^{14}C CYCLE IN THE HOT ZONE AROUND CHERNOBYL

NIKOLAI N. KOVALIUKH, VADIM V. SKRIPKIN

State Scientific Centre of Environmental Radiogeochemistry, Paladina 34, Kiev, 252680 Ukraine

and

JOHANNES VAN DER PLICHT

Centre for Isotope Research, University Groningen, Nijenborgh 4, 9747 AG Groningen
The Netherlands

ABSTRACT. Radiocarbon from the Chernobyl accident was released mainly in two forms: fine dispersed reactor graphite, and carbon dioxide from burning graphite. The CO_2 was partly assimilated by annual and perennial vegetation. Reactor graphite dispersed over a wide territory was taken up biochemically by micromicetes, transforming non-organic carbon of the reactor graphite into organic matter. Organic matter of micromicetes is the main nutrition product for soil organisms such as bacteria, worms, larvae of insects, small beetles, *etc.* The following relatively independent trophic chains are considered: 1. carbon dioxide → leaves, grass → insects; 2. graphite → micromicetes, protozoa, insects. The ^{14}C content in beetles of different species sampled in the 30-km hot zone of the Chernobyl accident site in 1986–1988 agrees well with the contamination levels of insect habitats as well as with their biology.

INTRODUCTION

The Chernobyl catastrophe in 1986 led to a large release of radiocarbon into the environment. Practically the entire graphite moderator (*ca.* 1.7×10^6 kg) was thrown out of the reactor limits after the reactor was destroyed. Table 1 shows the main radionuclide activities. Large fragments of graphite were lying close by the reactor. Small particles, however, were dispersed in the atmosphere several kilometers away from the Chernobyl Nuclear Power Plant (ChNPP). At the first stage of the ChNPP accident the entire ^{14}C content present in the form of a gaseous compound was thrown into the atmosphere. Next, burning of graphite at high temperatures (up to 3000°C) produced CO and CO_2, which were transported to the upper atmosphere. Only a small part of CO_2 was assimilated by vegetation (Kovalyukh 1994). ^{14}C from the reactor traveled in a radioactive cloud over considerable distances, and has been found as far away as Sweden and Finland (Salonen 1987). A total of 4×10^{13} to 6×10^{15} Bq of ^{14}C was released into the environment (DOE 1987; Gofman 1994; Kovalyukh *et al.* 1996). The main form of ^{14}C fallout in the zone of near the NPP was finely dispersed reactor graphite, which had been changed into organic carbon by assimilation *via* microorganisms. At the same time, the ecosystem was contaminated this way by ^{14}C.

Radiocarbon Cycle in the Accident Zone

The main factor determining the ^{14}C cycle in the zone of the Chernobyl accident is the "destruction" of dust-like reactor graphite due to biochemical processes (Fig. 1) combined with the assimilation of transformed carbon by the ecosystem. Laboratory experiments have shown that during an emergency release, reactor graphite in combination with water is destroyed effectively by various types of microfungi (Kovalyukh *et al.* 1994, 1995). As a result, ^{14}C is transformed into organic compounds, bodies of fungi, and into the products of their vital activity such as CH_4 and CO_2. We investigated the ^{14}C contents in the trophic chain: reactor graphite → organic substance of the soil → insects and plants. The most interesting factor in the accident's region is the release of "Chernobyl" ^{14}C out of this trophic chain. The secretion products of the biological objects CO_2, methane and water-soluble organic substances are assimilated only partly by the plants, and then they rejoin the cycle. Only a small part of this ^{14}C exchanges from the chain into the global reservoir. The organic substances of soil, insect larvae, and mature insects yield information on the dynamics of ^{14}C. Lar-

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vae of insects assimilate ^{14}C from a limited local zone, the surface-soil layer, and are the indicators for local levels of soil contamination. Adult insects feed from a large area and can adsorb ^{14}C from a large ecosystem zone (Fig. 1). Correlation of the ^{14}C contents of the surface-soil layer, larvae of insects and adult insects as a function of time, yields information on the ^{14}C transmission processes from inorganic into organic form. In addition, we can determine the Chernobyl ^{14}C release velocity from different ecosystems in the accident zone.

TABLE 1. Radionuclide Composition of Graphite Particles Ejected from ChNPP

	Activity of radionuclides (10^4 Bq g^{-1})			
Sample no.	^{90}Sr	^{134}Cs	^{137}Cs	^{14}C
1	3.7	14	180	1.5
2	27	17	200	3.1
3	96	100	1200	4.3
4	140	130	1400	6.2

Methods and Sample Locations

The ^{14}C content of the samples was measured by liquid scintillation counting (LSC) in Kiev (soils and insects, conventional samples), and by accelerator mass spectrometry (AMS) in Groningen (small samples, insects). The Kiev laboratory applied a new method for samples with a high mineral content (such as soils): thermodestruction with lithium carbide. This method also enabled us to select the different carbon fractions from the soil samples and as well as reactor graphite (Skripkin and Kovalyukh 1994, 1998). For more details, refer to Skripkin and Kovalyukh (1998). The ^{14}C content of small samples (insects) was measured by the Groningen AMS facility (Gottdang *et al.* 1995). To avoid possible contamination with "hot material", the samples were not combusted and graphitized by our standard setup. The insects were cleaned by acid, sealed under vacuum in Pyrex and heated at 300°C for several hours. The amorphous carbon obtained this way was mixed with silver powder and pressed into the sample holders for the AMS ion source. Sample material was taken from near the villages of Shepelichi, Krasnitsa and Leljov, located 6 km west, 16 km west-northwest and 10 km southeast from the Chernobyl site, respectively (Fig. 2).

These sites represent the three standard landscape structures of the Poliesia area. Krasnitsa is located near the end of the Pripyat River terrace, above the floodplain which gradually turns into a leveled morainic apron. In place of the former agrolandscape with soddy podsols, there is now a succession of herbage and grasses with birch and pine subgrowth. Shepelichi is located on the pine forest outskirts, within the pine forest terrace limits consisting of thick sands and sandy loams with soddy and sandy podsols. Forest litter formation is well developed for this ecosystem. Leljov is located within the limits of the high sandy floodplain of the Pripyat River. Floodplain soils (soddy, gley and sandy) are developing under the grain-herbage meadow vegetation. Chemical and biological investigations show an increasing soil formation process. This is typical for the soils of this floodplain.

For these three sites, we selected soil samples at 0–10 cm intervals, containing the lower and upper sections of forest litter, grasses, and insects existing there during 1986–1988. The mixed composition of radioactive fallout was evident by the domination of dispersed particles (*ca.* 75%) near the sampling sites. These particles represent both products of the destroyed fuel elements, as well as the graphite cover of the reactor, which accumulated ^{14}C during operation of the ChNPP.

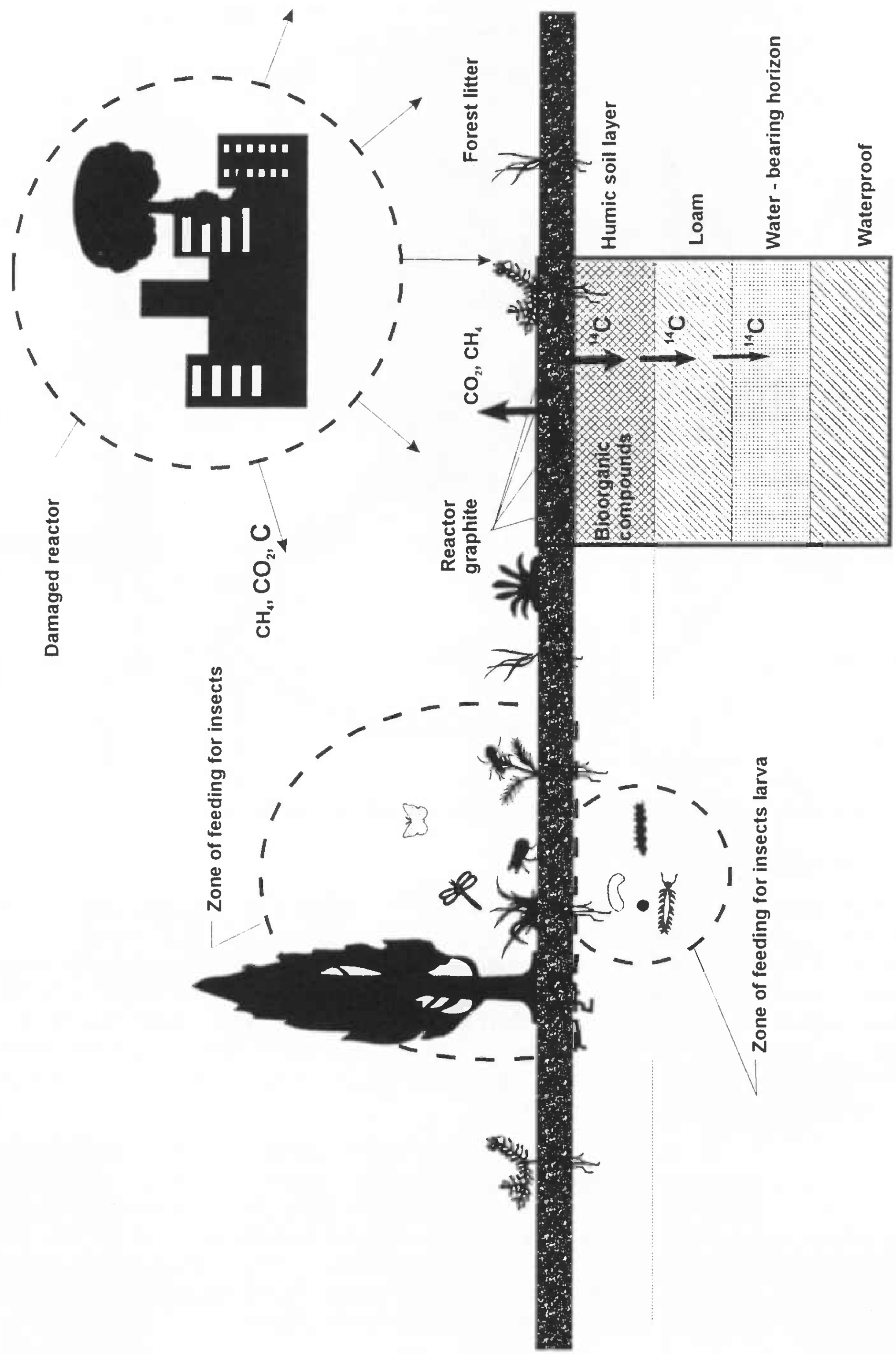

Fig. 1. Schematic depiction of radiocarbon cycle around the Chernobyl NPP

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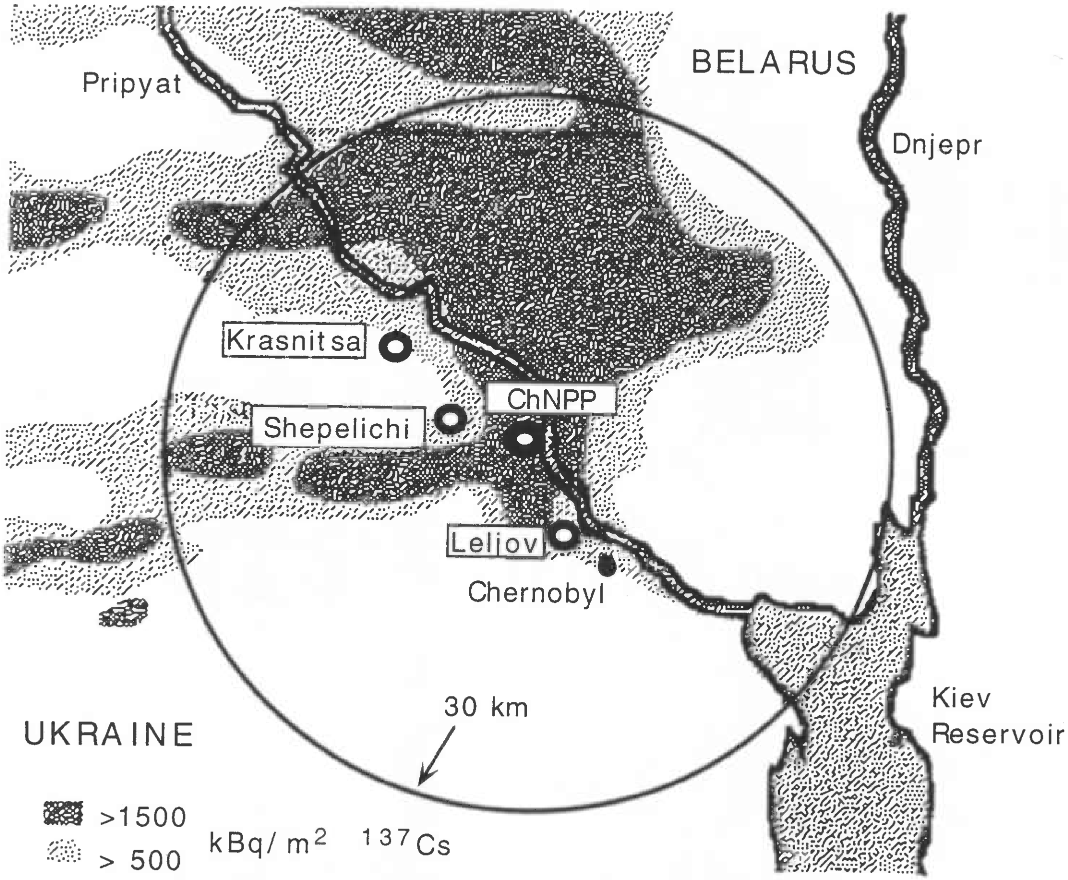

Fig. 2. Map showing the locations of the sampled areas. The contamination levels are indicated.

RESULTS AND DISCUSSION

We have studied the distribution of ^{14}C from the ChNPP accident at three locations (Fig. 2) in the "hot zone" during 1986–1988 for the following parts of the ecosystem: soil, litter bottom, litter top, grasses and insects. The series of insects was measured both in Kiev and Groningen with results shown in Table 2. The highest ^{14}C activities in the insects were, of course, found in 1986, with pMC values ranging from 187 (Shepelichi), 162–174 (Leljov) and 124–134 (Krasnitsa). Insects of the species *Ophanus rufipes* are the most important for our investigation because their diet consists mainly of microfungi. Table 3 and Figure 3 show the results, including the other parts of the ecosystem (soil, litter, grass and selected insects).

The 1986 samples were taken in the autumn. The highest ^{14}C activities (250–390 pMC, depending on the location) are present in the lower part of the litter. This represents the main part of the fallout, corresponding to the initial phase of the accident (April–May 1986). The time period between accident and sampling is five months. Soil contamination occurred as a result of graphite particles penetrating through the litter. The shedding of leaves and thorns in 1986 formed a new litter layer, which adsorbed gaseous and dustlike components of the fallout. The ^{14}C content here appears to be less by a significant amount (*ca.* 50%). The same is true for the grasses.

The beetles inhabiting the area were contaminated simultaneously with the litter, soil and grass (Table 2, Fig. 3). The ecosystem activities for the three investigated sites (Fig. 3) show the dependence on locality. The site closest to the NPP (Shepelichi) indeed shows the highest ^{14}C activity levels. At the site farthest from the NPP (Krasnitsa) the lowest activities are measured, only moderately enriched in ^{14}C compared to 1986 atmospheric values. For the years 1987 and 1988, the ^{14}C results obtained from soil, litter and grass show the redistribution among these elements of the ecosystem as a function of time (Table 3).

TABLE 2. Radiocarbon Content of Insects from the Chernobyl Accident Zone

Site name	Site no.	Insect species	Year	Lab no.	^{14}C content (pMC)
Leljov	1	*Amphimallou solstialis*	1986	GrA-5495	171.7
	2	*Ophonus rufipes*	1986	Ki-6099	174.1
	3	*Nicrophorus vespillo*	1986	Ki-6096	162.3
Krasnitsa	4	*Nicrophorus vespillo*	1986	GrA-6006	128.2
	5	Staphilinidae	1986	GrA-5573	133.6
	7	*Broscus cephalotes*	1986	GrA-6002	134.4
	8	*Ophonus rufipes*	1986	GrA-6005	124.9
	9	*Ophonus cephalotes*	1986	Ki-6097	130.8
	10	*Ophonus cephalotes*	1986	Ki-6098	134.6
	11	*Silpha obscura*	1987	GrA-6008	122.2
	12	*Silpha obscura*	1987	GrA-6009	121.3
	13	*Amphimallou solstialis*	1987	GrA-6004	128.8
	14	*Ophonus rufipes*	1988	GrA-5494	117.2
	15	*Silpha obscura*	1988	GrA-5498	119.0
	16	*Nicrophorus vespillo*	1988	GrA-5497	122.7
	17	*Silpha Obscura*	1988	GrA-5500	120.7
Shepelichi	18	*Ophonus rufipes*	1986	Ki-6104	187.3
	19	*Corabus coriace*	1988	GrA-6007	168.9
	20	*Geotrupes stercorosus*	1988	GrA-5493	131.9
	21	*Corabus arceusis*	1988	GrA-5496	139.6
	22	*Geotrupes stercorosus*	1988	GrA-5499	132.0

TABLE 3. Radiocarbon Content (in pMC) in the Chernobyl Accident Zone

Year, site	Soil	Bottom litter	Top litter	Grass	Insects
1986					
Shepelichi	217.1	392.3	201.5	206.8	187.3
Leljov	205.4	320	186.8	190.4	174.1
Krasnitsa	180.7	249.6	137.3	143.1	124.9
1987					
Shepelichi	233.2	301.4	137.6	134.3	
Leljov	142.8	194	133.9	132.4	
Krasnitsa	182.8	191.8	128	127.4	122.2
1988					
Shepelichi	239.4	272.6	132.7	122.9	139.6
Leljov	140.6	188.5	129.3	121.8	
Krasnitsa	177.3	182.6	123.8	120.3	120.7

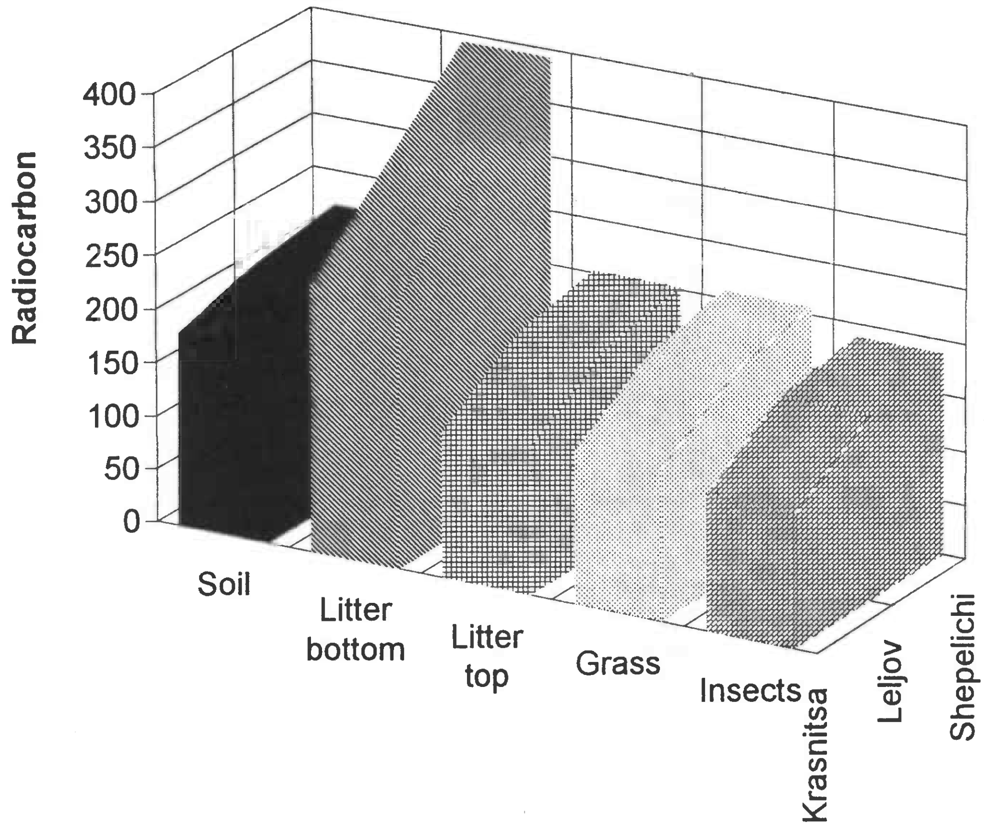

Fig. 3. Radiocarbon content in the "soil-litter-grass-insects" system in the Chernobyl accident zone (fall 1986)

Conclusion

We have investigated ^{14}C released by the 1986 Chernobyl accident in the "hot zone" ecosystem, by measuring soil, litter, grass and insects at three selected sites. Cumulative ^{14}C accumulation occurred in the soddy podsols of the forest (Shepelichi site) while the activity in other materials diminished. Redistribution of ^{14}C in the old-arable successional system (Krasnitsa site) is not large. The flood-plain (Leljov site) shows a decreasing ^{14}C activity in all samples. The most convenient study objects for the ecosystem contamination are the insects *Ophonus rufipes*, since microfungi are part of their diet. These microfungi assimilate reactor graphite. The levels of ^{14}C contamination of connected ecosystems form a basis for radiochemical monitoring of trophic chains and can be used for forecasting rehabilitation processes of contaminated areas. The present and necessary future investigations make it possible to understand the distribution dynamics of ^{14}C in the natural environment.

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