THE FIRST DATA OF DENDROGEOCHEMICAL ANALYSIS OF THE MULTI-YEAR DYNAMICS OF POLLUTION OF PETROZAVODSK

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An attempt to analyze the dynamics of a regionôs pollution using data on the content of chemical (radioactive) elements in annual rings of a tree (pine) was undertaken for the first time in 1993 in the Medvezhegorsk Region [13]. The content of elements in the annual rings was analyzed using local laser spectrometry method [10].

Similar studies have continued on three core samples of 32-33 year-old pines taken on February 26, 1998 in Petrozavodsk (two samples) and near the Silicate Factory, which is not currently in operation, on the outskirts of the city (one sample). Cores with a diameter of 0.5 cm were extracted using a special auger 25-30 cm above the ground surface. The content of 34 elements was analyzed in each annual ring and in the tree bark (a total of 104 samples analyzed) by the above-mentioned laboratory method: U. Th, Ra, W, Ac, Sr, Pu, Tl, Bi, Tc, Co, Ni, V, Pb, P, Mn, La, Zn, Ba, Sc, Ti, Ag, Cr, Cd, [illegible], Hg, Y, Ga, Ge, Zr, Mo, Nb, Ta, and As.

Pb, Sr, V, Co, Ni, Zn, Cr, Ti, Mn, Ba, and P are present in various concentrations in the annual pine rings. The content ranges are shown in Table 1. Maximum error in determining the element content was 12%. This method detected certain elements only in the tree bark (Table 1).

The following elements were not detected either in the wood or in the bark of the pines (the threshold of detection is shown in parentheses): Pu, La (10^{-3}% mass), U, Th, Ra, Ac, Tc, Bi, Y, Hg, Ga, Ge, Zr, Mo, Nb, Ta, As (10^{-4}% total mass), Sc (10^{-4}% total mass), Ti (10^{-4}% mass).

A brief description is given below of the dynamics of the distribution of the most important pollutants in the annual pine rings: Pb, Sr, V, Co, Ni, and Cr, as well as a nutrient v. element, such as P.

During the study, a factor analysis of samples was performed separately on each sample to identify the specific features of the relationship between the elements as they accumulated in the pine wood.

Lead

The virtually ubiquitous Pb pollution of the environment is caused by the use of its compounds as additives to increase the octane rating of gasoline. At present, Pb emissions from vehicles have been significantly reduced in many Western European cities thanks to the increase in the market share of unleaded gasoline [12].

On the national government level, the Interdepartmental Commission of the Russian Federation Security Council examined the problem of lead pollution in Russia in May of 1995 [15]. The Commission concluded that a regional approach was necessary ñto identify predominant sources of lead pollution (vehicular transport, industrial enterprises, etc.) and their effect on public health.ò

The studies we have done showed that Pb content in annual rings of trees growing in Petrozavodsk [illegible]% total mass Pb (Table 1).

Table 1. Content of several elements in the annual rings (a.r.) and the bark of pines (o.b.l. ï outer layer of bark, i.b.l. ï inner layer of bark), % total mass Ā10^{-4}

Substance Pb W Sr Co Ni V Zn Ti Mn He Ag Cr Cd Ba P, % Ā10^{-3}

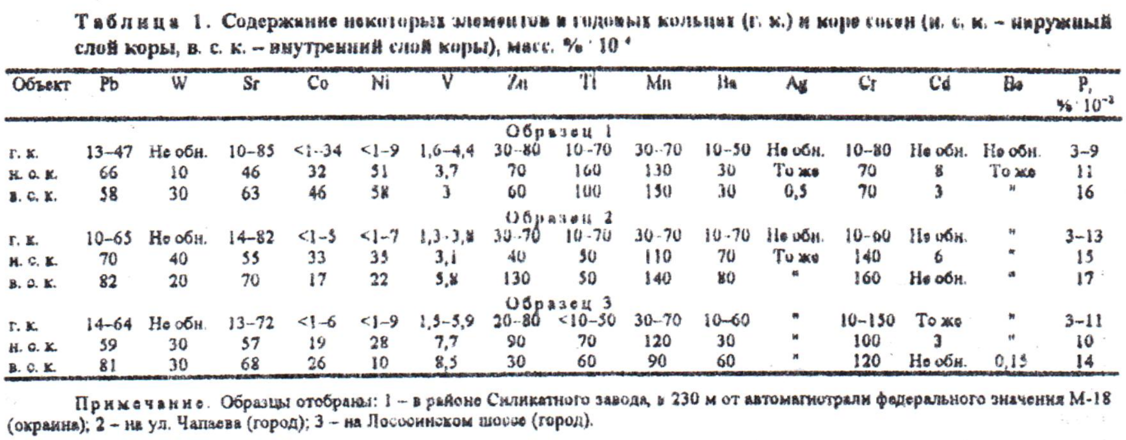

Таблица 1. Содержание некоторых элементов в годовых кольцах (г. к.) и коре сосен (н. с. к. – наружный слой коры, в. с. к. – внутренний слой коры), масс. % · 10^{-4}

Объект	Pb	W	Sr	Co	Ni	V	Zn	Ti	Mn	Hg	Ag	Cr	Cd	Ba	P, % · 10^{-3}
							Образец 1								
г. к.	13–47	Не обн.	10–85	<1–34	<1–9	1,6–4,4	30–80	10–70	30–70	10–50	Не обн.	10–80	Не обн.	Не обн.	3–9
н. с. к.	66	10	46	32	51	3,7	70	160	130	30	То же	70	8	То же	11
в. с. к.	58	30	63	46	58	3	60	100	150	30	0,5	70	3	"	16
							Образец 2								
г. к.	10–65	Не обн.	14–82	<1–5	<1–7	1,3–3,8	30–70	10–70	30–70	10–70	Не обн.	10–60	Не обн.	"	3–13
н. с. к.	70	40	55	33	35	3,1	40	50	110	70	То же	140	6	"	15
в. с. к.	82	20	70	17	22	5,8	130	50	140	80	"	160	Не обн.	"	17
							Образец 3								
г. к.	14–64	Не обн.	13–72	<1–6	<1–9	1,5–5,9	20–80	<10–50	30–70	10–60	"	10–150	То же	"	3–11
н. с. к.	59	30	57	19	28	7,7	90	70	120	30	"	100	3	"	10
в. с. к.	81	30	68	26	10	8,5	30	60	90	60	"	120	Не обн.	0,15	14

Примечание. Образцы отобраны: 1 – в районе Силикатного завода, в 230 м от автомагистрали федерального значения М-18 (окраина); 2 – на ул. Чапаева (город); 3 – на Лососинском шоссе (город).

Note. Samples taken: 1 ï in the area of the Silicate Factory, 230 m from Federal Highway M-18 (outskirts); 2 ï on Chapaev Street (city); 3 ï on Lososinskoe Highway (city).

Analysis of distribution of Pb content in annual rings revealed a general trend toward an increase in lead pollution of Petrozavodsk in all samples from the end of the 1970s to the present (Figure 1). There was an especially significant increase (four- to six-fold) in samples taken within a 50-meter strip along the edges of the city highways. For one of them (Lososinskoe Highway), a small reduction in Pb content is noted only in the last two annual rings. Most likely the reduction in level of Pb pollution in this zone is due to the temporary closure of the bridge (along Gogol Street) for repair, guaranteeing the heaviest automobile traffic over that highway. The zone, located on the outskirts of the city 230 m from Federal Highway M-18, is characterized by a somewhat lower level of lead pollution. Nonetheless, even here the Pb content has increased by a factor of 2.3 over the last 20 years, from 1965 to the present ï by a factor of 3.6.

The author of this paper, Dmitry Rybakov, is a geologist from Karelia, Russia. The technique detailed in his study reveals past contamination by taking samples from trees, allowing dates of radioactive releases to be determined scientifically. The paperôs first presentation in America by Mr. Rybakov was on September 7, 2004 in Augusta, Georgia. His visit was sponsored by the Blue Ridge Environmental Defense League working under a grant from the ISAR Open World Russia Civic Program in Washington, DC. For more information, contact:

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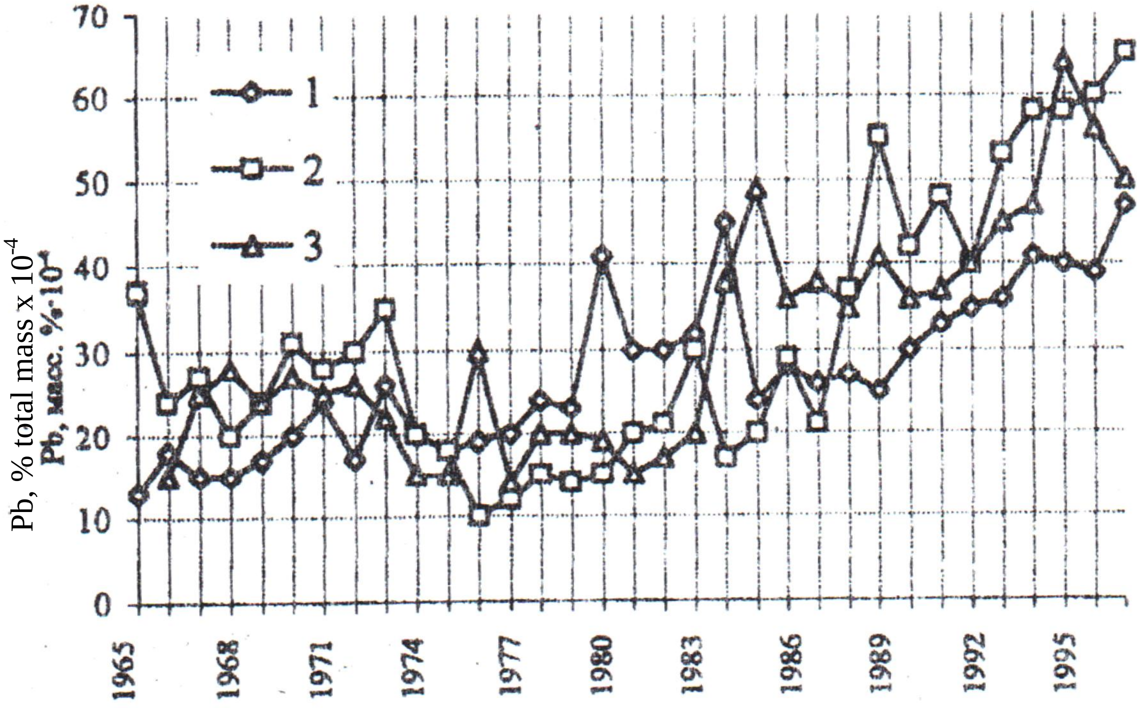

Рис. 1. Динамика распределения Pb по годовым кольцам сосен:

1–3 – номера образцов: 1 – район Силикатного завода, 2 – улица Чапаева, 3 – Лососинское шоссе

Figure 1. Dynamics of Pb distribution in annual rings of pines:
1-3 ï sample numbers: 1 ï Silicate Factory area; 2 ï Chapaev Street; 3 ï Lososinskoe Highway

The general systematic increase in lead pollution, of which automobile emissions are obviously the main cause, shows almost no local spikes associated, possibly, with other sources of pollution, for example, the cityôs coal-fueled railroad enterprises, etc., operating in the area. It might be possible to judge the role of these sources in the dynamics of Petrozavodsk Pb pollution after the problems of lead pollution from vehicular transport are solved.

The average lead content in the annual rings of pines growing in Petrozavodsk and its surroundings has increased by a factor of 3.5 from the mid-70s to the present, reaching reached 0.0054% total mass in 1997 (Figure 2). The control region, where there is no high-level lead pollution (1.2 km south of Pindush Lake, the Medvezhegorsk Region of Karelia Republic) exhibited no similar increase.

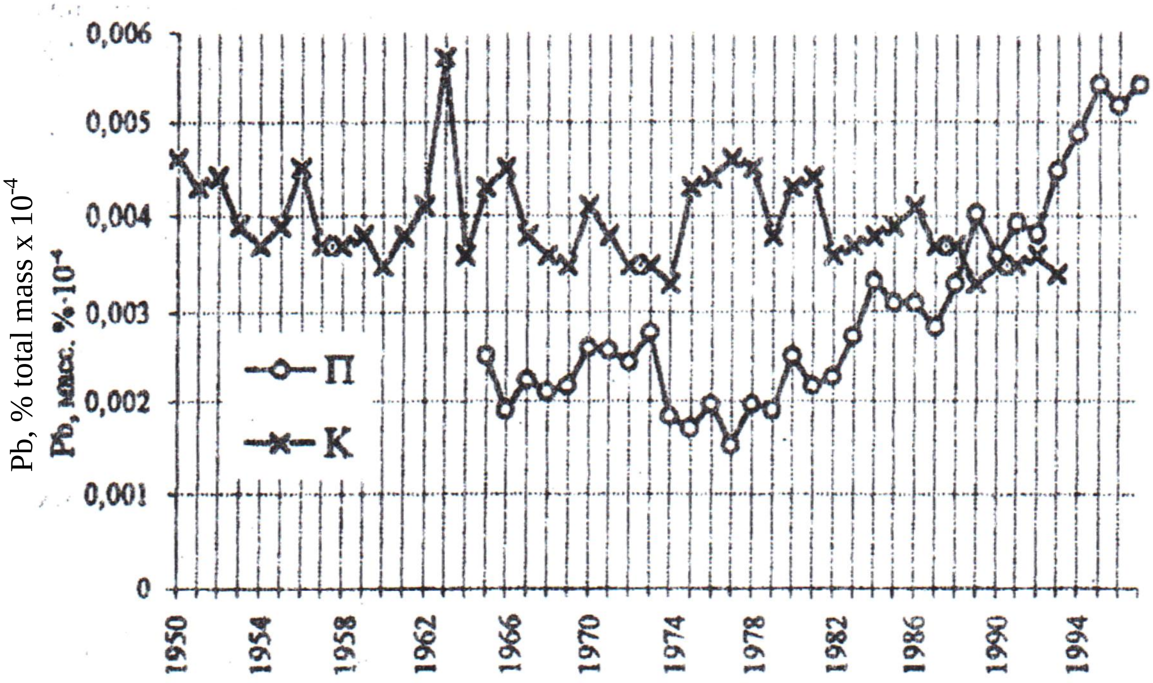

Рис. 2. Распределение Pb по годовым кольцам сосен:
П – Петрозаводск (интегрированный график по трем образцам); К – контроль (Медвежьегорский район)

Figure 2. Distribution of Pb in annual rings of pines:
ɬ ï Petrozavodsk (integrated graph for three samples); K ï control (Medvezhegorsk Region)

Thus, the dynamics of Pb distribution in annual pine rings indicates an ominous trend toward an increase in pollution of the capital of Karelia by this extremely hazardous toxic heavy metal.

Strontium

The main anthropogenic source of Sr entering the environment is industrial production [1]. In the meantime, the increased Sr content that we recorded earlier [13] in the annual rings of a pine tree from Medvezhegorsk Region corresponding to 1985-1986 is presumably associated with pollution of the territory by a radionuclide of ^{90}Sr as a result of the Chernobyl accident in April 1986. It was possible that there were other potential sources of global or regional pollution. Study of the three pine core samples taken in Petrozavodsk revealed a similar pattern.

The Sr content in the annual pine rings varies from 0.001 to 0.0085% total mass (Table 1). The bark contains Sr in the following quantities: 0.0046-0.0057% total mass (outer layer) and 0.0063-0.007% total mass (inner layer).

Maximum Sr concentrations in all samples occurred in 1985 (Figure 3). As shown by the graphs, the Sr content in rings corresponding to that year sharply increases in comparison with preceding years (by a factor of approximately 2 to 5.5) and then decreases, while the rate of reduction in the element proves to be different for different zones. While the Sr content in annual pine rings from the area around the Silicate Factory has decreased more than fivefold over the last 3-4 years in comparison with the 1985 ring, there has been a similar reduction, but only twofold, in a pine growing in the city (sample 3) over approximately 10 years.

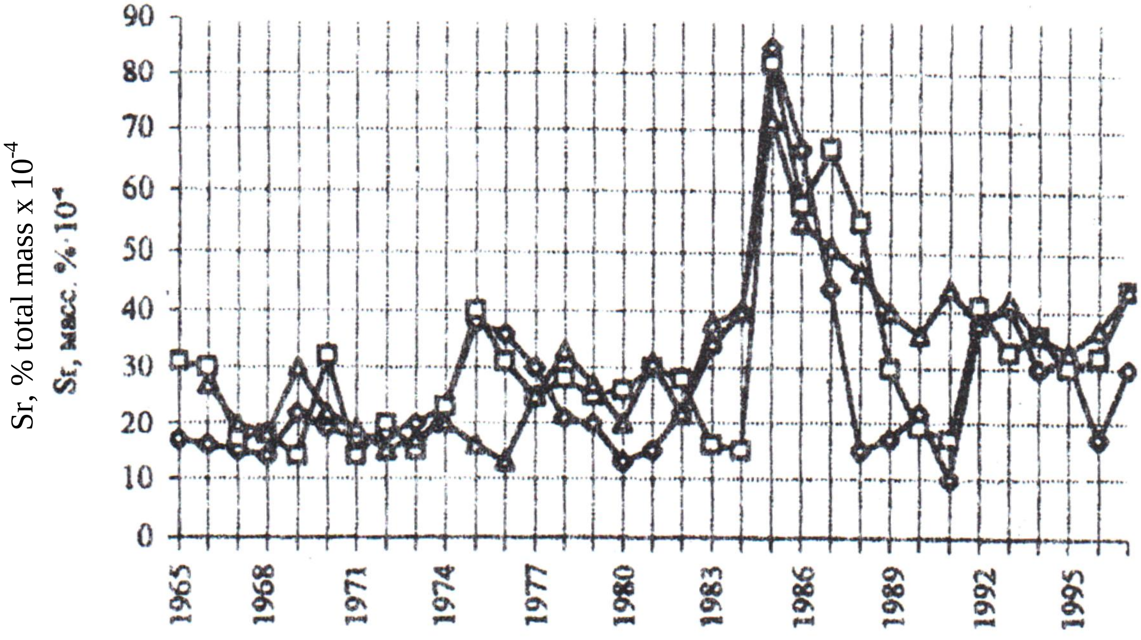

Рис. 3. Динамика распределения Sr по годовым кольцам сосен

Здесь и на рис. 4–8 обозначения, как на рис. 1

Figure 3. Dynamics of Sr distribution in annual pine rings
Here and in Figures 4-8, the legend is the same as in Figure 1

The hypothesis that Sr in the form of radioactive isotope ^{90}Sr was deposited in the wood and the bark of pines after the Chernobyl accident looks fairly accurate. In the spring of 1986, pollution evidently moved through the external tissues (epidermis and periderm) because of the rapid movement of substances taken up through pores and lenticels via the phloem, initially adjacent to the 1985 layer, and then to the 1986 growing layer. In subsequent [illegible line] of ^{90}Sr entering the wood through the tree roots. The difference in the rate of reduction of Sr concentrations in annual pine rings may be associated, on the one hand, with the differing degree of availability of ^{90}Sr depending on the particular soil conditions (Ca concentrations in the soil, acidity, and content of organic matter [14]). On the other hand, it may depend on the degree of pollution of different zones by heavy metals that destroy the physiological barriers in plants. The latter hypothesis will be discussed below.

Additional (secondary) pollution of pines by the radionuclide at the beginning of the 1990s possibly occurred due to redistribution of Sr in the surrounding root environment as a result of processes associated with natural reworking of the forest floor.

For public safety, special research must be undertaken in connection with the pollution of different habitats by ^{90}Sr and other radionuclides. It is necessary to pay special attention to the soil, plants, residues, fish, the flesh of animals, and human biosubstrates.

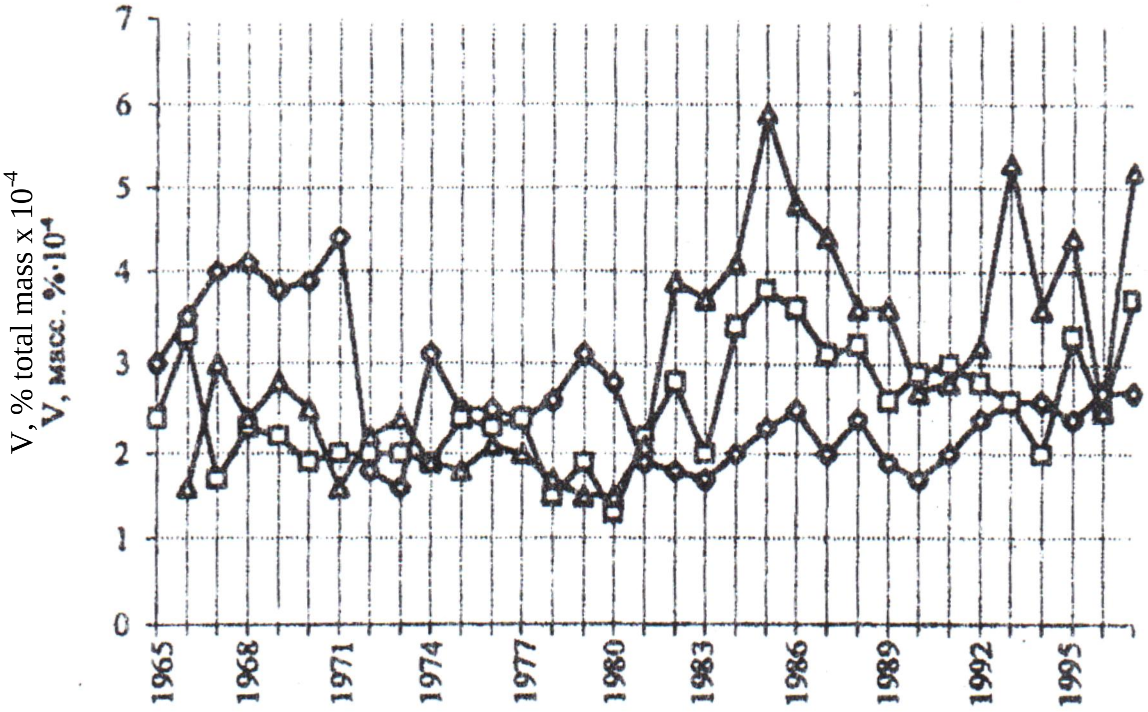

Рис. 4. Динамика распределения V по годовым кольцам сосен

Figure 4. Dynamics of V distribution in annual pine rings

Vanadium

The most serious source of vanadium pollution is the burning of fuel oil [2, 9 et al.]. Other sources may be coal burning, ore concentration enterprises, ventilation emissions of different industrial processes, and transport, including diesel and aviation.

The vanadium content in annual rings of pines ranges from 1.3-1.6 Ā10^{-4} to 3.8-5.9 Ā10^{-4}% total mass. The tree bark contains from 3 Ā10^{-4} to 8.5 Ā10^{-4}% total mass of V (Table 1).

The end of the 1970s marked the beginning of intensive operation of the Petrozavodsk thermal power plant, which operated until the end of 1997 exclusively on fuel oil. The startup in 1979-1981 of three power-generating boilers (alongside two hot-water units) is reflected in the increase in V content in annual pine rings, beginning with the ring that formed in 1981 (Figure 4). The degree of pollution of the bio-subject (pines) by V depends on the prevailing wind directions in relation to the thermal power plant [11]: Lososinskoe Highway has exhibited the maximum fluctuation in V content since the beginning of the 1980s; the Silicate Factory area, the minimum. In the latter case the relatively high V content before 1980 in comparison with the later period may be explained by the effect of emissions from the Silicate Factory boiler. The relatively sharp increase in V content in samples from Lososinskoe Highway and Chapaev Street in rings corresponding to 1985 are probably related to the destruction of physiological barriers due to stress caused by radioactive Chernobyl fallout in the spring of 1986.

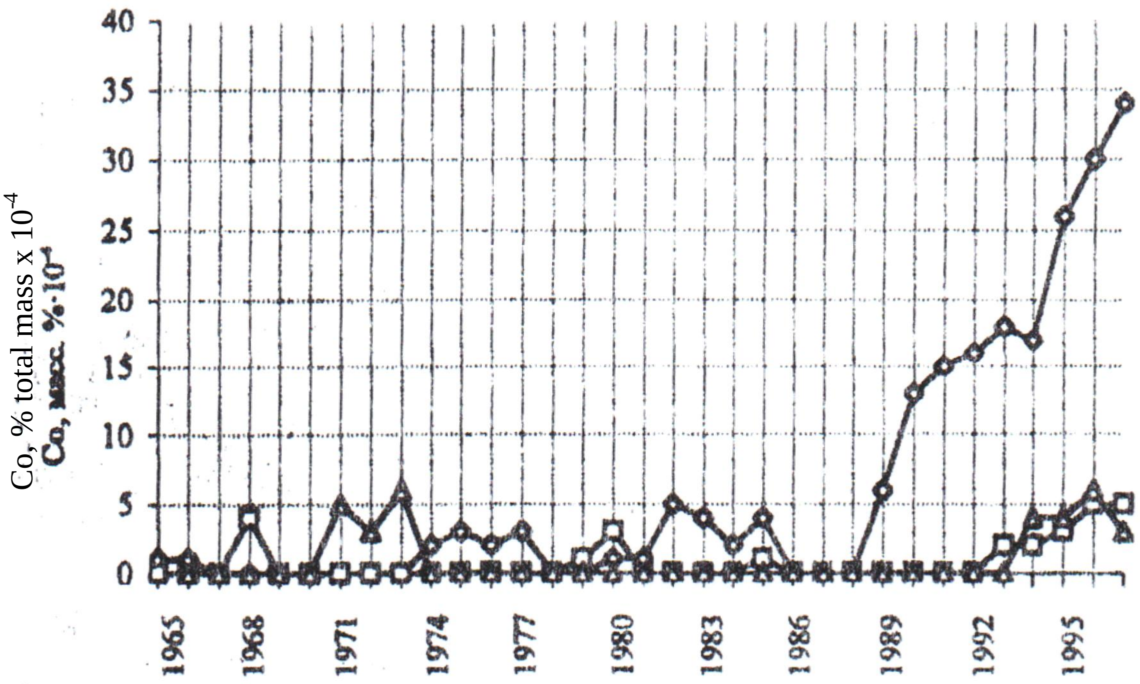

Рис. 5. Динамика распределения Co по годовым кольцам сосен

Figure 5. Dynamics of Co distribution in annual pine rings

Variations in V content in annual pine rings may depend on the quantity and quality of the fuel oil burned in the thermal power plant and on partial replacement of the type of fuel. The change in V content (% total mass) from 1993 through 1996 for samples taken directly in the city is comparable to the changes in overall quantity (thousands of tons) of emissions of the Petrozavodsk thermal power plant, for example, for sample 3 (sources with respect to emissions [4-7]): 1993 ï 5.3 Ā10^{-4} and 19.515; 1994 ï 3.6 Ā10^{-4} and 14.904; 1995 ï 4.4 Ā10^{-4} and 15.799 (supply of low quality fuel oil); 1996 ï 2.5 Ā10^{-4} and 13.487 (conversion of two boilers to natural gas). Sample 2 exhibits similar changes in V content (1993 ï 2.6 Ā10^{-4}; 1994 ï 2 Ā10^{-4}; 1995 ï 3.3 Ā10^{-4}; and 1996 ï 2.5 Ā10^{-4} % total mass). It is likely that these circumstances may attest also to the mainly superficial introduction (through protective tissues) of V into the pine wood. The predominance of this mechanism in relation to these two samples is indirectly confirmed by the large accumulation of V in the inner layer of the tree bark in comparison with the outer layer (Table 1). For the zone less polluted by V (sample 1) the ratio is inverse.

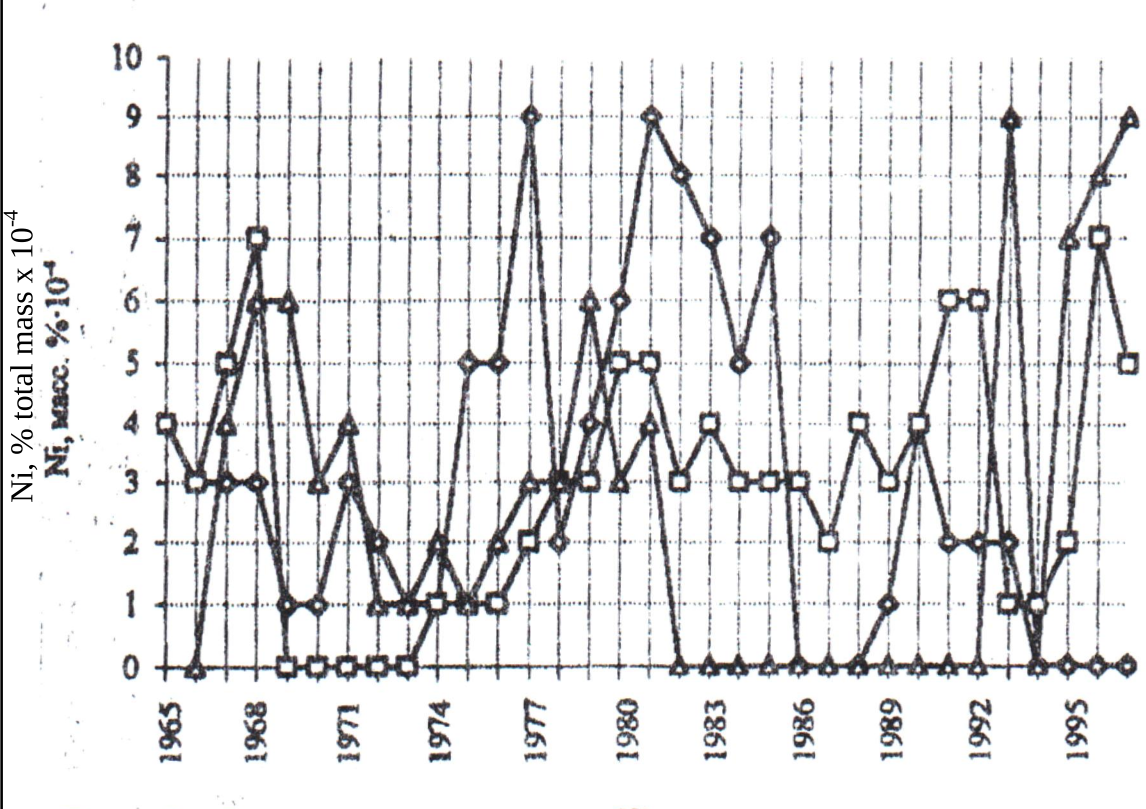

Рис. 6. Динамика распределения Ni по годовым кольцам сосен

Figure 6. Dynamics of Ni distribution in annual pine rings

Cobalt

One of the reasons for Co pollution of the environment is the burning of fuels containing this metal during industrial production. The smelting of nonferrous metals is a significant source of Co pollution [2].

The Co content in annual pine rings of pines occurs in the following ranges: for samples taken in the city ï from $<10^{-4}$ to 5-6 Ā10^{-4} % total mass; for a sample from the Silicate Factory area ï from $<10^{-4}$ to 34 Ā10^{-4} % total mass. The tree bark contains Co from 17 Ā10^{-4} to 46 Ā10^{-4} % total mass (Table 1).

As shown in the diagram (Figure 5), local periodic Co pollution of the Petrozavodsk urban environment was recorded throughout the entire studied period. The largest (anomalous) increase in Co content has occurred since the end of the 1980s in the sample from the Silicate Factory area, where the maximum content of the metal, is tens, and perhaps, even hundreds of times higher than the primary (background) level recorded in the annual ring corresponding to 1997. A similar, but lesser increase in Co content beginning later occurs in samples taken directly in the city. The latter circumstance suggests that the increase in Co content in the city environment may be associated not only with local sources, but also with regional factors. This in particular is confirmed by the research of N. G. Fedorets and others on mosses and the forest floor [8].

Nickel

The most important sources of Ni pollution of the environment are mining, nonferrous metallurgy, machine-tool, and metalworking enterprises. Ni also occurs in the biosphere as a result of burning fuel oil, coal, diesel fuel and from other industrial causes [2, 9].

[illegible line] of the Ni content in annual pine rings may be judged by the graphs in Figure 6. Analysis shows the extremely broad spread of the element content (from $<10^{-4}$ to 7-9 Ā10^{-4} % total mass) and the presence of several peaks in its level of concentration for each sample.

For the sample taken in the Silicate Factory area, these peaks, corresponding to approximately 1977 and 1981-1985, may be associated with Ni-containing emissions of the boiler factory. It is possible that it was the shutdown of the enterprise that subsequently caused the gradual reduction in Ni content in annual pine rings to the point of undetectable concentrations (1994-1997). On the other hand, it has been seen [2] that ions of several elements (Co, Cu, Fe, Zn) inhibit absorption of Ni by 25-42%. However, it is also possible that the ñdisappearanceò of Ni was facilitated by exceedingly high concentrations of Co in this specimen. Thermal power plant and vehicular emissions may make a certain contribution to nickel pollution of the two relatively elevated city zones (Lososinskoe Highway and Chapaev Street). Meanwhile, for all samples, excluding the ñcobalt anomalyò mentioned above, the variations in Ni and Co content closely coincide (Figures 5, 6). The Ni content in the pine root is significantly higher than in the wood (Table 1).

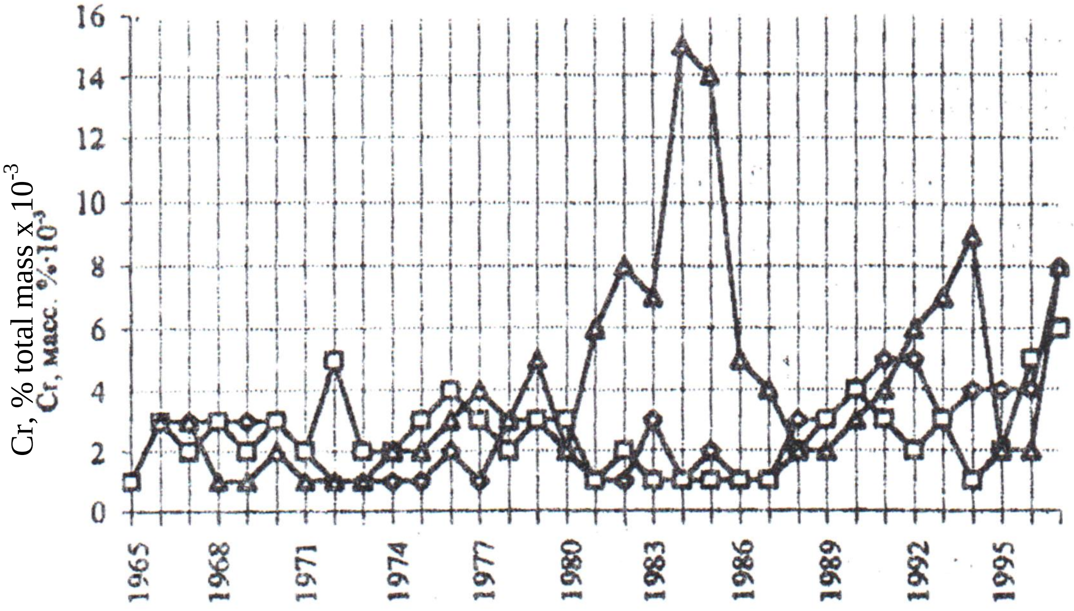

Рис. 7. Динамика распределения Cr по годовым кольцам сосен

Figure 7. Dynamics of Cr distribution in annual pine rings

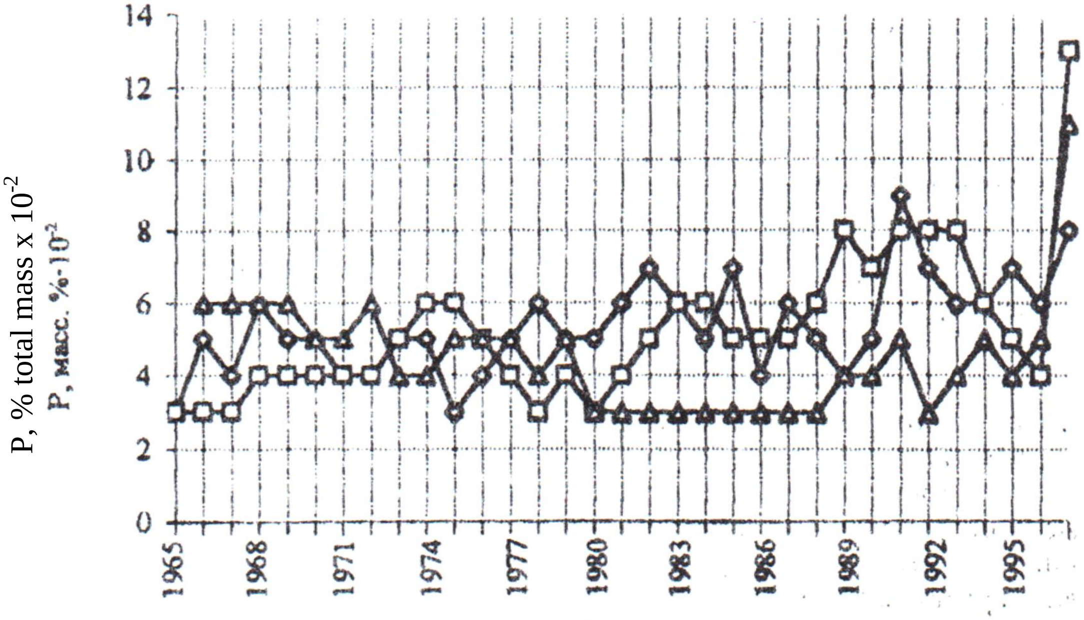

Рис. 8. Динамика распределения P по годовым кольцам сосен

Figure 8. Dynamics of P distribution in annual pine rings

Chromium

Natural ambient air contains an extremely insignificant quantity of Cr; virtually all of it is confined to the very fine dust particles [3]. The main anthropogenic sources of Cr and its compounds in the atmosphere are emissions of enterprises where they mine, produce, process and use Cr and its compounds. Active dispersion of Cr is associated with burning mineral fuel, mainly coal. Significant amounts of Cr enter the environment with industrial wastes [2].

The Cr content in annual pine rings varies rather widely ï from 0.001 to 0.006-0.015% total mass. The root contains 0.007-0.016% total mass of Cr (Table 1).

Anomalous periodic increases in Cr content in annual pine rings were recorded for the sample taken on the Lososinskoe Highway (Figure 7): in the rings corresponding to 1981-1984 ï up to 0.015% total mass, against a background of about 0.001-0.002% total mass (ñfirst chromium anomalyò) and 1991-1994 ï up to 0.009% total mass (ñsecond chromium anomalyò).

Phosphorus

Phosphorus, along with Ca, Mg, K and N, is a key component of food, and its deficit is reflected in the growth and development of plants. The balance of P and other nutritive substances is critical for plants [9].

The content of P in annual rings of the pine cores varies from 0.03 to 0.09-0.13% total mass. The tree roots contain 0.1-0.17% total mass.

Peak concentrations of P were observed in the last annual ring, 1997, for samples taken on the Lososinskoe Highway (0.11% total mass) and on Chapaev Street (0.13% total mass). In comparison with the previous ring, concentrations increased suddenly by a factor of 2.2 and 3.25 [two lines illegible] amounted to 1.3 times. Such a correlation may indicate an additional accumulation of P at the end of the growing season.

Results of factor analysis (FA)

Samples for FA were taken after rejecting the values for content of various elements that were anomalous according to criterion 3s. Table 2 presents the factor weights for three samples for several elements according to factors I and II, which describe a total of about 50-60% of the total dispersion. This matrix shows the change in the correlations of elements from sample to sample. For samples 1 and 2, taken in zones less polluted by emissions by the Petrozavodsk thermal power plant, the factor weights for Pb and P for factor I have the same sign (Pb accumulates in plants in the form of ortho and pyrophosphates [9]). At the same time, for sample 3 (Lososinskoe Highway), the factor weights have opposite signs. The direct correlation of Pb and P for the last sample is noted only factor II. The nature of the relationship with the other elements (Zn, V, Sr, Mn, and Ti) also changes consistently.

Table 2. Factor Weights for Three Samples

Factors Elements Sample 1 Sample 2 Sample 3

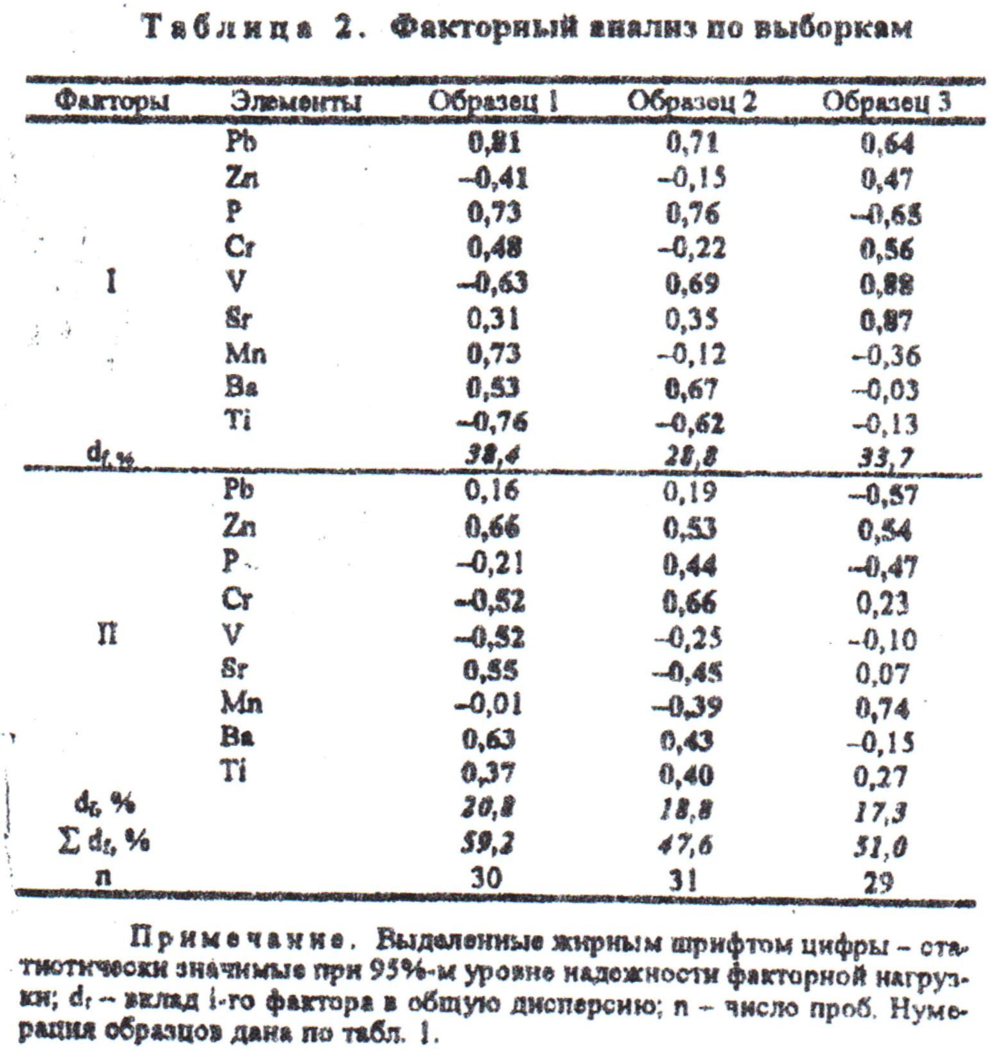

Таблица 2. Факторный анализ по выборкам

Факторы	Элементы	Образец 1	Образец 2	Образец 3
I	Pb	**0,81**	0,71	**0,64**
	Zn	**–0,41**	–0,15	**0,47**
	P	**0,73**	**0,76**	**–0,65**
	Cr	**0,48**	**–0,22**	**0,56**
	V	**–0,63**	**0,69**	**0,88**
	Sr	0,31	0,35	**0,87**
	Mn	**0,73**	–0,12	–0,36
	Ba	**0,53**	**0,67**	–0,03
	Ti	**–0,76**	**–0,62**	–0,13
d_{I}, %		*38,4*	*28,8*	*33,7*
II	Pb	0,16	0,19	**–0,57**
	Zn	**0,66**	**0,53**	**0,54**
	P	–0,21	**0,44**	**–0,47**
	Cr	**–0,52**	**0,66**	0,23
	V	**–0,52**	–0,25	–0,10
	Sr	**0,55**	**–0,45**	0,07
	Mn	–0,01	**–0,39**	**0,74**
	Ba	**0,63**	**0,43**	–0,15
	Ti	**0,37**	**0,40**	0,27
d_{I}, %		*20,8*	*18,8*	*17,3*
Σd_{I}, %		*59,2*	*47,6*	*51,0*
n		30	31	29

Примечание. Выделенные жирным шрифтом цифры – статистически значимые при 95%-м уровне надежности факторной нагрузки; d_{I} – вклад i-го фактора в общую дисперсию; n – число проб. Нумерация образцов дана по табл. 1.

Note. The numbers highlighted by boldface type are statistically significant at the 95% level of factor weight reliability; d_1 ï contribution of first factor to total dispersion;
n ï number of specimens. Enumeration of samples is given in Table 1.

Comparing these data with the data obtained by analyzing the diagrams of dynamics of element distribution, it is possible to conclude that the degree of penetration of pollutant elements into the wood of plants depends both on the level of pollution of the respective areas and probably on the specific composition of the pollutants. Judging from all of the above, the simultaneous penetration into a plant of heavy metals and radioactive elements significantly aggravates destruction of the physiological barriers within it. This also promotes the additional accumulation of V, Sr, Pb, Cr and Zn in the annual pine rings, leading in turn to a reduction in concentrations of elements that are vital for plants, such as P and Mn.

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A brief description of the dynamics of distribution of key pollutant elements (Pb, Sr, V, Co, Ni, Cr) and P in the annual rings of pines growing in Petrozavodsk and in the immediate surroundings. To identify the specific nature of the relationships of Pb, Zn, Sr, V, Cr, Mn, Ba, Ti and P as they accumulate in wood, a factor analysis is performed on samples taken separately on each of three of the core samples. The ranges of concentrations of microelements in the annual rings and the root of pines are presented.