DISPERSION IN THE ENVIRONMENT IN THE VICINITY OF THE CHERNOBYL NUCLEAR POWER PLANT

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ABSTRACT. This paper estimates the radioactive graphite dispersion on the land surface (forest litter and soil) as a result of the Chernobyl Nuclear Power Plant (NPP) release. Graphite mass was calculated using an estimated average concentration of 2.5×10^7 Bq/kg C (carbon). The sample collection method, sample origin and its mass, and sample preparation procedure used for preparation of benzene were taken into account to obtain the optimum sensitivity of the method. Thus, the sensitivity of the corresponding method for graphite detection in forest litter was estimated to be 0.2 mg/m². All analyses gave a range of deposited graphite from 0.12 to 52.6 mg/m². The maximum value was observed at a site located 9 km west of the Chernobyl NPP. The results of the study indicate the importance of studying the upper layer of soil (0–5 cm) in addition to the lower layer of forest litter.

INTRODUCTION

Reconstruction of the processes that occurred at the 4th reactor of the Chernobyl Nuclear Power Plant (NPP) in the initial period of the accident (26 April to 5 May 1986) could clarify some answers to a number of important questions about the accident. ¹⁴C ($T_{1/2} = 5730$ yr) is one of the long-lived radionuclides released during nuclear accidents. ¹⁴C accumulated in the graphite stacks of the Chernobyl NPP during the operation of the reactor. Due to the accident, the reactor graphite partially burned and dispersed, forming the 2 main components of ¹⁴C release: a gaseous component (CO₂) and an aerosol component (graphite dust). This combination formed a trace in the environment that remained for a long period of time. This is the ¹⁴C accumulated from CO₂ in annual tree rings and graphite dust deposited in the system “forest litter – soil.” Most of the investigations of ¹⁴C originating from the Chernobyl NPP were performed using tree-ring analysis. Results of the tree-ring analysis allowed us to estimate a total gaseous release and its daily variations during the accident (Buzinny et al. 1998). The ¹⁴C variation analyzed for a wide time span (1978–1999) for 2 trees in the vicinity of Chernobyl NPP also allowed for estimating ¹⁴C operational releases (Buzinny and Talerko 2000b).

Other types of environmental samples analyzed were plants and grasses collected in 1986. These samples show very high levels of ¹⁴C at different observation sites (Buzinny et al. 1993). The ¹⁴C activity of samples was formed by both gaseous (structural contamination) and aerosol (surface contamination of the samples) components, which cannot be separated.

Some aspects of ¹⁴C behavior in the forest ecosystem around the Chernobyl NPP are described by Kovaliukh et al. (1998). We assume that the system “forest litter – soil” is stable enough to preserve graphite dust for the period since the accident up to the time of sampling, in the summer of 1997. As the stability and behavior of graphite are not the topic of this paper, this issue should be studied separately. Our study of graphite deposition in the environment was aimed at the determination of the peculiarities of the graphite distribution.

Evaluation of ¹⁴C Deposition in the Reactor Graphite

Analysis of the dispersion of ¹⁴C components deposited in the environment during the accidental release is complicated by 2 factors. The first factor is the uncertainty of ¹⁴C activity deposited in the reactor graphite at the moment of the accident, and the second factor is the uncertainty of measurements of the gaseous and aerosol components from the releases.

The stack of the 4th Chernobyl NPP reactor contained 1850 tons of graphite (Gaiko et al. 1985), and the activity of ^{14}C accumulated in graphite during the operation of the 4th reactor was estimated to be 100 TBq (Begichev et al. 1990). Direct measurements of 4 samples of graphite fragments from the 4th reactor of the Chernobyl NPP gave a maximum value of 2.5×10^7 Bq/kg, and the corresponding estimate of total activity in the graphite was 46 TBq (Buzinny et al. 1993). The deposition intensity of ^{14}C for the RBMK-1000 reactor was estimated to be 3300 GBq/TWt(E) (Salonen 1987). Hence, the ^{14}C deposition equals 187 TBq during the 570 d of the 4th reactor's operation (for the average reactor power of 10.9 MWt/day). According to the estimates given by Borovoi (1995), a significant part of graphite burned during the active stage of the accident, and a lesser part was discharged as finely dispersed dust and deposited in the environment. About 700 tons of graphite (~38%) was estimated to have remained in the reactor. Using high estimates of total ^{14}C activity in graphite (100 and 187 Bq), the remaining graphite in the reactor was estimated to have ^{14}C in the range of 38 and 68 TBq, respectively (Buzinny et al. 1998).

Estimation of the Amount of ^{14}C Gaseous Discharges

To determine the specifics of ^{14}C spatial dispersion in the accidental releases at the Chernobyl NPP, Buzinny et al. (1993, 1998) performed a study of ^{14}C deposited in annual tree rings (1985–1987). The results covered more than 60 sites where wood was collected and allowed us to determine both daily releases during the active stage of the accident and the spatial dispersion of the releases (Buzinny and Talerko 2000a). We determined that the total ^{14}C gaseous discharges were ~50 TBq. The aerosol fraction deposited in the environment covered a wide range from 12 to 62 TBq (Buzinny et al. 1998).

METHODS

To estimate the activity of the dispersed ^{14}C and the graphite, we used 2 components of ^{14}C : ^{14}C graphite activity and the activity of the ^{14}C natural component in plant material. To evaluate the 2 methods of sample preparation (a conventional one including prior pyrolysis of the organic matter and a second method of direct synthesis of lithium carbide [Skripkin and Kovalyukh 1998]), we performed a sample count rate comparison for both ^{14}C components in forest litter (graphite and natural carbon) in corresponding samples using Equations 1 and 2:

$$CPM_{\text{graphite}}(\text{count} \cdot \text{min}^{-1}) = M_{\text{litter}} \times A_{\text{graphite}} \times \frac{\sigma}{M_s} \times \frac{S}{S_0} \times E_{\text{graphite}} \times E_{\text{LSC}} \times 60 \quad (1)$$

$$CPM_{\text{litter}}(\text{count} \cdot \text{min}^{-1}) = M_{\text{litter}} \times A_{\text{litter}} \times E_{\text{litter}} \times E_{\text{LSC}} \times 60 \quad (2)$$

where $A_{\text{graphite}}^1 = 2.5 \times 10^7$ Bq/kg; only one of the 3–4 estimations mentioned above is in the range $(2.5\text{--}10.0) \times 10^7$ Bq/kg; $M_{\text{litter}} = 8$ g or 40 g (mass of litter sample used by 2 different methods); the subscript litter is the density of graphite deposition onto the land surface (mg/m^2); M_s is the mass in grams the forest litter sample (50×50 -cm sampling square); $S/S_0 = 0.25$ is the part of the area where forest litter was sampled; $E_{\text{graphite}} = 0.8$ (the chemical yield of benzene synthesis in forest litter); $A_{\text{litter}} = 0.5 \times 0.92 \times 0.226$ Bq/g, 0.5 is the fraction of carbon in forest litter, 0.92 is the fraction of carbon in benzene, and 0.226 Bq/g is the specific activity of ^{14}C “modern” carbon of objects in the environment); $E_{\text{litter}} = 0.45$ and 0.12 (the chemical yield of benzene synthesis for the 2 applied methods); and E_{LSC} is the efficiency of ^{14}C counting in benzene (80%).

The expected count rate for samples undergoing both methods of preparation is given in Table 1. The table shows that the ratio signal/background (the natural level of ^{14}C is background) is the best one for the “conventional” method. This method is the most sensitive as it can be applied to samples

¹Only one value is used to show the order of graphite deposition mass (mg/m^3).

with a density of graphite deposition of $n \times 10^{-4}$ g/m² compared to $n \times 10^{-3}$ g/m² used for the other method (Buzinny et al. 2000). Unfortunately, high uncertainty and low reproducibility (during pyrolysis some graphite dust could burn) in the chemical yield of benzene from graphite during sample preparation encouraged us to adopt the second method (see Skripkin and Kovaliukh 1998). Therefore, the sensitivity of this alternative method (Skripkin and Kovaliukh 1998) is sufficient for our sample analyses, even when considering the most pessimistic variant of density for graphite deposition.

Table 1 Expected count rate (CPM) for forest litter samples containing deposited graphite for different methods of samples preparation.

σ , g/m ²	Conventional method			Alternate method (Skripkin and Kovaliukh 1998)		
	Graphite	Forest litter	Ratio	Graphite	Forest litter	Ratio
10 ⁻²	240	28.8	8.33	48	21.6	2.22
10 ⁻³	24	28.8	0.83	4.8	21.6	0.22
10 ⁻⁴	2.4	28.8	0.083	0.48	21.6	0.02

The amount of ¹⁴C deposited in each layer and for each collecting site was estimated by determining ¹⁴C activity according to the equation:

$$^{14}C_S = C_S \times A_{^{14}C} \quad (3)$$

where C_S is the mass (kg) of carbon in the analyzed layer and $A_{^{14}C}$ is the ¹⁴C concentration in forest litter in the analyzed layer.

The background content of ¹⁴C is determined by Equation 4:

$$^{14}C_{BG(S)} = C_S \times 226 \times Modern \quad (4)$$

where 226 Bq/kg C expresses the concentration of the “modern level” of carbon corresponding to the background level of the mass of carbon, and *Modern* is a relative concentration of ¹⁴C that can be observed in the forest litter expressed as a percent of the Modern level.

Thus, the ¹⁴C fraction caused by graphite is determined as:

$$\Delta^{14}C = ^{14}C_S - ^{14}C_{BG(S)} \quad (5)$$

where 2.5×10^7 is the ¹⁴C graphite concentration; 1000 is the recalculation coefficient (mg/kg); and S_{site} is the collecting area (m²) for the forest litter samples. The density of graphite deposition on each studied site has to be summed for all analyzed layers.

Sampling and Sample Preparation

The amount of deposited graphite in the environment is estimated from the results of the excess ¹⁴C activity above the modern natural level (Buzinny and Talerko 1999, 2000b) in the forest litter and/or in the soil samples. A method based on direct synthesis of lithium carbide prior to benzene (Skripkin and Kovaliukh 1998) and subsequent liquid scintillation counting (LSC) was used to measure the activity of ¹⁴C. The measurements were carried out using a modern low-level liquid scintillation (LS) spectrometer, Quantulus 1220™ (Wallac Oy, Finland).

Sampling Network

More than 60 sites were included in this ^{14}C activity study. The samples covered a distance up to 60 km from the reactor in different directions around the Chernobyl NPP (see Figure 1). All these sites were located near settlements (Buzinny et al. 1993; Buzinny and Talerko 1999, 2000a). We collected samples of forest litter (different layers of the forest litter according to its morphological structure) and soil at 41 locations.

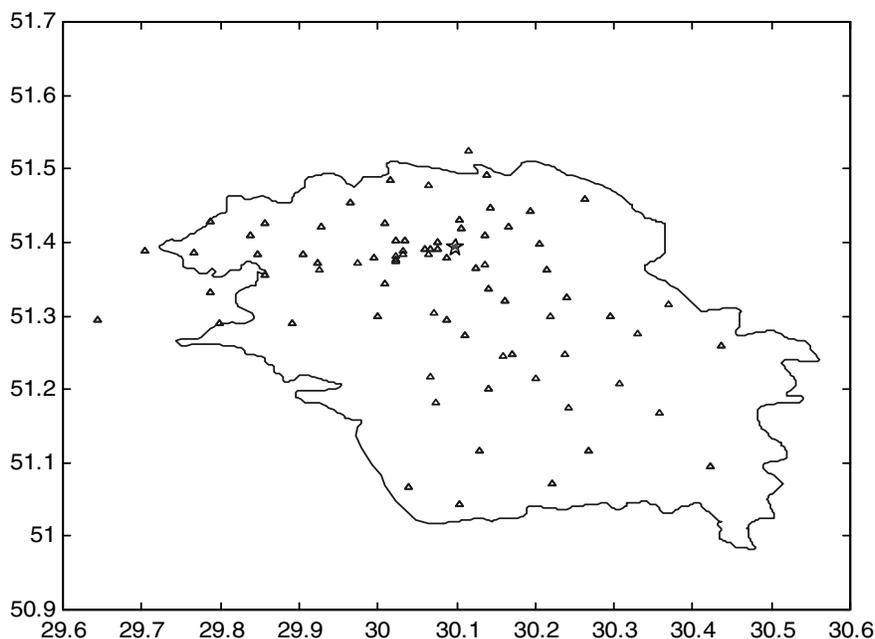


Figure 1 Sampling map for ^{14}C study (Buzinny et al. 1998; Buzinny and Talerko 1999, 2000a). The border in the map corresponds to border of the former Chernobyl district. The axes represent the latitude and longitude coordinates given in Table 2.

The forest litter samples were collected from the surface and were sampled from an area of 2500 cm^2 ($50 \times 50\text{ cm}$). The samples were stratified by 3 layers according to their morphological state: A – upper layer: fresh sagged needles; B – middle layer: old needles without structural changes; C – lower layer, adjacent with upper soil layer: destroyed needles and other humus-like substances. The relative average distribution of the mass of forest litter samples for 38 sites was found to be $A = 13 \pm 5\%$; $B = 29 \pm 10\%$; and $C = 58 \pm 11\%$. Samples of total litter were collected in 3 sites where we could not stratify the forest litter by layers. The soil samples were collected from 2 layers following one another at a depth of 5 cm each for all sites. Soil sampling was performed using the same area used for forest litter sampling ($15 \times 25\text{ cm}$). According to our research hypothesis, we assumed that graphite had been deposited into the lower layer of the forest litter (C); hence, we used the corresponding A and B layers as a background.

RESULTS

Analyses of Forest Litter

I believe that the method chosen to estimate the amount of graphite deposited on the land surface (Equations 1 and 2) is valid up to 0.2 mg/m^2 . Forest litter samples were collected at 41 sites. A total

of 59 samples of forest litter were involved for the study and 75 analyses of ^{14}C activity were carried out, including replicates. The results obtained show that the majority of samples have a ^{14}C activity exceeding the background (Table 2) and cover a wide range.

Table 2 Dispersion of ^{14}C (pMC)^a in the samples of various layers of forest litter for particular sites of observation. Coordinates of wood collection sites and forest litter samples for studying ^{14}C in the vicinity of the Chernobyl NPP (E longitude, N latitude) (Buzinny et al. 1998, 1999, 2000).

Site	Longitude (E)	Latitude (N)	Layer	pMC
97/27	29°58'30"	51°22'15"	A	115.0
97/27	29°58'30"	51°22'15"	B	127.9
97/27	29°58'30"	51°22'15"	C	108.4
96/09	30°10'0"	51°25'15"	A	118.8
96/09	30°10'0"	51°25'15"	B	125.4
96/09	30°10'0"	51°25'15"	C	235.0
96/33	30°3'38"	51°23'25"	A	122.5
96/33	30°3'38"	51°23'25"	B	126.9
96/33	30°3'38"	51°23'25"	C	132.7
97/06	30°6'20"	51°25'10"	A	123.0
96/04	30°5'15"	51°22'40"	Total	385.3
97/24	30°1'52"	51°23'3"	A	126.8
97/24	30°1'52"	51°23'3"	B	147.8
97/24	30°1'52"	51°23'3"	C	561.2
97/20	30°6'50"	51°31'30"	A	116.0
97/20	30°6'50"	51°31'30"	B	123.4
97/20	30°6'50"	51°31'30"	C	235.2
97/08	30°0'55"	51°29'5"	A	116.1
97/08	30°0'55"	51°29'5"	B	121.3
97/08	30°0'55"	51°29'5"	C	126.8
97/06	30°6'20"	51°25'10"	B	123.8
96/36	30°4'30"	51°24'0"	Total	188.1
96/28	29°54'18"	51°23'3"	A	108.3
96/28	29°54'18"	51°23'3"	B	132.9
96/28	29°54'18"	51°23'3"	C	442.5
96/02	30°1'20"	51°22'25"	A	113.7
96/02	30°1'20"	51°22'25"	B	141.0
96/02	30°1'20"	51°22'25"	C	180.0
TL ^b	—	—	A	130.0
TL ^b	—	—	B	272.0
TL ^b	—	—	C	378.0
97/06	30°6'20"	51°25'10"	C	149.1
96/34	30°4'30"	51°23'26"	Total	135.5

^apMC is the percent of "modern" ^{14}C , a relative unit that expresses the ^{14}C activity in the carbon of the oak wood of 1890 growth (100 pMC = 226 Bq/kg C).

^bThe sampling site is outside our network.

Forest Litter

Reproducibility of the ^{14}C activity determinations of the samples of forest litter was determined for the 5 most contrasting samples (5 replicates for each). We found that the relative standard deviation ranged from 5.1 to 13.7% (Table 3). There are discrepancies in the results for the same collecting

sites and corresponding layers in Tables 2 and 3, because the results of single measurements are presented in Table 2 and average results for the same samples are demonstrated in Table 3.

Table 3 Relative content of ^{14}C (pMC) in forest litter samples for 5 of the most contrasting observation sites (the average results of 5 replicates for each sample).

Site	Layer	^{14}C	Standard deviation	Relative deviation (%)
97/27	C	938.9	128.7	13.7
96/28	C	417	43.1	10.4
TL	C	414.0	42.9	10.4
96/04	Total	326.3	43.1	13.2
96/02	C	181	9.1	5.1

A particularly high difference is observed for site TL (Table 3), where ^{14}C levels for A, B, and C layers are 135, 275, and 380 pMC, respectively. The A and B layers of forest litter apparently have significant ^{14}C contamination. A similar increase in the ^{14}C level of layer B was found at site 97/24. Meanwhile, a series of observation sites located near the Chernobyl NPP have either very low levels of ^{14}C or values close to the detection limit. It was assumed that decontamination work might have been done at those sites. In this case, the layer of litter containing the deposited graphite had been collected and removed to the burial ground. It is possible that the graphite was moved to the upper layer of the soil. Tests of the soils (0–5-cm layer) and new collections of forest litter samples for secondary tests will help in future comparisons.

Soil Study

Soil samples corresponding to the forest-litter collection sites that contained the maximum determined levels of the deposited graphite were analyzed to determine the amount of ^{14}C . All soil samples were initially dried at 150 °C. The amount of organic matter for each soil sample was estimated by burning 1 g of sample material. The samples (200 g) were then mixed with an adequate amount of pyrolusite (MgO_2) at the upper part of the reactor of carbide lithium synthesis. The calculations of graphite balance for soil were done in a similar way as the calculations of graphite balance for forest litter. These results are shown in Table 5.

Comparing the values obtained of the graphite deposition density (Tables 4 and 5) for samples 97/27, 96/13, and 96/04, we received a corresponding series of values for the graphite fraction of the soil: 4.9%, 19.4%, and 1.5%. We expect the most likely explanation for this interval is the consistency of samples from the different strata during sample collection.

Table 4 shows the ^{14}C and graphite balance in the forest litter for different observation sites. The results include the sites where the graphite content does not exceed 0.1–0.2 mg/m² (the detection level). Excessive levels of ^{14}C cannot be determined for a number of sites due to the discrepancy in the ^{14}C level for different layers. The maximum value (52.6 mg/m²) of graphite is determined for a site 9 km west of the Chernobyl NPP.

CONCLUSIONS

This paper demonstrates an application of ^{14}C study for practical reconstruction of accidental releases from a nuclear power plant that used a graphite reactor. The sensitivity of the method is adequate for conducting analyses of samples collected at distances up to 10 km. Both components of accidental releases of ^{14}C of the Chernobyl NPP can be used for research on ^{14}C distribution.

Table 4 Balance of ^{14}C and graphite in the forest litter for different sites of observation.^a

Site code	Distance (km)	Nr of layers	Sample mass (g)	Carbon mass (g)	^{14}C (Bq)	Background ^{14}C (Bq)	A ^{14}C (Bq)	σ (mg/m ²)
97/27	8.98	3	900	149.4	368.1	39.3	328.8	52.6
96/04	1.89	1	215	32.4	28.4	8.5	19.8	21.1
97/24	4.85	3	926	223.4	174.5	58.8	115.6	18.5
96/28	13.52	3	440	81.1	87.2	21.4	65.8	10.5
97/20	14.67	3	1005	200.5	107.1	52.8	54.3	8.6
96/36	1.87	3	1440	364.6	120.9	96.0	24.9	3.9
96/02	5.78	3	828	176.9	60.3	46.6	13.8	2.2
96/13	2.31	3	697	150.0	49.8	39.5	10.3	1.6
96/38	6.89	1	570	126.0	39.9	33.2	6.7	1.0
97/26	12.47	1	450	94.6	29.7	24.9	4.8	0.77
96/17	3.63	3	585	126.8	38.0	33.4	4.7	0.74
97/23	3.68	1	270	60.5	20.0	15.9	4.1	0.65
96/09	5.56	3	570	68.0	21.9	17.9	4.0	0.64
97/22	13.35	1	1300	130.7	38.2	34.4	3.8	0.60
96/23	22.67	1	480	57.8	18.6	15.2	3.4	0.54
96/33	2.73	3	485	93.3	27.3	24.6	2.7	0.43
97/06	2.95	3	709	72.4	21.5	19.1	2.4	0.39
96/34	1.73	1	42	7.6	2.3	2.0	0.3	0.36
97/08	11.73	3	600	110.3	30.7	29.1	1.6	0.26
96/24	17.45	1	380	57.4	16.3	15.1	1.1	0.18
96/10	7.24	1	330	42.1	12.1	11.1	1.0	0.16
96/18	9.15	1	445	95.1	26.0	25.0	1.0	0.16
97/21	9.62	1	345	74.7	20.6	19.7	0.9	0.15
96/25	17.27	1	600	105.7	28.6	27.8	0.8	0.12

^aNote: The specific ^{14}C activity of graphite used for the calculation was 2.5×10^7 Bq/kg; samples are sorted according to the level of deposited graphite on the land surface.

Table 5 Balance of ^{14}C and graphite (C, mg/m²) in soil samples (0–5) for different sites.

Site	Mass (g)	^{14}C (pMC)	C ₆ H ₆ (g)	Excess ^{14}C (Bq) ^a	Graphite (mg/m ²)
97/27	1525	367.8	0.637	2.55	2.72
96/13	1420	147.5	1.129	0.53	0.56
96/04	1925	141.8	0.569	0.30	0.32
96/13	1420	132.5	0.885	0.22	0.23
97/14	2065	128.7	0.561	0.15	0.16
96/02	1625	137.0	0.473	0.17	0.18
96/13	1420	128.7	0.669	0.13	0.13
97/14	2065	123.2	0.561	0.09	0.09
96/02	1625	132.8	0.473	0.13	0.14
96/24	1710	121.9	0.441	0.05	0.05

^aExcess ^{14}C in Bq per soil sample collected (sampling area 15 cm × 25 cm × 5 cm).

We should consider that various natural and human activity factors can affect the behavior and transport of the graphite in the environment. As a result of this ^{14}C study that includes the analyses of different layers of forest litter and soil in the vicinity of the Chernobyl NPP, we can estimate the amount of graphite deposited on the land surface and we have the information required to forecast

the implications of this deposition. A comparison of graphite dispersion in forest litter and soil revealed that the fraction of graphite deposited in the soil reached 20% for the sampling conducted in 1997.

The release of graphite due to the accident at the Chernobyl NPP has caused the formation of the contaminant traces in the environment reaching a density of 52.6 mg/m² observed in a site 9 km west of the Chernobyl NPP.

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