Environmental radioactive pollution: biogeochemical assessment of ^{90}Sr in trees using geostatistical methods. Theory, methodology and a case study

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Abstract

To measure the degree of environmental radioactive pollution and the chemical composition of the soil, two kinds of methods can be used: the direct or physico-chemical methods, consistent in the extraction of samples from the soil and their subsequent analysis and the indirect or biochemical methods, based on the measurements of certain elements in the living beings, and particularly in plants.
Direct methods have been used more widely to date; however, indirect methods have some advantages when compared to the direct methods:

1. Sampling is much more simple and cheap, even though there are certain limitations of the period in which it can be done.
2. The sampling grid can be much denser and so more representative.
3. Additional information is allowed since morphological changes made by radioactive pollution of the environment on plants can tell us about the mutations on living beings.

The use of some elements in vegetation as indicative of the state of radioactive pollution of the environment in the surroundings of deposits of radioactive wasting materials or any other toxic materials has the same methodological basis as that applied to the study of mineral deposits. It is known that minerals create dispersion halos of certain chemical elements in soil and, according to their concentration in plants, the deposits can be identified. In this way, buried radioactive waste also form dispersion halos of radioactive and toxic elements, which are accumulated in plants. So a new methodology is possible to be developed and applied to environmental pollution assessment, based on the use of some parts of the plants as geochemical indicators.
Nevertheless, to develop correctly the application of indirect methods in the monitoring of the environment pollution, some aspects have to be still studied: what

plant that can be more effective for such monitoring, what parts of those organisms reflect more faithfully the pollution of the soil, evaluation of the depth from where the roots absorb the radioactive pollutants and to description of the dispersion halos in the neighbourhood of the waste deposits.
In this work, theory, methodology and application of the indirect methods are studied from a geostatistical point of view. A case study of the Exclusion Zone of the Chernobyl's Central Atomic (CCA) in Ukraine, using different parts and analyzing different kind of trees to assess ^{90}Sr radio nuclide concentration is included.

Introduction

After Chernobyl accident many radioactive materials were disposed and buried. The piling up of radioactive waste has become an environmental problem because radioactivity has travelled to aquifers, ground waters and soils. It is usual to control the pollution sampling soil, rocks or water. But those direct methods are expensive and difficult to carry out. The indirect monitoring of pollution is easiest to do and cheaper. Nevertheless, to develop correctly the application of indirect methods in the monitoring of the environmental pollution some aspects have still to be studied:

1. The plant that can be more effective for such monitoring.
2. It is necessary to determine what parts of those organisms reflect more faithfully the pollution of the soil.
3. It must be evaluated the depth from where the roots absorb the radioactive pollutants.
4. The dispersion halos in the neighbourhood of the waste deposits should be described.

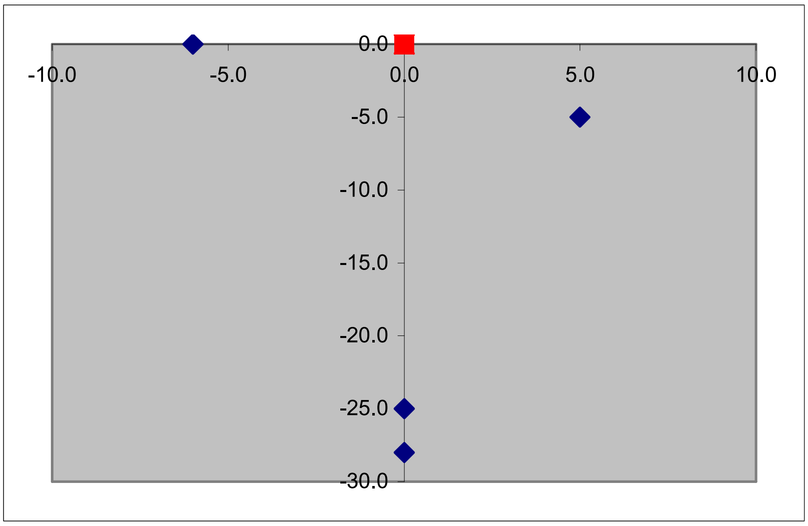
So, indirect monitoring is based on the capability of plants to absorb radioactive pollutants from soils and aquifers. Theoretically, biogeochemical monitoring methods are based on the same laws as usual biogeochemistry. Nevertheless, the piling up of radio nuclei in the soils and their correlation in some parts of the plants has to be carefully studied. It has been stated that some different species of trees can absorb radio nuclei in different amounts and that those amounts are not the same, depending on which part of the tree is analyzed. On top of that, absorption of ^{90}Sr depends strongly on humidity of the soil. In this work some results based on monitoring Chernobyl Exclusion Area during years 1995-2000 are studied; from this study, biogeochemical methods to monitor ^{90}Sr are developed. It must be said that available data set is very small; nevertheless this work is a first attempt on that direction.

The studied zone

All samplings were done in the period 1995 to 2000 in four standardized polygons (see Figure 1). They show the typical natural conditions in natural woods at Ukrainian Poléss`e. In all of them soils have quite similar humidity conditions, landscape, soil cover, vegetal cover and radioactive superficial pollution. Short descriptions of each of them are the following:

- Ditiáytki-1 (D1) polygon 28 km south from CCA, automorphic landscape, dry soils, mixed forest (pine, birch and oak trees). Exposition dose's power to γ-rays (PED) on soil surface in 1998 was 0.036 mR/hour. Soil surface pollution from ^{90}Sr in 1998 was $5.6.10^{10}$ Bk/m^2.
- Ditiáytki-3 polygon (D3). 26 km south from CCA, hydromorphic (humid or marshy), mixed forest (alder, poplar, pine, birch, oak trees). PED on soil surface in 1998 was of 0.031 mR/hour. Soil surface pollution from ^{90}Sr in 1998 was $7.4.10^{10}$ Bk/m^2.
- Kopachí-2 polygon (K2). 7 km south-east from CCA, automorphic landscape, flat and with a good drainage. Forests can be of pine or mixed pine and birch trees. PED in soil surface in 1998 was of 0.29 mR/hour. Soil surface pollution from ^{90}Sr in 1998 was of $1.1.10^{12}$ Bk/m^2.
- Shepélichi-1 polygon (Sh1). 6 km west from CCA, automorphic landscape, flat and with a good drainage. Forests can be of pine or mixed pine and birch trees and also just birch or just oak trees. PED on soil surface in 1998 was of 2.88 mR/hour. Soil surface pollution from ^{90}Sr in 1998 was $1.9.10^{13}$ Bk/m^2.

Figure 1. Polygon location; distances in km from CCA



Materials and Methods

In investigated polygons, the most important source of ^{90}Sr in plants is polluted soils by Chernobyl precipitations in 1986. Also, another ^{90}Sr source for plants at CCA zone is its absorption through leaves and barks directly from the air. Atmospheric pollution comes from the spreading of polluted dusts, from ground fire fumes and from atmospheric emissions from CCA (which was still working until 15th December 2000). So in 1995 at a distance of 5-8 km from CCA the average annual contents of ^{90}Sr in air was of 3.7.10^{-5} – 8.5.10^{-5} Bk/m^3.

Polygons were quite uniform from a radiological point of view (level and composition of radioactive pollution) and also for edaphologic conditions. Naturally, those parameters changed slightly from one border to another for each polygon. But this handicap is minimised and it is impossible to absolutely avoid it.

The most widespread tree species is pine tree (Pinus silvestris L.) which was collected in all polygons. The second one, in importance, was birch tree (Betula pendula L.), which was collected in three polygons (D1, D2, Sh1). At last, oak tree (Quercus robur L.) was just collected in D1 and D3.

For each polygon, the more typical trees were felled to get the samples. Every year the most up to sample trees were chosen (in trunk thickness, height, canopy size). They were called model trees.

Felled model trees were cut in their principal morphological parts: log, leaves, branches, bark and roots. For pine trees leafs and branches were classified in three ages (1, 2 or 3 year old). For caduceus trees, branches were classified into young (diameter < 1 cm) and old ones (diameter > 1 cm). Log and bark were collected from low parts (less than 1 m tall), from medium parts and from high parts. In some cases it was possible to separate branches and leafs of low, medium and high parts. In other cases branches and leafs were collected just from the medium height. Tree height was just trunk length, not trunk and canopy.

Samples to analyze were taken from model trees. Biologic matter was homogenized and aliquot simples were analyzed in laboratory following standard methods. Sampling was done every year alter finishing vegetation active processes (July-August). Sampling of birches' sap was done in April, when up going rate of sap inside the trunk is very high.

Statistical analysis

As it is stated in the introduction, one of the goals of this work is to find out what part of the studied organisms reflects more faithfully the pollution of the soil. To do so, some statistical analysis has to be done; then, the correlation coefficient between ^{90}Sr contents in the different parts of the trees and different soil level, and its cover, has been calculated. In next three tables (Table 1 for pine trees, Table 2 for birch trees and Table 3 for oak trees) those coefficients are shown. As there is a poor data set (just one data for each sample, year and polygon), those tables can be read just from a qualitatively point of view. To begin with, results for SO4, soil at depth greater than 20 cm, cannot be taken into account because data are too poor; they were samples just for two years. Sometimes there is a lag, so correlation has not been calculated. Nevertheless, those tables show that there is a correlation among some parts of the trees and soil pollution.

It can also be observed from those tables that birches absorb better ^{90}Sr than other trees do; this is because birches need more calcium than other trees and so it absorbs it better (Sr is chemically analogous to Ca). This is also stated in our previous work, see Jarauta-Bragulat et al (2001 a and b).

Table 1. Correlation coefficients of ^{90}Sr contents for pine tree

	L	R	B	BR1	BR3	LE1	LE3
YCO	0.99	0.98	0.98	0.93	0.97	0.90	0.93
OCO	0.98	0.99	0.96	0.97	0.99	0.95	0.96
HUM	0.96	0.90	0.96	0.80	0.87	0.76	0.83
SO1	0.96	0.91	0.96	0.81	0.88	0.78	0.84
SO2	0.80	0.71	0.84	0.55	0.65	0.50	0.62
SO3	0.70	0.60	0.74	0.42	0.53	0.35	0.49
SO4	0.96	0.96	0.97	0.95	0.95	1.00	0.95

L = log; R = root; B = bark; BR1 = branches 1 year; BR3 = branches 3 years; LE1 = leaves 1 year; LE3 = leaves 3 years; YCO = young cover; OCO = old cover; HUM = humus; SO1, soil at depth 0-5 cm; SO2, soil at depth 5-10 cm; SO3, soil at depth 10-20 cm; SO4, soil at depth greater than 20 cm.

Table 2. Correlation coefficients of ^{90}Sr contents for birch tree

	L	R	S	B	YBR	OBR	LE
YCO	1.00	-0.31	0.98	0.96	1.00	0.92	1.00
OCO	1.00	-0.29	0.93	0.99	0.97	0.97	1.00
HUM	0.93	-0.31	1.00	0.84	0.98	0.77	0.93
SO1	0.93	-0.34	1.00	0.84	0.98	0.77	0.93
SO2	0.74	-0.31	0.91	0.58	0.83	0.47	0.72
SO3	0.63	-0.27	0.83	0.45	0.74	0.34	0.61
SO4	1.00		1.00	1.00	1.00	1.00	1.00

L = log; R = root; S=sap, B = bark; YBR = young branches, OBR = old branches; LE = leaves; YCO = young cover; OCO = old cover; HUM = humus; SO1, soil at depth 0-5 cm; SO2, soil at depth 5-10 cm; SO3, soil at depth 10-20 cm; SO4, soil at depth greater than 20 cm.

Table 3. Correlation coefficients of ^{90}Sr contents for oak tree

	L	B	YBR	OBR	LE
YCO	0.92	0.74	0.81	0.89	-0.45
OCO	0.12	-0.39	0.82	0.72	-0.48
HUM	0.49	0.03	0.98	0.95	-0.57
SO1	0.97	1.00	0.24	0.38	-0.39
SO2	0.77	0.99	0.06	0.21	0.09
SO3	-0.48	0.00	-0.98	-0.93	0.98
SO4	0.99	0.99	0.36	0.50	-0.33

L = log; B = bark; YBR = young branches; OBR = old branches; LE = leaves; YCO = young cover; OCO = old cover; HUM = humus; SO1, soil at depth 0-5 cm; SO2, soil at depth 5-10 cm; SO3, soil at depth 10-20 cm; SO4, soil at depth greater than 20 cm.

The storage of ^{90}Sr in trees depends on the intensity of metabolic processes. Young organs and tissues, which grow up quickly, store ^{90}Sr with more intensity than older ones. However, in pine leaves just the reverse tendency is seen. When pollution is very high this intensity is relatively less than it is when pollution is mean or low. This can be stated from the next three tables (Table 4 for pine trees. Table 5 for birch trees and Table 6 for oak trees). From those tables, looking at different polygons one can verify that ^{90}Sr contents in pine trees is greatly influenced by underground water level. In wet conditions (hydromorphic- D3) the storage of ^{90}Sr in pine trees is higher than in dry conditions (automorphic-D1, K2, Sh1). This feature can also be seen in birch trees, to a lesser extent, but it is not seen in oak trees.

Table 4. ^{90}Sr contents in parts of pine trees, depending on polygon and year.

POL	YEA	L	R	B	BR1	BR3	LE1	LE3
	1996	38.28	170.31	188.01	92.47	105.89	69.10	99.41
	1997	62.41	680.33	440.76	201.75	216.07	123.69	
	1998	33.06	283.60	158.54	140.00	108.00	83.00	215.00
	1999	71.07	323.85	167.21	100.00	150.00	81.00	95.00
D1	2000		370.00	154.02				
	1996	223.49	1344.40	624.25	471.78	609.53	285.40	593.29
	1997	63.75	546.03	426.32	206.76	499.79	94.37	154.91
	1998	131.47	722.23	432.30	540.00	280.00	118.00	391.00
	1999	96.23	564.15	401.41	140.00	240.00		90.00
D3	2000	78.84	465.67	333.68	140.00	190.00	60.00	80.00
	1996	7417.29	46740.61	12123.96	15715.45	22881.53	12945.98	16141.98
	1997	2223.21	24512.96	10346.65	4877.12	7039.91	4237.80	6371.30
	1998	2572.50	19690.81	14704.29	9376.00	7257.00	4156.00	8290.00
	1999	1837.10	13605.48	3974.01	3432.00	3662.00	2236.00	2178.00
K2	2000	5506.34	57782.00	13809.49	21860.00	22750.00	14370.00	22870.00
	1996	32425.17	185312.33	37520.12	30310.64	50167.00	11584.48	19261.22
	1997	26393.07	165926.81	33762.13	21066.95	16888.00	10670.96	8648.00
	1998	26295.31	193719.32	29389.95	67619.00	58642.00	23407.00	61770.00
	1999	35202.63	293098.57	50291.65	118889.00	130925.00	57498.00	75044.00
Sh1	2000	26257.37	165022.99	40977.69	43800.00	64740.00	20380.00	34580.00

L = log; R = root; B = bark; BR1 = branches 1 year; BR3 = branches 3 years; LE1 = leaves 1 year; LE3 = leaves 3 years.

Table 5. ^{90}Sr contents in parts of birch trees, depending on polygon and year.

POL	YEA	L	R	B	BR1	BR3	LE1	S
	1998	392.23	2576.44	394.35	3827.00	1911.00	3217.00	34.00
	1999	406.62	1200.00	1754.99	3200.00	2400.00	3700.00	15.00
D1	2000	334.54						14.00
	1997	357.79		1778.74	3919.00		5883.00	8.50
	1998	195.98	1091.43	282.14	1818.00	281.00	1545.00	11.50
	1999	241.25		880.06	920.00	1900.00	1600.00	16.00
D3	2000	241.97		771.18	2180.00	1140.00	2240.00	32.00
	1996	63311.70		230819.93	374494.81	381576.28		4678.00
	1997	45538.89		146447.88	295857.08		328321.15	2601.00
	1998	46623.42	193700.95	15485.07	550992.00	171587.00	399505.00	3360.00
	1999	47695.93		196825.56	239183.00	267995.00	314210.00	3009.00
Sh1	2000	30079.53		85766.72	201620.00	85190.00	200280.00	3167.00

L = log; R = root; B = bark; BR1 = branches 1 year; BR3 = branches 3 years; LE1 = leaves 1 year; S = sap.

Table 6. ^{90}Sr contents in parts of oak trees, depending on polygon and year.

POL	YEA	L	R	B	BR	LE
D1	1996	202.12	1509.98	511.53	558.47	
D1	1997	31.83	181.57	300.00		215.00
D1	1998	34.19	750.97	317.00	215.00	510.00
D1	1999	63.19	880.44	1200.00	1100.00	
D1	2000	44.72				835.00
D3	1997	41.49	2410.20		811.00	570.00
D3	1999	145.92	2025.99	1100.00	1100.00	
D3	2000	49.32	1076.41	580.00	350.00	

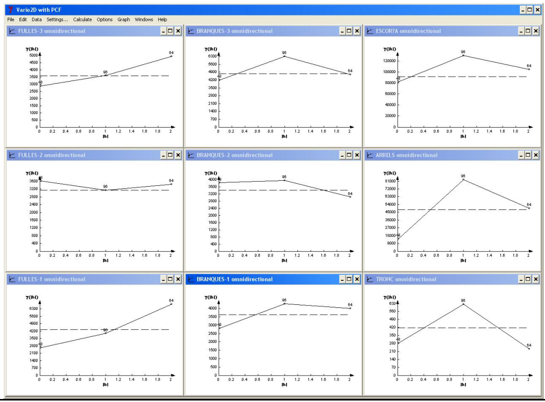
L = log; R = root; B = bark; BR = branches; LE = leaves.

Geostatistical analysis

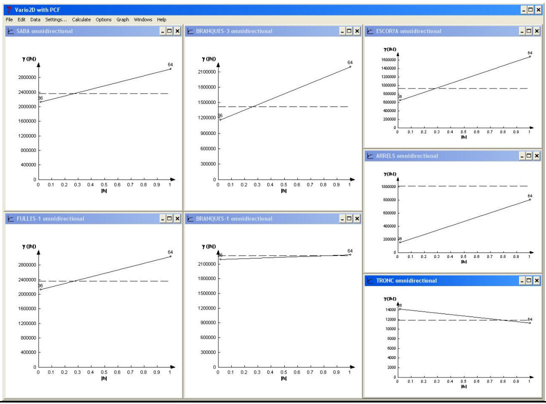
To do the geostatistical study, that is, to analyse the behaviour of the different tree species depending on the polygon and its characteristics, we firstly observe that polygons Sh1 and K2 cannot contribute to any significant interpretation because there is not enough information. So study has been centred in polygons D1 (automorphic) and D3 (hydromorphic). In Figures 2 and 3 their corresponding variograms are shown. From the study of those variogram structures, our aim is to find out what species is a better indicator of soil pollution.

Figure 2. Polygon D1: variograms of ^{90}Sr contents of studied different parts in pine (a), birch (b) and oak (c) trees.

(a)



(b)



(c)

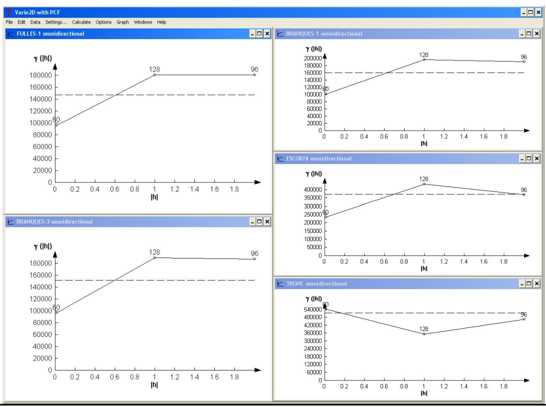
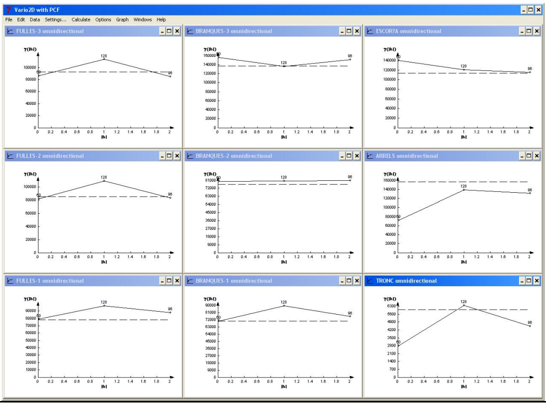
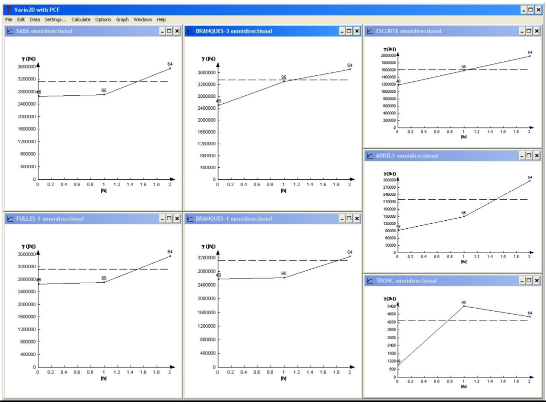


Figure 3. Polygon D3: variograms of ^{90}Sr contents of studied different parts in pine (a), birch (b) and oak (c) trees.

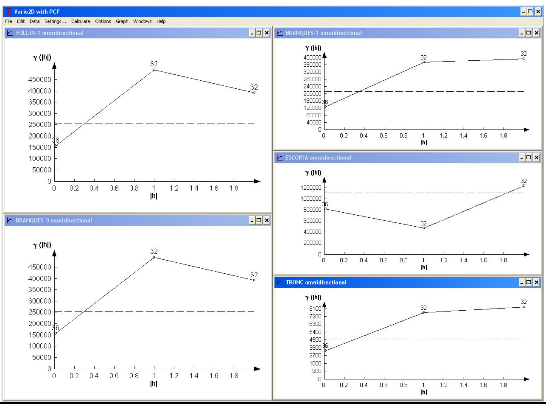
(a)



(b)



(c)



From the observation of those variograms one can point out the following:

1. Pine trees variograms do not always show a structure and show great differences from one part to another one.
2. Birch trees variograms are relatively well structured and their behaviour is much more homogeneous among different parts.
3. Oak trees variograms show a better behaviour than pine trees variograms but they are not so well structured as birches and have some anomalies in bark and log.

From those remarks one can conclude that birches are better indicator of soil pollution than others. This conclusion is coherent with results obtained in Jarauta-Bragulat et al (2001 a and b).

Table 7. ^{90}Sr contents (Bk/kg on dry weight) in soils on years 1999-2000

POL	YEA	YCO	OCO	HUM	SO1	SO2	SO3	SO4
D1	1999	2586	3830	5086	536	86	19	
	2000	1730	1790	2260	550	190	70	
D3	1999	3478	2416	4174	2564	288	23	11
	2000	1530	1850	2330	830	150	30	
K2	1999	9973	28319	20576	1280	336	170	44
	2000	9890	21360	40770	2780	920	271	
Sh1	1999	435490	758777	1186412	18539	1874	523	112
	2000	317290	411190	1388670	21070	4190	1845	

POL, polygon; YEA, year; YCO, young cover; OCO, old cover; HUM, humus; SO1, soil at depth 0-5 cm; SO2, soil at depth 5-10 cm; SO3, soil at depth 10-20 cm; SO4, soil at depth greater than 20 cm.

Results and discussion

As moisture was very different from one year to another, it was necessary to measure ^{90}Sr contents in soil to assess its radial migration; results are shown in Table 7. Atmospheric precipitation in 1999 was about 556.8 mm and in 2000 it was 633.5 mm. From radiogeochemical research in Chernobyl zone, one knows that humidity conditions have a great influence on ^{90}Sr release from organic matter (boughs, twigs and leafs that make a soil coverage), its radial migration to the surroundings and its transfer to soil solutions [Ovsiyannikova et al, 2000; Mixailovsakaiya et al., 1999; Agapkina, 1999]. For example, its release from organic matter in hydromorphic conditions (very wet) is about 9-10 times greater than in normal humidity conditions (automorphic) [Mixailovsakaiya et al., 1999]. However, up going ^{90}Sr migration rate from low soil soaked sheet to aquifer is smaller than in drier conditions [Ovsiyannikova et at., 2000]. Those features can be easily seen in our data set. ^{90}Sr release from organic matter and its transfer to mineral soil in a normal humidification year (1999) in D3 polygon (hydromorphic) is 5.4

times greater for 0-5 cm layer and 4.8 greater for 5-10 cm layer than in D1 polygon (automorphic). This law was violated for a wet year (2000) due to up going migration processes, when D3 was quite a marsh.

For all automorphic polygons (D1, K2, Sh1) another feature of ^{90}Sr migration rate in wet conditions can be seen when comparing 2000 and 1999 data: rate was higher in wet conditions (2000) than in drier ones (1999) for all soil layers (0-5, 5-10, 10-20 cm). In marshy conditions (D3) this trend was inverted. So we can verify the great influence that moisture conditions have on ^{90}Sr migration rates. This moisture can come from aquifers and form atmospheric precipitations.

Conclusions

1. ^{90}Sr contents in tissues and organs in pine, birch and oak trees in Chernobyl Zone (Ukraine) is in correlation to soil radioactive pollution.
2. ^{90}Sr in pine trees is greatly influenced by underground water level. In wet conditions (hydromorphic) the storage of ^{90}Sr in pine trees is higher than in dry conditions (automorphic). This feature can also be seen in birch trees, to a lesser extent, but it is not seen in oak trees.
3. The incoming of ^{90}Sr to a plant comes from the polluted soils through the roots and from the air through bark and leaves. Some vegetal tissues react to radical or aerial pollution. The aerial incoming of ^{90}Sr in a tree depends essentially of climatic conditions. In dry periods ^{90}Sr enrichment is seen for high bark (especially for oak which is highly rough).
4. The storage of ^{90}Sr in trees depends on the intensity of metabolic processes. Young organs and tissues, which grow up quickly, store ^{90}Sr with more intensity than older ones. However, in pine leaves just the reverse tendency is seen. There can also be observed that birches absorb better ^{90}Sr than other trees do; this is because birches need more calcium than other trees and so it absorbs it better (Sr is chemically analogous to Ca). When pollution is very high this intensity is relatively less than it is when pollution is mean or low.
5. Looking to trees as a whole, we can observe that birches are better indicator than pine or oak trees of soil and underground water ^{90}Sr pollution.

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