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Industrial Pollution Reduces the Effect of Trees on Forming the Patterns of Heavy Metal Concentration Fields in Forest Litter

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Abstract—An analysis is made of the effect of large spruce and birch trees on the spatial pattern of the fields of heavy metal concentrations (Cu, Pb, Cd, Zn) and pH in the forest litter formed in tree stands exposed to long-term pollution with emissions from the copper smelter in Revda, Sverdlovsk oblast. The fields formed by trees growing in the background area have a regular spatial structure: the concentrations of elements decrease with increase in the distance from the tree trunk to the edge of canopy gap, with the position of sampling point relative to the trunk accounting for more than half of total variance. In polluted areas, the regular component of the field structure is very weakly expressed, and the main role is played by higher-order heterogeneity related to the mosaic pollution pattern on the scale of tens to hundreds of meters.

Keywords: habitat-forming ability, edificators, ecosystem engineers, phytogenic field, ecological field, distance from tree trunk, stemflow, throughfall, soil, air pollution, point polluters, acidity, microscale spatial distribution, variance components

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Trees in the forest are a classic example of ecosystem engineers (Jones et al., 1994) that significantly modify the physical and chemical environment and thereby shape the spatial structure of forest communities. A well-known fact is that tree crowns redistribute atmospheric precipitation and alter its chemical composition (Uchvatov and Glazovskii, 1984; Bergkvist et al., 1989; Levia and Frost, 2003, 2006; Keim et al., 2005; Staelens et al., 2006; Andre et al., 2011; Bialkowski and Buttle, 2015; Frischbier and Wagner, 2015; Levia and Germer, 2015) and produce an effect on the light, thermal, and wind regimes of habitats (Ipatov and Kirikova, 2001), the moss and herb–dwarf shrub vegetation layers, and tree stand regeneration (Coates, 2000; Kryshen', 2000; Lebedeva et al., 2005; Miller et al., 2006). All this has influence on morphological, physicochemical, and biological properties of soils (Zinke, 1962; Mina, 1967; Karpachevskii, 1981; Seiler and Matzner, 1995; Rhoades, 1997; Dmitriev et al., 1999; Vorobeichik and Pishchulin, 2009, 2011; Ma et al., 2014).

In most cases, the habitat-forming role of trees has been studied in background areas, i.e., those not exposed to impacts from point polluters. Only a few studies have been performed in the vicinities of nonferrous smelters in Finland (Nieminen et al., 1999), Chile (Ginocchio et al., 2004), England (Watmough and Dickinson, 1995), and Spain (Avila and Rodrigo, 2004), as well as in Russia: in the Kola Peninsula (Dem'yanov, 1992; Nikonov and Lukina, 2000; Lukina et al., 2003; Zvereva and Kozlov, 2004, 2007) and the Middle Urals (Dulya, 2006; Vorobeichik and Pishchulin, 2009, 2011).

A common concept is that polluted territories are characterized by high spatial variability of pollutant contents. However, this concept is based either on fragmentary data or on the results of studies performed on several hundred meter to kilometer scales. Strange as it may appear, relevant studies on a microscale (tens of centimeters to few meters) are scarce (Salminen and Haimi, 1999), even though characteristics of the biota at this spatial level are important for understanding the patterns of its functioning in polluted areas (Watmough and Dickinson, 1995; Vorobeichik and Pozolotina, 2003; Gongalsky et al., 2009, 2010). This is why researchers are paying increasing attention to spatial ecology (Ettema and Wardle, 2002). However, a factual finding of increased variability under conditions of pollution is only the first step in the study of this phenomenon, which should be followed by the analysis of mechanisms responsible for it. In the case of forest ecosystems, this means the necessity to evaluate the effect of trees on the spatial distribution of pollutants.

The purpose of this study was to analyze changes in the role of trees in forming the spatial structure of concentration fields of heavy metals (Cu, Pb, Cd, Zn) and hydrogen ions in the forest litter under the impact of strong industrial pollution. The starting hypothesis to be tested was that the effect of trees on the spatial dis-

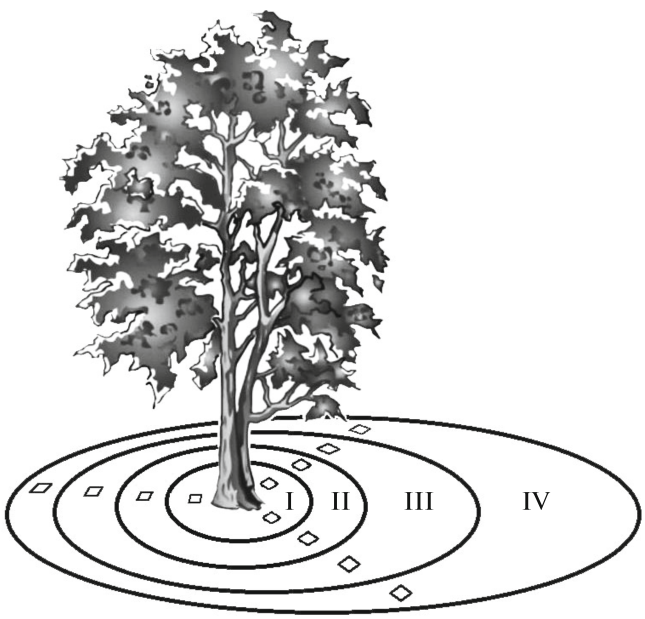

Fig. 1. Scheme of positions of sampling points relative to tree trunk: (I) at tree base, (II) in the middle of crown cover, (III) at the edge of crown cover, (IV) in canopy gap.

tribution of these elements should be attenuated in polluted areas, compared to control (background) areas. This hypothesis followed from the well-documented fact that the growth of trees exposed to pollution is suppressed, which is manifested in decreasing stand density and thinning of tree crowns (Innes, 1993), and, on the other hand, from the existence of positive feedback between the strength of the habitat-forming ability of a tree and the level of development of its crown (Levia and Frost, 2003, 2006; Levia and Germer, 2015). The dependence of the concentrations of the above elements on the distance from tree trunk at different pollution levels is considered in detail in our previous study (performed on the same material) (Vorobeichik and Pishchulin, 2009); here, attention is focused on the spatial pattern of their concentration fields under the forest canopy.

The basic methodological principle of our research is in using a representative sample of specially selected model trees allowing us to analyze, per se, the gradient of intensity of phytogenic field (in A.A. Uranov's terms; see Kryshen', 2000), or ecological field (Walker et al., 1989). Sampling points are distributed at different distances from the model tree trunk so as to represent a series of microbiotopes along a decreasing gradient of phytogenic field intensity: tree-base site → the middle of crown cover → the edge of crown cover → canopy gap (Fig. 1). In each microbiotope, several samples are taken, which are regarded as independent replicates. Such an approach allows the total spatial variance to be split into three components: the variance related to the position of sampling point relative to the tree trunk, the variance due to differences between individual trees, and the residual variance. The first component characterizes the effect of phytogenic field on the distribution of elements; the second, spatial heterogeneity (patchiness) within the test area; and the third, variation unrelated to the above factors (caused by random effects). If our starting hypothesis is true, then the proportion of the first variance component in the polluted area should decrease, compared to that in the control area, while the proportions of the second and third components should increase.

It should be noted that this approach complements but does not negate the traditional scheme of geostatistical analysis that is commonly used to assess spatial variation (Ettema and Wardle, 2002). The point is that it is aimed at analyzing the mechanisms of formation of concentration fields rather than at describing these fields and evaluating their structural parameters.

MATERIAL AND METHODS

The material was collected in 2003 in the territory exposed for a long time to emissions from the Middle Ural Copper Smelter (MUCS) located in the outskirts of Revda, Sverdlovsk oblast. The MUCS (in operation since 1940) has been, until recently, one of the largest sources of airborne pollutants in Russia. The main pollutants are sulfur dioxide and dust containing heavy metals and other toxins (Cu, Pb, Cd, Zn, Fe, Hg, As, etc.). The annual amount of emissions in 1980 reached 226×10^3 t, but it subsequently decreased to 148×10^3 t in 1990, 63×10^3 t in 2000, 34×10^3 t in 2003, and was reduced to no more than 3×10^3 t after 2010. Nevertheless, vegetation recovery and soil purification from heavy metals near the MUCS have not yet been achieved (Vorobeichik et al., 2014; Trubina et al., 2014).

Studies were performed in the area located west of the smelter (opposite to the prevailing wind direction), in two variants of biotopes: spruce–fir forests and secondary birch forests of different plant associations. Their soil cover was represented by combinations of brown, sod-podzolic, and gray forest soils (groups Cambisols, Retisols, and Phaeozems according to *World Reference Base*..., 2014) transformed to different degrees under the impact of technogenic factors (Kaigorodova and Vorobeichik, 1996). With regard to the state of higher vegetation, three zones with different pollution loads were delimited: the impact zone (1 km from the smelter for birch forests and 2 km for spruce–fir forests), the buffer zone (5 and 4 km), and the background zone (20 and 30 km, respectively).

Among various aspects of technogenic degradation of forest ecosystems, attention in this context should be paid to suppression of the tree layer (decrease in timber stock and crown closure and increase in the proportion of dead standing trees); degradation of the herb–dwarf shrub layer (reduction of its species richness and abundance); increase in the coverage of mosses, which in the impact zone are represented by a single species (Vorobeichik and Khantemirova, 1994; Vorobeichik et al., 2014; Trubina et al., 2014); reduction in the rate of organic matter decomposition because of suppressed activity of soil saprotrophs (elimination of earthworms and decrease in the abundance of microfungi) (Vorobeichik, 1991; Vorobeichik and Pishchulin, 2011); and, as a consequence, two- to threefold increase in the depth of forest litter (Vorobeichik, 2003).

Ten model trees were selected in each pollution zone and biotope variant. The main selection criterion was their proximity to a gap in the forest canopy (but not to a large glade or forest edge). In spruce–fir forests, these were Siberian spruce (*Picea obovata* Ledeb.) trees; in birch forests, downy birch (*Betula pubescens* Ehrh.) or silver birch (*B. pendula* Roth.) trees of similar habit (trunk height no less than 15 m, diameter no less than 15 cm in birch and 30 cm in spruce, well-developed crown without any visible damage). The crown projection area in spruce trees averaged 17.9 m^2 in the background zone, 29.6 m^2 in the buffer zone, and 23.0 m^2 in the impact zone; the respective values for birch trees were 26.3, 20.8, and 14.2 m^2 (Vorobeichik and Pishchulin, 2009). The distance between neighboring model trees within the same biotope varied from 15 to 80 m in the background and buffer zone and from 10 to 150 m m in the impact zone. The total area surveyed in the background zone was about 2.5 ha; in the buffer zone, 0.95 ha; in the impact zone, 0.8 ha.

Samples of the forest litter from under the trees were taken within a circular plot with a radius of 4–6 m around the trunk, depending on crown size. Sampling points were arranged along three arbitrary straight lines (not oriented with respect to the cardinal directions) extending from the trunk at angles of no less than 45° (in most cases, 90–120°) relative to each other, with four points on each line (Fig. 1). Point I was near the tree base (0.2–0.3 m from the trunk); point II, in the middle of crown cover (1.2–1.8 m); point III, at the edge of crown cover; and point IV, in the canopy gap. Thus, 12 samples were collected from each model tree (a total of 720 samples from 60 trees).

The samples were cut out with a sharp knife within a 10 × 10-cm frame, all the way through the litter horizon, and packed in plastic bags. In the laboratory, they were dried to air-dry state at room temperature for 30 days, cleared of foreign matter (large branches, cones, green moss, stones, etc.), and ground in a mechanical grinder into small particles (1–2 mm). Each sample was analyzed for the concentrations of acid-soluble Cu, Cd, Pb, and Zn (extracted with 5% HNO_3, 1 : 10 w/v, for 24 h after a single stirring step) and pH of water extract (1 : 25 w/v). Metal concentrations were determined by atomic absorption spectrometry in an AAS 6 Vario instrument (Analitik Jena AG, Germany), pH_{water} was measured ionometrically. Measurements were made in our laboratory accredited in the State System of Analytical Laboratories (certificate no. ROSS.RU0001.515630). To estimate litter stocks and measure pH, samples were weighed with an accuracy of 0.01 g; aliquots for extracting metals (about 2 g) were weighed with an accuracy of 0.0001 g.

Metal stocks in the litter (g/m^2 or mg/m^2) were calculated by multiplying metal concentration (mg/kg) in the sample by litter stock (kg/m^2) at the sampling point. Some samples in the impact zone were devoid of the litter (5 out of 120 samples in spruce–fir forests and 14 samples in birch forests). In such cases, litter stocks were taken to be zero, and data on metal stocks, to be missing.

The components of variance were evaluated by means of two-way ANOVA, with the variant of sample position relative to the tree trunk being the first factor, and the tree (differences between individual trees), the second factor (below, referred to as factors "position" and "tree"). Before analysis, the values of metal stocks and concentrations were transformed logarithmically. We used a random effect model without interaction, but such an approach needs clarification. Formally, evaluation of variance components can only be performed in a random effect model, but consideration of the first of the above two factors as a random-effect factor is not obviously appropriate and should be substantiated (which is not the case with the second factor). We proceeded from the fact that the question of criteria for distinguishing between random- and fixed-effect factors is controversial and has long been discussed by statisticians (Gelman, 2005). It is now accepted that this depends not only on the scheme of experiments but also on the purpose of research: if the total variance is to be estimated, the factor can be justly regarded as random; if the purpose is to evaluate differences between certain gradations, the factor should be regarded as fixed (Bolker et al., 2009). We did not aim at estimating differences in the concentrations of elements between certain variants of sample position relative to the tree trunk, because our interest was focused on variation in their concentrations among all variants. Furthermore, these variants were uniformly distributed along the decreasing gradient of phytogenic field intensity, while the location of sampling point within each variant was random. In other words, the variants were specified not arbitrarily but so as to characterize four levels of field intensity out of a potentially infinite set. Therefore, such an experimental scheme allows the sample variance to be used as an estimate of variance in the general population. Taking into account both these circumstances, it appears correct to regard the variant of sample position relative to the trunk as a random effect and estimate the proportion of variance it explains.

RESULTS

As expected, pollution resulted in a significant increase in litter stock, acidity, and heavy metal contents (Table 1). In spruce–fir forest of the impact zone, the recorded concentrations of metals in the litter exceed the background levels by factors of 52.3 for Cu, 14.6 for Pb, 4.4 for Cd, and 2.8 for Zn; the respective values in the buffer zone are 17.8, 7.7, 2.5, and 1.8. In birch forests, the differences between polluted and background areas are even greater: the concentrations of Cu in the impact and buffer zones exceed the background values by factors of 94.8 and 37.1; of Pb, by factors of 17.6 and 9.3; of Cd, by factors of 6.3 and 9.2; and of Zn, by factors of 2.0 and 3.8, respectively. Moreover, the concentrations of Zn and Cd in the buffer zone proved to be 1.9 and 1.4 times higher, respectively, than in the impact zone.

The range of litter stock and metal contents expands with increase in pollution, and this range in the impact zone is almost the same as that in the entire pollution gradient. It is noteworthy that, in the case of Cd and Zn, the minimum concentrations recorded in the impact zone are even lower than their maximum concentrations in the background zone. The opposite tendency is observed for pH: its range narrows as pollution increases.

The variation coefficients of concentrations and stocks of test elements are mostly within the ranges of 20–40% and 50–80%, respectively. However, their changes with increasing pollution show different tendencies: variation in pH and Pb concentrations remains almost the same; variation in litter stock, Cu concentration, and Cu and Pb stocks decreases; and that in the concentrations and stocks of Cd and Zn increases. An exception from the general trend is the birch forest growing in the impact zone, where variation in all test parameters is markedly lower than in other areas.

In most cases, factor "position" has a significant effect on the test parameters, and the same is true of factor "tree" (Table 2). However, the relative contributions of these factors to the total variance are different. For the litter stock in spruce–fir forests, their ratio is similar in all pollution zones, with the first and second factors explaining more than 50% and only 10–15% of the total variance, respectively (Fig. 2A). In birch forests, the contribution of factor "position" decreases from 60% in the background zone to 20–30% in the buffer and impact zones, while that of factor "tree" increases from 2 to 10–15%.

The ratio of variance components for pH in spruce–fir forests changes unevenly as pollution increases: the proportion of variance explained by factor "position" reaches a maximum (about 50%) in the buffer zone, being significantly lower either in the impact (20%) or in the background zone (10%) (Fig. 2B). The picture observed in birch forests is different: upon transition from the background to the impact zone, the proportion of the first component decreases almost threefold, while that of the second component increases up to 50%.

Taken together, the two factors explain roughly half (30–80%) of the total spatial variance in metal concentrations (Fig. 3A). In spruce–fir forests, the ratio of variance components consistently changes with increasing pollution: the main role in the background zone is played by factor "position," but its role decreases in the buffer zone and is reduced almost to zero in the impact zone. The proportion of the first variance component in the background zone is the highest for Cu (70%) and the lowest for Pb (20%), with the values for Cd and Zn being intermediate (40–50%). Conversely, the proportion of the second component increases upon transition from the background to the impact zone, reaching 50–80%.

In birch forests, the proportion of the first component of variance in metal concentrations in the background zone is generally lower than in spruce–fir forests. As pollution increases, the ratio of these components for particular elements changes in different ways: for Cu and Pb, the role of factor "position" decreases, as in spruce–fir forests, whereas in the case of Cd и Zn there is a kind of inversion between the impact and buffer zones: the contribution of this factor to the total variance is minimum in the buffer zone but significantly increases in the impact zone.

The proportion of the first component of variance in metal stocks, compared to metal concentrations, is higher, reaching almost 80% in the background zone (Fig. 3B). In spruce–fir forests, the contribution of factor "position" decreases upon transition from the background to the impact zone but remains appreciable (about 40%). In birch forests, the proportion of the first variance component is generally lower, as in the case of metal concentrations, and the inversion of the ratio between variance components in the impact versus background zone is observed not only for Cd and Zn but also for Cu and Pb.

DISCUSSION

The spatial pattern of concentration fields of chemical elements in the forest litter is formed as a result of numerous differently directed processes. The content of an element in a given spatial point is a reflection of balance between the input and output components of the element flow. The first component is due to direct surface deposition of aerosols and liquid and solid precipitation with dissolved elements and suspended dust particles; stemflow and throughfall, which are enriched with elements contained in dust particles and leached out of plant tissues; and input with plant debris. The output component involves vertical migration of elements to mineral soil horizons due to leaching, biological decomposition of organic matter, transfer of plant detritus by soil animals, and root absorption, and their horizontal migration due to transfer by water, wind, and animals.

Table 1. Forest litter stocks, pH, and heavy metal contents in different biotopes and pollution zones (accounting unit: sample, $n = 120$)

Parameter	Biotope and zone					
	Spruce–fir forest			Birch forest		
	background	buffer	impact	background	buffer	impact
	Mean ± *SE* (variation coefficient, %)					
Litter stock, kg/m^2	2.2 ± 0.1 (66.7)	5.5 ± 0.3 (60.3)	9.4 ± 0.4 (51.8)	1.2 ± 0.1 (67.7)	3.1 ± 0.1 (33.8)	3.9 ± 0.2 (60.9)
pH*	5.1 ± 0.03 (7.3/82.7)	4.6 ± 0.03 (6.0/83.9)	4.4 ± 0.04 (7.8/79.3)	5.8 ± 0.03 (5.7/100.7)	5.5 ± 0.02 (3.5/46.9)	4.5 ± 0.03 (6.1/62.2)
Concentration, μg/g:						
Cu	82.9 ± 3.5 (45.9)	1472.3 ± 41.7 (31.0)	4335.7 ± 119.0 (29.4)	58.1 ± 2.3 (44.3)	2156.2 ± 51.8 (26.3)	5538.6 ± 160.5 (30.0)
Pb	98.0 ± 2.3 (25.4)	754.1 ± 17.7 (25.8)	1423.9 ± 37.2 (28.0)	110.7 ± 2.4 (23.7)	1026.3 ± 17.9 (19.1)	1960.6 ± 62.0 (32.7)
Cd	3.3 ± 0.1 (30.0)	8.1 ± 0.3 (39.9)	14.4 ± 0.6 (42.9)	3.3 ± 0.1 (28.7)	30.4 ± 0.5 (16.4)	21.3 ± 1.1 (55.7)
Zn	214.9 ± 4.9 (24.9)	387.5 ± 13.2 (37.2)	610.6 ± 20.2 (35.4)	458.3 ± 9.6 (23.1)	1734.2 ± 21.8 (13.8)	931.2 ± 46.7 (51.8)
Metal stocks:						
Cu, g/m^2	0.2 ± 0.02 (106.9)	8.6 ± 0.6 (73.3)	43.0 ± 2.2 (55.6)	0.1 ± 0.01 (129.3)	6.9 ± 0.3 (45.5)	25.4 ± 1.5 (61.1)
Pb, g/m^2	0.2 ± 0.02 (79.9)	4 ± 0.2 (54.6)	13.8 ± 0.6 (49.7)	0.1 ± 0.01 (90.6)	3.2 ± 0.1 (35.3)	8.5 ± 0.4 (48.4)
Cd, mg/m^2	7.9 ± 0.6 (84.0)	47.3 ± 3.4 (79.1)	147.9 ± 9.4 (67.9)	4.3 ± 0.4 (104.2)	93.7 ± 3.0 (35.4)	98.0 ± 7.3 (76.4)
Zn, g/m^2	0.5 ± 0.04 (79.7)	2.3 ± 0.2 (76.3)	6.2 ± 0.4 (62.2)	0.6 ± 0.05 (86.6)	5.4 ± 0.2 (34.5)	4.3 ± 0.3 (74.2)
	Min–Max					
Litter stock, kg/m^2	0.5–8.3	0.7–16.3	0.0–20.9	0.2–4.2	1.0–6.3	0.0–11.2
pH	4.3–6.0	3.8–5.1	3.6–5.0	4.9–6.7	5.1–6.0	4.0–5.3
Concentration, μg/g:						
Cu	29.1–219.9	344.2–2929.3	1960.3–8520.7	29.3–193.4	638.6–3821	2308.7–9080.9
Pb	32.7–194.8	394.4–1377.9	704.2–2480.3	58.8–195.2	517.6–1495.3	465.8–3784.6
Cd	1.7–7.3	2.8–21.1	5.0–31.5	1.5–8.3	16.1–47.1	5.5–66.9
Zn	97.9–365.8	126.0–948.7	274.6–1274.4	167.9–698.6	1059.6–2291.1	294.8–3135.7
Metal stocks:						
Cu, g/m^2	0.02–1.5	0.3–30.3	2.7–101.7	0.01–0.8	0.6–16.1	4.2–78.7
Pb, g/m^2	0.03–1.0	0.3–10.9	1.5–34.1	0.02–0.8	0.5–5.7	1.3–20.4
Cd, mg/m^2	1.1–33.8	1.9–203.3	7.0–507.1	0.5–32.6	16.1–205.3	9.6–309.9
Zn, g/m^2	0.1–2.3	0.1–8.6	0.4–21.7	0.1–2.9	1.1–10.1	0.5–13.8

*Values before and after slash are variation coefficients for pH and hydrogen ion concentration, respectively.

Table 2. Results of two-way ANOVA (Fisher's *F* test) for the effect of factors on the distribution of litter stocks, pH, and heavy metal concentrations

Parameter	Biotope, zone					
	Spruce–fir forest			Birch forest		
	background	buffer	impact	background	buffer	impact
Differences between variants of position relative to tree trunk ($df_1 = 3$)						
Litter stock	66.0***	47.9***	58.5***	55.0***	13.5***	17.6***
pH	6.7***	37.9***	8.7***	15.1***	4.7**	6.9***
Concentration:						
Cu	101.2***	16.4***	2.0 ns	28.7***	3.1*	3.8*
Pb	10.6***	4.8**	0.8 ns	4.6**	11.6***	1.6ns
Cd	35.5***	7.1***	1.5 ns	22.4***	2.4ns	10.1***
Zn	35.0***	8.2***	1.9 ns	17.8***	1.7ns	15.2***
Metal stocks:						
Cu	139.7***	76.8***	34.1***	59.9***	2.6ns	13.2***
Pb	93.0***	31.0***	29.1***	39.9***	1.8ns	9.4***
Cd	126.4***	46.1***	20.4***	66.3***	6.3***	21.1***
Zn	143.2***	49.6***	21.4***	68.2***	6.9***	26.5***
Differences between individual trees ($df_2 = 9$)						
Litter stock	5.4***	3.4***	3.2**	1.7ns	5.3***	4.4***
pH	9.9***	3.1**	5.0***	3.4**	5.2***	13.2***
Concentration:						
Cu	7.1***	5.3***	40.5***	2.2*	6.7***	15.0***
Pb	6.3***	4.4***	11.9***	3.2**	4.3***	2.4*
Cd	4.5***	5.8***	11.6***	1.9ns	4.8***	4.8***
Zn	4.6***	7.5***	10.8***	5.0***	4.5***	7.8***
Metal stocks:						
Cu	8.6***	6.1***	14.2***	1.7ns	5.1***	13.5***
Pb	9.0***	3.9***	7.9***	2.4*	4.5***	5.8***
Cd	6.5***	5.0***	8.2***	2.6*	2.8**	5.9***
Zn	5.2***	5.6***	7.3***	3.9***	3.5***	9.0***

$df_{error} = 107$; ns, $p > 0.05$; * $p < 0.05$, ** $p < 0.01$, ***$p < 0.001$.

The ratio of the input and output components varies with distance from the tree trunk and depends on a number of factors, including the size and pattern of the crown, the density and structure of tree stand, the size and configuration of canopy gaps, meso- and microtopographic features, and weather conditions. Vertical components of the element flow enhance spatial heterogeneity, while its horizontal components reduce it (in the absence of distinct topographic gradients). The increased input of chemical elements in technogenically transformed areas drastically complicate this a priori complex situation, since pollution can directly or indirectly affect all components of the above balance as well as all factors that modify fluxes of chemical elements.

Our previous observations in the background area have revealed a distinct trend that the litter stock and concentrations of hydrogen ions and all metals are the highest near the tree trunk and gradually decrease in the direction to canopy gap (Vorobeichik and Pishchulin, 2009). As a result, a regular horizontal pattern of concentration fields is formed under the effects of individual trees, which consists of concentric zones with decreasing contents of chemical elements. This is why the greater part of spatial variance in test parameters is accounted for by factor "position." Since the study by Zinke (1962), such a regular pattern has been repeatedly described for the distribution of many soil parameters, including the concentrations of macroelements and heavy metals (Wittig, 1986; Falkengren-Grerup, 1989; Skrivan et al., 1995; Rhoades, 1997;

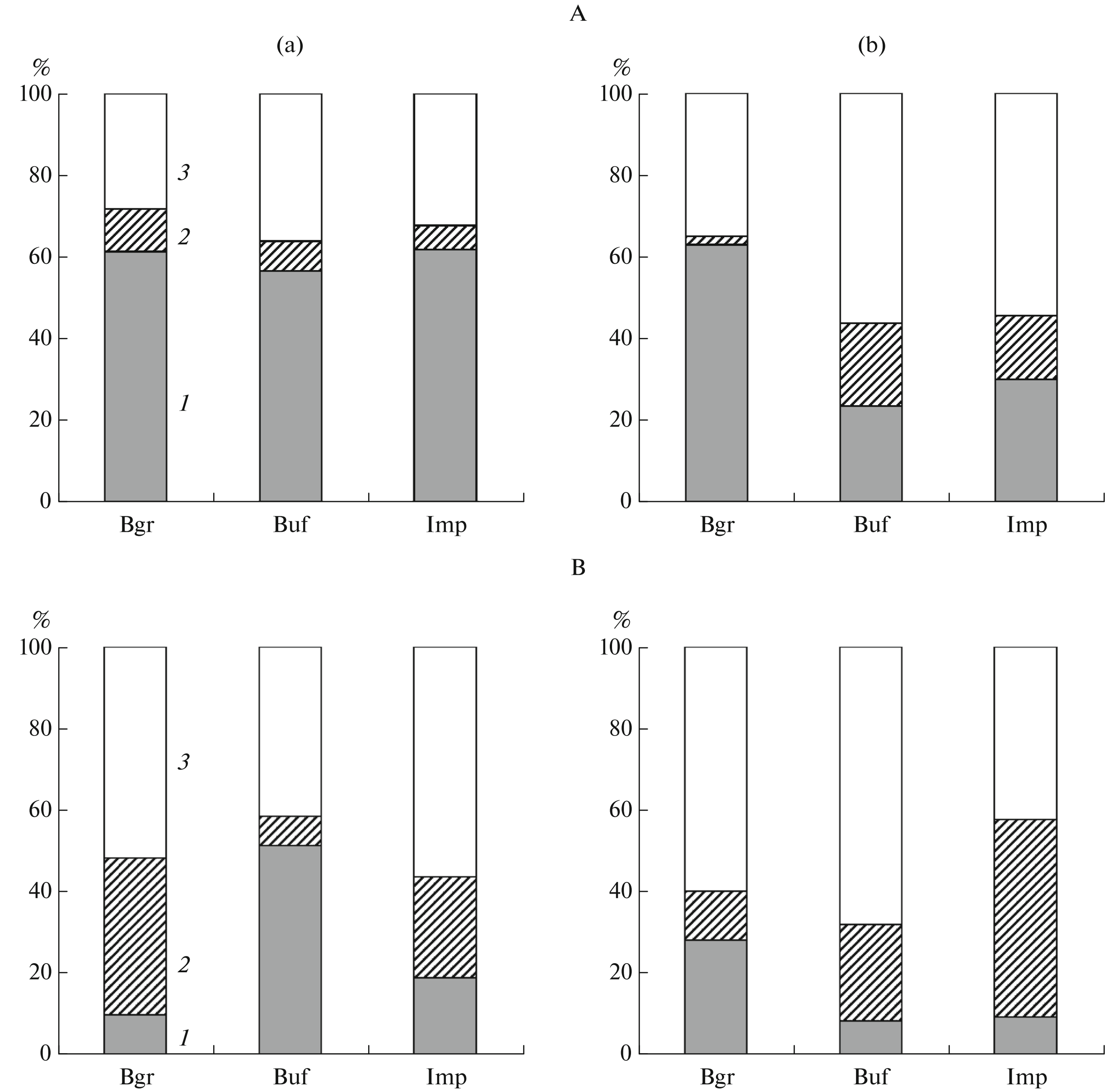

Fig. 2. Components of spatial variance (%) in (A) stock and (B) pH of the litter in (a) spruce–fir and (b) birch forests in background (Bgr), buffer (Buf) and impact (Imp) zones. Here and in Fig. 3, different shading (hatching) indicates variance depending on (*1*) position of sampling point relative to tree trunk, (*2*) differences between individual trees, and (*3*) effect of other factors (residual variance).

Ginocchio et al., 2004; Blagodatskaya et al., 2008). The majority of authors explain this descending concentration gradient by increased input of chemical elements with precipitation and aerosols that are intercepted by tree crowns and then reach the ground as throughfall and stemflow (Bergkvist et al., 1989; Levia and Frost, 2003, 2006; Bialkowski and Buttle, 2015; Levia and Germer, 2015), enrichment of stemflow with elements leached out of foliage and bark (Mina, 1967; Levia and Frost, 2003, 2006), and heterogeneous distribution of litterfall with its prevailing accumulation under tree crowns, at least in conifer forests (Hirabuki, 1991).

Such a consistent decrease in metal concentrations away from the tree trunk is not observed in polluted areas (Vorobeichik and Pishchulin, 2009). Therefore, regularity in the spatial structure of concentration fields (characterized by the first variance component) is weakly expressed or absent, with the leading role being played by higher-order heterogeneity related to the mosaic pollution pattern on the scale of tens to hundreds of meters, which is reflected in the increased proportion of the second variance component. An

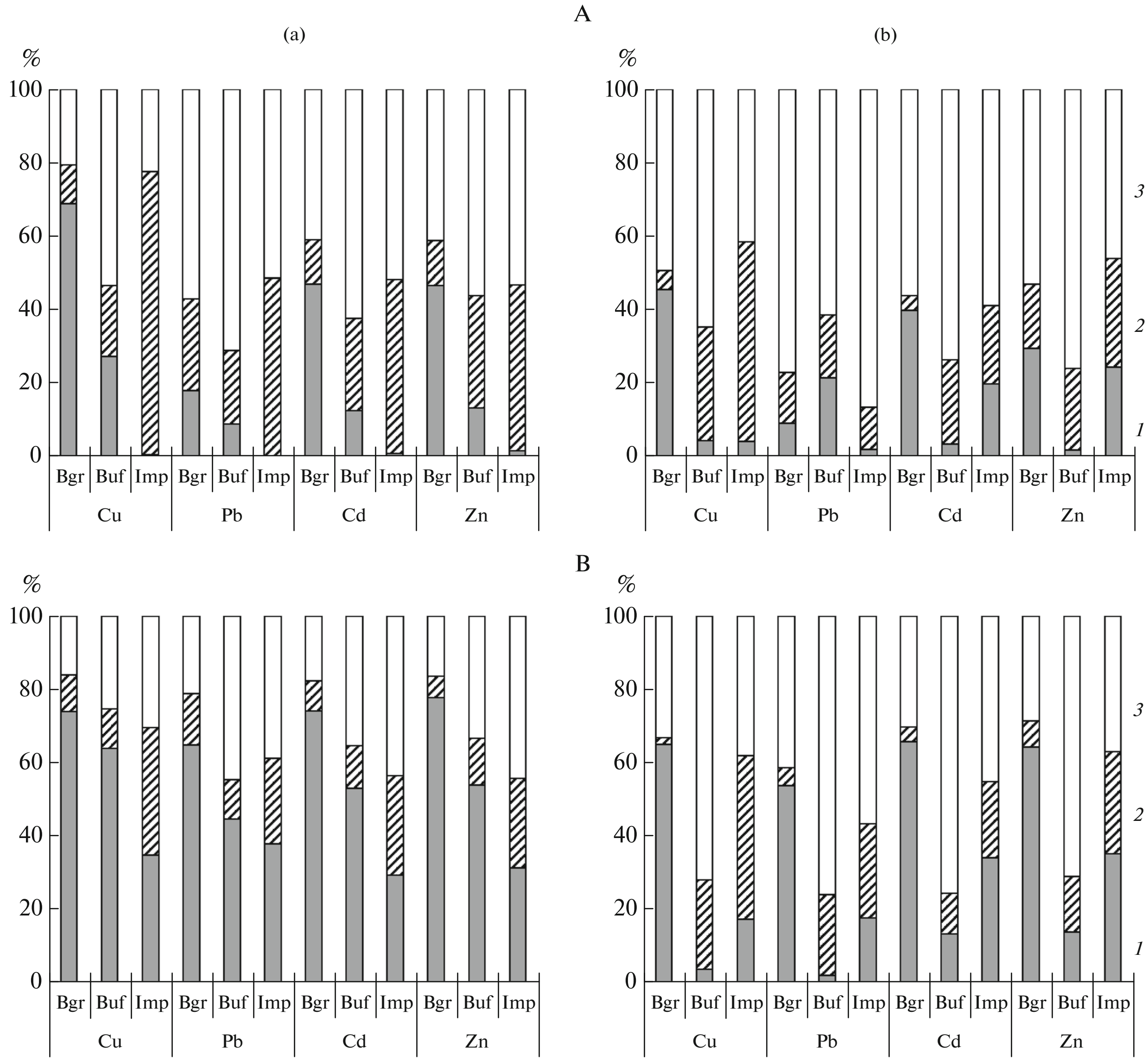

Fig. 3. Components of spatial variance (%) in (A) concentrations and (B) stocks of heavy metals in (a) spruce–fir and (b) birch forests in background (Bgr), buffer (Buf) and impact (Imp) zones.

increase is also observed in the residual variance, which mainly characterizes random variation on the scale of tens of centimeters (rather than the analytical error in concentration measurements, which did not exceed 5–10% in our experiments). All this is evidence in favor of our starting hypothesis that the role of trees in forming the spatial pattern of element concentration fields decreases in polluted areas.

It appears that the primary cause of this phenomenon is the suppression of trees exposed to pollution, with consequent thinning of their crowns. As a result, they less effectively intercept precipitation and aerosols, and the chemical composition of stemflow and throughfall is altered to a lesser extent relative to that of precipitation falling directly on the litter surface. Direct deposition of large dust particles on the litter surface may also have a considerable effect. This process is hardly influenced by the phytogenic field gradient, especially in polluted areas where the shielding role of the herb–dwarf shrub layer is reduced (Dulya, 2006). It is also not excluded that, after being deposited, such particles are redistributed due to wind transfer, which decreases the heterogeneity conditioned by the phytogenic field gradient. In addition, the aforementioned decrease in the role of trees in polluted areas may be explained by the leveling of differences between zones of the phytogenic field in the rate of organic matter decomposition (Vorobeichik and Pishchulin, 2011) and, hence, in biogenic migration of chemical elements.

The decrease in the intensity of tree influence is less pronounced for metal stocks than for metal concentrations, because, in all pollution zones, the stocks of the litter in tree-base sites are markedly greater than in canopy gaps (Vorobeichik and Pishchulin, 2009) due to prevailing accumulation of litterfall under tree crowns (Hirabuki, 1991). Correspondingly, differences in metal stocks between the variants of position relative to the trunk depend primarily on the stock of the litter than on the concentrations of metals. In other words, metal stocks reach a maximum near the trunk not so much because the litter is more strongly polluted as because its amount in this point is greater.

Special attention should be paid to the "strange behavior" of birch forest in the buffer zone that is responsible for the "inversion" between this zone and the impact zone: the effect of the tree decreases upon transition from the background to the buffer zone but increases upon transition from the buffer to the impact zone (Fig. 3). This is observed for only two elements (Cd and Zn) when their concentrations are considered but applies to all test metals when dealing with their stocks. It should be primarily noted that the average concentrations of Cd and Zn, compared to other elements, are higher in the buffer than in the impact zone (see Table 1); therefore, if consideration is given only to the absolute values of concentrations (without regard to pollution zones), then no inversion is observed, and everything fits together: the effect of trees on the distribution of Cd and Zn concentrations regularly decreases with increasing pollution, as in the case of other elements. Such a situation with Cd and Zn may probably be explained by specific features of their transport in the atmosphere: these elements, compared to Cu, are associated with smaller particles (Tsibul'skii and Yatsenko-Khmelevskaya, 2004; Zdanowicz et al., 2006) and are therefore transferred to greater distances from the emission source; hence, the pattern of their distribution in the buffer zone is similar to that of Cu in the impact zone.

It is more difficult to explain the inversion with respect to stocks, which is observed for all test metals. The main role in this case is probably played by unusually low variation of litter stock in the birch forest of the buffer zone (see Table 1), where the increased accumulation of the litter in the tree-base site is less manifested than in other zones (Vorobeichik and Pishchulin, 2009). This, in turn, can be explained by more active horizontal transfer of the litter, which is facilitated due to the suppression of herb–dwarf shrub layer and almost complete absence of moss layer in this biotope, unlike in the birch forest of the impact zone where the moss layer is well developed (Dulya, 2006; Vorobeichik and Pishchulin, 2009).

Several methodological aspects of the study should also be discussed. Firstly, the formation of concentration fields is a complex process involving a number of divergent components, but we have analyzed it from only one aspect—the effect of individual trees—without considering many other factors. Secondly, our attention was focused on the forest litter as the horizon characterized by the maximum deposition of technogenic pollutants, while the pattern of formation of concentration fields in underlying mineral horizons may be different. Thirdly, it must be kept in mind that the models of concentric zones of influence of individual trees (Zinke, 1962) and soil tessera (Karpachevskii, 1981) significantly simplify the actual situation (Dmitriev et al., 1999). The overlap of tree crowns in the forest may blur the influence of individual trees, and it is therefore necessary to discriminate between the habitat-forming effects of individual trees and tree stand as a whole. Fourthly, we emphasize that our conclusions are based on analysis of specially selected model trees with well-developed crowns, while the occurrence of such trees in polluted areas is likely an exception rather than the rule. Nevertheless, even these trees lose their significance as leading determinants of the pattern of concentration fields. It is logical to assume that the influence of smaller and/or suppressed individuals forming the bulk of tree stand in the impact zone is even less prominent.

Knowledge of trends in the formation of the pattern of concentration fields under industrial pollution is important from several aspects. In particular, it is prerequisite for (1) modeling biogeochemical fluxes of elements in forest landscapes; (2) developing correct sampling schemes necessary for unbiased estimation of pollution levels in a given area, and (3) gaining an insight into mechanisms providing for the survival of biota under heavy toxic loads. Laboratory experiments and observations in the field have shown that plants can survive in microsites where the level of toxicity is relatively low (Watmough and Dickinson, 1995; Ginocchio et al., 2004), and soil animals can avoid microsites with increased concentrations of toxicants (Gongalsky et al., 2009, 2010). In turn, spatial heterogeneity of the environment in polluted areas accounts for high beta-diversity of the biota (Trubina and Vorobeichik, 2012) and its relatively rapid recovery after reduction of technogenic load (Vorobeichik and Nesterkova, 2015).

A detailed analysis of changes in the pattern of spatial variation in the contents of test elements was beyond the scope of this study. Nevertheless, it can be stated that a rise in pollution level may lead not only to the widening of variation range but also to its narrowing, with the variation coefficient increasing or decreasing. Therefore, the accepted opinion that spatial variation in the contents of metals increases in polluted areas should be updated, at least on the microscale.

CONCLUSIONS

In the background area, more than half of spatial variance in the concentrations and stocks of test elements is explained by position relative to the trunk. As the level of pollution increases, the proportion of this variance components decreases, while differences between individual trees become greater. Thus, our starting hypothesis is true: the role of trees in forming a regular pattern of concentration fields (characteristic of undisturbed forests) is weakly expressed in polluted areas. Therefore, trees exposed to heavy industrial pollution become ineffective as ecosystem engineers, at least with respect to fluxes of chemical elements. This may be interpreted as evidence for the replacement of the main determinant of then pattern of concentration fields, with biotic regulation yielding "the palm of victory" to abiotic regulation.

The results of this study have methodological significance. To correctly estimate the degree of soil pollution in areas with a background level of pollutant fallout, it is important to take into account the positions of sampling points relative to the tree trunk, paying less attention to differences between individual trees. In polluted areas, conversely, attention should be focused primarily on the mosaic pattern of pollution, while the relative positions of sampling points may almost be disregarded.

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