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Article

Study of vertical migration of heavy metals in soil contaminated with petroleum products

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Abstract. This study investigates the effects of soil contamination by used motor oil on the migration and accumulation properties of heavy metals under field experiment conditions. Radial contrast coefficients are calculated for zinc, copper, lead, nickel, and cobalt based on two years of monitoring during the 4 month period of observations (June–September 2019–2020). It is shown that soil contamination with petroleum products leads to active accumulation of zinc in the surface soil layer (0–10 cm). The migration capacity of copper increases proportionally to the increase in the amount of waste oil entering the soil. Petroleum products have practically no effect on the accumulation and mobility of Co, Pb and Ni.

Keywords: radial contrast coefficient, petroleum hydrocarbons, zinc, copper, accumulation, pollutants, monitoring

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Научная статья

Изучение вертикальной миграции тяжелых металлов в загрязненной нефтепродуктами почве

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Аннотация. Исследовано влияние загрязнения почвы отработанным машинным маслом на миграционные и аккумуляционные свойства тяжелых металлов в условиях полевого опыта. Рассчитаны коэффициенты радиальной контрастности для цинка, меди, свинца, никеля и кобальта на основе двухлетнего мониторинга в течение 4 месяцев наблюдений (июнь–сентябрь 2019–2020 гг). Показано, что загрязнение почвы нефтепродуктами приводит к активному накоплению цинка в поверхностном слое почвы (0–10 см). Миграционная способность меди растет пропорционально увеличению дозы вносимого в почву отработанного масла. На накопление и подвижность Co, Pb и Ni нефтепродукты практически не влияют.

Ключевые слова: коэффициент радиальной контрастности, нефтяные углеводороды, цинк, медь, аккумуляция, поллютанты, мониторинг

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Introduction

The monitoring of soil contamination by oil and refined petroleum products in Russia is an important issue which is under constant regulatory oversight. According to the State Report of the Ministry of Natural Resources of Russia for 2021 (O sostoyanii..., 2022), a study of the territories of Western Siberia, the Republics of Tatarstan, Udmurtia and Chuvashia, as well as Irkutsk, Nizhny Novgorod, Orenburg and Samara oblasts showed a high level of soil contamination with petroleum products (PP). Thus, the PP concentrations in soils within the city of Kazan exceeded the background values (B) by 3–5 times; in Samara, it was found that the average concentration of PPs in soils was 940.7 mg/kg, which is 18.8B, with maximum concentrations reaching 47B; for soils in Nizhny Novgorod, the corresponding values were 452 mg/kg and 2202 mg/kg (22B). The results of a soil study in the city of Glazov in the Udmurt Republic indicate dangerous levels of pollution: the average concentration of PPs in soils was found to be 1351 mg/kg (48B), and the maximum – 345B (9727 mg/kg).

Oil products entering the soil lead to changes in its physical, chemical and microbiological properties, resulting in a decline or loss of its fertility. There is a risk of pollutants accumulation by plants, which may eventually have a negative impact on human health and other living organisms (Fu et al., 2018; Tiwari et al., 2023). No less dangerous is the change in migration activity of heavy metals (HM) under conditions of anthropogenic pollution. Patterns of HM migration into plants on soil substrates containing wastes from copper smelters (Kotelnikova et al., 2022) as well as HM mobility in soil and their bioavailability for plants in the impact zone of mining and processing enterprises (Lipina and Alexandrova, 2013) have been studied. Research has also been done on the modeling of HM migration under conditions of technogenic pollution based on the results of long-term monitoring (Timofeev et al., 2021), using the machine learning method (Wang et al., 2023), as well as in laboratory experiments with soil columns (Fried and Borisochkina, 2020b; Gruzdkov et al., 2009).

Although there are data on the radial and spatial distribution of petroleum pollutants in hydrocarbon production areas (Pan et al., 2021; Zamotayev et al., 2015), the issue of HM migration in oil-contaminated soils is currently insufficiently studied.

In this regard, the aim of this paper is to evaluate the results of a two-year monitoring of the radial migration of HMs in soil contaminated with petroleum products during the four-month periods of observation (from June to September of 2019–2020).

Materials and methods

In 2019, a small-plot field experiment (a plot area of 1 m^2) was conducted in Vyaznikovsky raion of Vladimir oblast to simulate an unauthorized spill of petroleum products and assess the impact of this pollution on the ecological state of soil. An area of the land withdrawn from agricultural turnover in 1992 and partially overgrown with tree and shrub vegetation was selected for this experiment. Waste oil of the M-8DM brand for automobile and tractor diesel engines, manufactured according to GOST 8581-78[1] was used as a pollutant. Three parallel experiments were conducted on two plots: PPs were spilled on the surface of the plots at concentrations of 10, 20 and 30 g/kg of soil, respectively (distances between the experimental plots were 3–4 m). The depth of the pollutant penetration did not exceed 10 cm of the topsoil. At a distance of at least 5 m from the experimental sites, two control uncontaminated plots were marked out (Fig. 1).

The soil type in the study area is sod-podzolic sandy loam underlain by heavy loams, the physical clay content (< 0.01 mm) is 10–20% (Komarov et al., 2019), the mass fraction of humus at a depth of 0–10 cm is 1.37–1.83%, at a depth of 10–20 cm – 0.8–1.1%. A soil transect was laid out to a depth of 0.6 m to study the soil profile. The obtained parameters are presented in Table 1.

Soil samples were taken at a depth of 0–10 cm and 10–20 cm using the envelope method in accordance with GOST 17.4.3.01-2017[2]. Samples were collected in the middle of each of the four months (June–September) for two years (2019–2020). In the middle of September of each year, an additional sample was taken at a depth of 20–30 cm. A total of 144 soil samples were analyzed.

Hydrogen index of soil salt extract was determined according to GOST 26483-85[3].

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[1] GOST 8581-78. Motor oils for automobile and tractor diesel engines. Technical conditions.

[2] GOST 17.4.3.01-2017. Nature protection. Soils. General requirements for sampling.

[3] GOST 26483-85. Soils. Preparation of salt extract and determination of its pH by Central Institute of Nitrogen and Organic Fertilizers method.

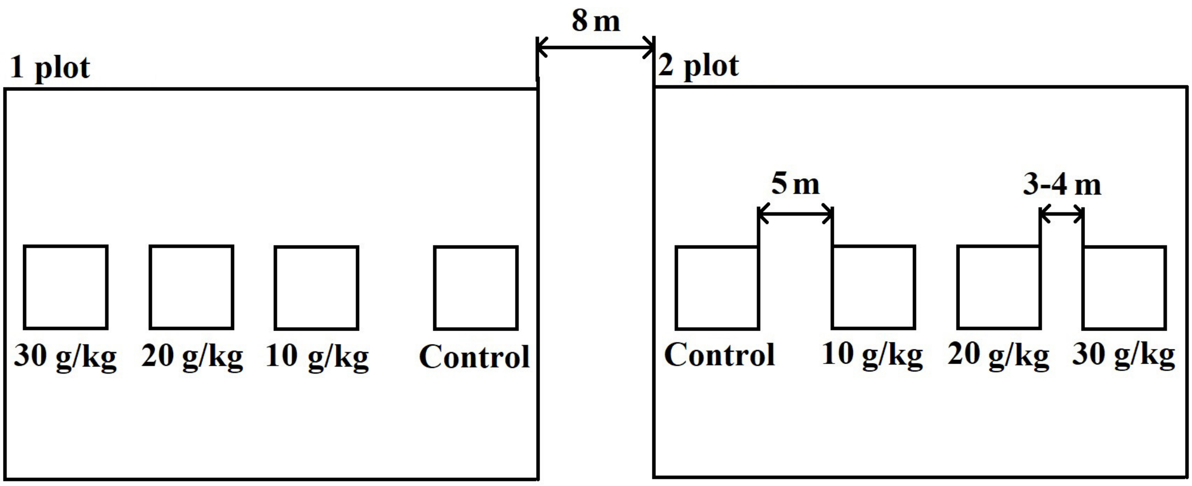

Fig. 1. Scheme of the experiment.

Table 1. Soil parameters of the experimental site.

Genetic soil horizon	Granulometric composition	Distance between upper and lower boundaries of the horizon / power of the genetic horizon, cm	pH_{KCl}
O – litter	Organic plant remains	0–2/2	5.59
AO – coarse humus	Sandy loam	2–6/4	5.59
A – humus	Sandy loam	6–12/6	5.59
EL – eluvial	Sandy loam	12–40/28	5.47
ELBt – transition	Sandy loam	40–60/20	5.07
Bt – textured	Heavy loam	60+...	4.73

The hydrocarbon composition of waste oil was determined by infrared spectrometry on an IR Fourier spectrometer "InfraLUM FT-08" according to PU 03-2002 "Instructions for the identification of petroleum products using IR Fourier spectrometer "InfraLUM FT-02". Each sample of used oil, previously dehydrated using calcined sodium sulfate was placed in a one-piece cuvette (0.05 mm) with an injection syringe. Then, the IR absorption spectrum of the oil product was recorded using the spectrometer (Fig. 2).

The percentage composition of alkane, naphthenic, aromatic, and oxidized structures in the waste oil was calculated based on the optical densities of their characteristic absorption bands (Trifonova and Kurbatov, 2023).The results are shown in Table 2.

Weather conditions during the study period varied: the amount of precipitation in 2019 was significantly higher than the long-term average, but the thermal regime was quite close to the mean annual (Table 3). In 2020, the weather was different: due to the warmer winter, the average air temperature was slightly higher than in 2019, and the amount of precipitation was comparable to the annual average.

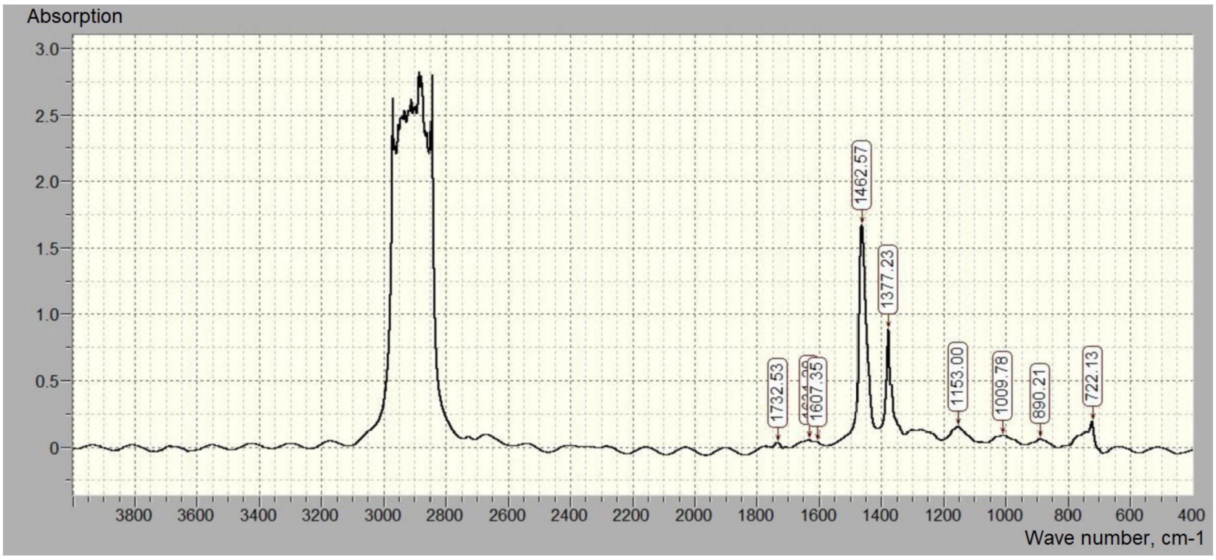

Fig. 2. The IR spectrum of waste oil with the main characteristic absorption bands.

Heavy metal concentrations were determined using a «Spectroscan MAX-G» wavelength dispersive X-ray fluorescence spectrometer according to the method M-049-PDO/18 (FR.1.31.2018.32143)[4] in two replicates for each sample. The sample preparation procedure involved grinding the soil in an agate mortar to a particle size ≤ 63 μm, followed by sifting through a sieve. A laboratory hydraulic press was used to form a boric acid substrate into which the prepared soil sample was pressed; the resulting "tablet" was then chemically analyzed.

To characterize the accumulation and migration of HMs in soil horizons, the radial contrast coefficient R was calculated using the following formula (Mikhalchuk, 2017)::

$$R = C_n / C_c,$$

where C_n is the element concentration in the horizon n, C_c is the weighted average element concentration in the set of soil horizons.

R coefficient values > 1 indicate the element accumulation in the soil horizon under consideration and the presence of geochemical barriers (Table 4), whereas R values < 1 indicate the element mobility (Shabanov and Marichev, 2020).

Statistical data processing was performed in the program "STATISTICA 10". To test the significance of the differences between samples, the nonparametric Mann–Whitney U–test was used at a significance level of $p < 0.05$.

Results and discussion

The results of two-year monitoring of the gross content of HMs in soil samples are presented in Table 5. Soil contamination with petroleum products has little effect on the content of such heavy metals as nickel, lead and cobalt. The concentrations of these HMs were close to control values during all months of the two-year study period, regardless of the applied dose of pollutants. No significant deviations were observed.

Sampling depth of 0–10 cm

The following patterns are typical for depths between 0 and 10 cm: low doses of contamination (10 g PP/kg) significantly increase zinc concentrations in the soil. In August 2019 and July 2020, zinc concentrations up to 2.3B and 1.9B were recorded respectively. Copper exhibits significant mobility both accumulation up to 1.6 times the background value and migration from the upper soil horizon to nearly half the control value were observed in July 2020.

[4] M-049-PDO/18. Methodology for measuring the mass fraction of metals and metal oxides in powder samples of soils and bottom sediments by X-ray fluorescence method.

Table 2. Hydrocarbon composition of waste oils of the M-8DM brand.

Contents of structural groups, %			
Arenes	Alkanes	Naphthenes	Oxidized
6.73	87.36	4.73	1.18

Table 3. Average monthly and average annual air temperatures and precipitation in 2019–2020 in the territory of Vyaznikovsky raion of Vladimir oblast[5]. The values of monthly average temperature and precipitation during the observation period (June–September 2019–2020) are highlighted in bold.

Year	January	February	March	April	May	June	July	August	September	October	November	December	Per year
Average monthly and annual air temperatures													
2019	−9.0	−3.6	−0.9	5.8	14.9	**17.6**	**15.7**	**14.6**	**10.2**	7.2	−0.9	−1.7	5.8
2020	−1.7	−2.4	2.6	3.5	11.5	**16.5**	**19.0**	**15.7**	**12.1**	6.8	0.1	-8.3	6.3
Average annual rate	−8.3	−7.7	−2.3	5.8	13.0	16.6	19.0	16.9	11.4	4.9	−1.9	−6.1	5.1
Monthly and annual precipitation amounts													
2019	56	49	27	14	42	**121**	**71**	**97**	**65**	102	25	47	715
2020	47	40	34	50	87	**37**	**82**	**51**	**98**	29	28	19	603
Average annual rate	40.4	32.6	29.3	35.8	46.2	71.1	65.0	54.4	49.7	54.6	54.3	38.6	572

Table 4. Gradations of geochemical barrier contrast by the coefficient value.

Barrier characteristics	Radial contrast ratio
Low contrast barrier	R = 1.3–1.5
Contrast barrier	R = 1.5–3.0
High contrast barrier	R > 3.0

[5] Weather and climate. Web page. URL: http://www.pogodaiklimat.ru/history/27543.htm (accessed: 06.02.2023).

A PP dose of 20 g/kg leads to a significant increase in zinc concentration: almost 2-fold in July 2019 and 2.2-fold in September 2020. Copper migration intensifies – its concentration at a depth of 0–10 cm was decreasing and amounted to 0.2–0.9 of the background level over the entire observation period.

A higher PP dose (30 g/kg) has the greatest effect on the accumulation and migration of the metals. Thus, zinc concentrations increased significantly compared to the control: up to 2.1 times in August 2019 and 2.9B in September 2020. Under such contamination conditions, copper is practically not retained in the topsoil: in August and September of both years, its concentration values were not reliably measurable.

Sampling depth of 10–20 cm

In 2019, zinc concentrations recorded at a depth of 10–20 cm were practically the same as those at the control sites, but in 2020, they slight exceeded the control – up to 1.3 times in the case of PP contamination at a dose of 30 g/kg. There were no significant changes at the other sampling sites. Copper is more mobile – its concentrations varied from half the background level to 1.8B (2019). In 2020, soils at the sampling sites with low (10 g/kg) and moderate (20 g/kg) levels of pollution were also characterized by high copper mobility, with a decrease in its concentration up to 50% of the background level. However, in the case of a 30 g/kg dose, high rates of Cu leaching from 10-20 cm depths to lower soil layers were observed: compared to the background level, the metal concentration decreased by 36–71%.

Sampling depth of 20–30 cm

Soil samples collected in September 2019–2020 showed that the content of zinc at a depth of 20–30 cm was stable. The concentration of this metal did not differ from the control values at any level of pollution. However, there was a decrease in copper concentration in this soil horizon up to 50% of the background values for the same period.

It is well known that the gross zinc content in soil is most often directly proportional to the thickness of the humus horizon, which is due to high biological significance of this metal and its tendency towards bioaccumulation (Lesnykh, 2005). Consequently, when soil is contaminated with petroleum products, it creates favorable conditions for the active assimilation and accumulation of Zn by soil microbial communities. This phenomenon may be associated mainly with the intensification of the vital activity of hydrocarbon-oxidizing microorganisms, as evidenced by other researchers (Fu et al., 2018; Geng et al., 2022; Koolivand et al., 2022). In earlier studies carried out by the authors in the framework of this experiment, the enzyme activity of soils contaminated with petroleum products was investigated (Kurbatov and Skripchenko, 2020). It turned out that as the dosage of waste oil increased, the urease activity of soils also increased, indicating an enhanced activity of microorganisms. The waste oil used in this study contains 87.36% alkanes (Table 2), which are most easily degraded by bacteria. The pathways of these substances oxidation by microorganisms has been studied pretty well (Timergazina and Perekhodova, 2012). Under the conditions of the present experiment, zinc appears to be less susceptible to migration processes in the presence of increased precipitation and temperature (Tables 3, 5). Apparently, the presence of petroleum products in the soil binds Zn compounds in the humus horizon even more firmly, as evidenced by its consistently low concentrations at depths of 10–20 cm.

With regard to the gross concentration of copper, an inverse relationship is observed: this metal has low rates of biological absorption and practically does not accumulate biogenically, which results in leaching and active migration of copper into the underlying soil layers depleted of organic matter (Lesnykh, 2005). Additionally, favorable weather conditions, such as increased temperature and precipitation, may contribute to the intensification of migration or immobilization of the metals under study. Similar conclusions have already been made previously (Fried and Borisochkina, 2020a); in this work, the migration activity of copper was predominantly influenced equally by the pair of factors "carbonates–precipitation" and "carbonates–temperature". As sown in Table 3, an increase in air temperature and precipitation intensifies the process of copper migration from the humus layer of the soil, primarily to a depth of 10–20 cm. Soil contamination with waste oil only reinforces this trend.

To characterize the vertical migration of HMs in soils contaminated with petroleum products, we calculated their radial contrast coefficients at depths of 0–10, 10–20, and 20–30 cm (Table 6).

Nickel, lead and cobalt are characterized by a uniform distribution in the soil layers under consideration; their ratio and quantity do not depend on the doses of petroleum products.

Radial contrast with respect to zinc and copper is observed in the soils under study, and the value of this coefficient increases with an increase in the dose of PPs. In the control soil samples, no differences

Table 5. Gross content of heavy metals in soil at sampling depths of 0–10, 10–20 and 20–30 cm. LLOQ – lower limit of quantification.

Month	Sampling depth, cm	Gross content of the element, mg/kg									
		Zn		Cu		Ni		Pb		Co	
		2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
Control (0 g/kg)											
June	0–10	63.73 ± 1.87	60.41 ± 1.23	49.82 ± 8.43	27.79 ± 3.49	30.77 ± 2.15	28.99 ± 1.10	18.29 ± 2.04	15.15 ± 2.52	11.35 ± 0.40	11.74 ± 0.26
	10–20	68.06 ± 1.05	62.15 ± 1.71	66.91 ± 4.62	27.03 ± 4.35	32.72 ± 0.85	31.21 ± 2.11	16.93 ± 1.24	16.20 ± 1.75	12.32 ± 0.55	12.33 ± 0.28
July	0–10	62.63 ± 2.29	57.18 ± 4.12	51.56 ± 13.40	18.35 ± 1.10	30.90 ± 1.49	25.93 ± 2.16	28.58 ± 2.58	20.74 ± 1.80	11.81 ± 0.72	10.03 ± 0.82
	10–20	61.84 ± 1.91	62.40 ± 1.35	52.63 ± 6.94	46.68 ± 5.06	30.46 ± 1.29	30.74 ± 0.57	19.84 ± 2.80	17.90 ± 1.44	11.19 ± 0.77	10.74 ± 0.44
August	0–10	54.85 ± 1.39	58.20 ± 0.66	37.57 ± 10.07	21.27 ± 7.40	27.12 ± 0.61	29.73 ± 2.28	19.59 ± 1.94	13.82 ± 3.76	10.55 ± 0.76	10.35 ± 0.49
	10–20	58.48 ± 3.21	58.60 ± 1.30	68.56 ± 7.01	48.99 ± 2.73	30.05 ± 1.67	31.35 ± 2.06	18.54 ± 3.29	14.80 ± 2.48	10.61 ± 0.58	11.41 ± 0.39
September	0–10	67.91 ± 1.39	61.92 ± 3.55	59.24 ± 4.41	33.81 ± 7.56	33.10 ± 1.02	32.99 ± 3.03	22.27 ± 0.90	17.95 ± 3.09	11.53 ± 0.81	11.60 ± 0.63
	10–20	60.58 ± 1.84	58.85 ± 0.70	38.74 ± 5.03	24.85 ± 1.69	31.32 ± 0.26	29.80 ± 1.00	17.57 ± 2.46	20.45 ± 2.82	11.93 ± 0.35	11.41 ± 0.54
	20–30	62.78 ± 0.40	61.45 ± 1.73	30.86 ± 5.80	40.28 ± 5.44	32.04 ± 0.25	34.98 ± 1.86	18.65 ± 1.17	14.51 ± 1.03	11.04 ± 0.41	11.41 ± 0.77
10 g/kg											
June	0–10	107.48 ± 14.54	72.80 ± 5.22	49.88 ± 3.26	29.20 ± 1.67	31.02 ± 1.52	32.07 ± 1.73	15.84 ± 3.32	13.35 ± 1.30	10.69 ± 0.52	12.06 ± 0.53
	10–20	75.57 ± 0.76	64.50 ± 0.98	73.33 ± 4.98	32.15 ± 1.46	35.90 ± 0.95	31.32 ± 1.58	22.44 ± 3.17	19.03 ± 2.30	12.56 ± 0.59	12.52 ± 0.19
July	0–10	71.34 ± 3.55	106.97 ± 1.74	39.53 ± 9.42	28.69 ± 7.31	31.32 ± 1.80	31.41 ± 1.11	22.96 ± 4.10	19.94 ± 1.21	11.44 ± 0.49	12.00 ± 0.53
	10–20	59.61 ± 3.31	62.69 ± 3.30	51.42 ± 14.62	41.68 ± 2.82	31.42 ± 2.23	31.98 ± 1.06	13.64 ± 2.31	11.16 ± 2.75	11.47 ± 0.69	12.68 ± 0.54
August	0–10	123.42 ± 2.64	101.70 ± 11.44	42.05 ± 11.35	13.83 ± 2.87	29.76 ± 2.85	32.22 ± 0.88	19.27 ± 1.05	15.56 ± 0.76	10.09 ± 0.25	10.94 ± 0.39
	10–20	59.83 ± 3.45	63.18 ± 0.68	41.63 ± 14.51	27.56 ± 2.43	31.98 ± 3.07	32.42 ± 1.01	17.86 ± 2.07	12.87 ± 4.02	11.21 ± 0.77	12.48 ± 1.15
September	0–10	66.66 ± 0.18	95.40 ± 10.05	34.80 ± 0.79	17.83 ± 5.39	32.02 ± 0.20	32.83 ± 0.89	18.00 ± 1.90	21.32 ± 1.75	11.88 ± 1.66	10.75 ± 0.40
	10–20	64.48 ± 2.69	71.16 ± 1.71	53.98 ± 2.68	22.04 ± 4.54	35.60 ± 1.11	32.26 ± 1.30	22.36 ± 4.39	17.15 ± 1.61	12.48 ± 0.57	11.87 ± 1.09
	20–30	65.33 ± 1.36	63.58 ± 2.09	23.87 ± 2.91	31.49 ± 3.44	33.51 ± 1.45	34.50 ± 2.21	21.96 ± 2.83	22.37 ± 2.14	11.81 ± 0.61	11.76 ± 0.22

Month	Sampling depth, cm	Gross content of the element, mg/kg									
		Zn		Cu		Ni		Pb		Co	
		2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
20 g/kg											
June	0–10	89.65 ± 9.95	80.36 ± 2.15	17.58 ± 5.78	12.75 ± 2.31	28.91 ± 2.01	30.50 ± 1.46	16.11 ± 0.99	17.43 ± 1.39	9.36 ± 0.43	10.86 ± 0.37
	10–20	72.32 ± 8.06	63.73 ± 1.03	59.31 ± 5.65	29.35 ± 3.12	33.90 ± 1.31	34.88 ± 0.69	19.09 ± 1.93	17.80 ± 1.80	11.52 ± 0.64	11.62 ± 0.47
July	0–10	117.53 ± 22.55	88.91 ± 5.08	26.58 ± 8.22	16.01 ± 2.39	30.16 ± 2.02	32.13 ± 0.87	21.84 ± 2.90	16.46 ± 2.33	10.96 ± 0.45	10.44 ± 0.77
	10–20	59.31 ± 3.47	67.68 ± 3.99	44.34 ± 10.85	37.71 ± 3.56	30.86 ± 2.34	34.04 ± 2.58	20.56 ± 2.39	18.52 ± 2.67	11.12 ± 0.50	12.18 ± 0.71
August	0–10	65.93 ± 2.13	115.16 ± 15.95	14.99 ± 2.32	< LLQQ	27.07 ± 0.20	29.40 ± 1.31	20.60 ± 0.56	18.51 ± 1.86	10.58 ± 0.17	10.14 ± 0.35
	10–20	67.33 ± 5.56	65.03 ± 1.29	51.84 ± 10.01	27.54 ± 3.29	33.86 ± 0.81	31.82 ± 1.68	17.67 ± 3.04	16.32 ± 1.57	11.35 ± 0.59	10.38 ± 0.92
September	0–10	87.76 ± 6.60	132.88 ± 18.51	37.05 ± 6.31	28.16 ± 6.80	36.09 ± 2.96	36.81 ± 2.44	20.04 ± 1.51	24.37 ± 2.76	11.86 ± 0.54	10.72 ± 0.50
	10–20	59.40 ± 6.07	68.02 ± 1.69	67.95 ± 16.71	21.79 ± 6.05	35.54 ± 3.15	32.70 ± 2.38	18.68 ± 1.57	16.57 ± 1.85	11.67 ± 0.70	11.72 ± 0.73
	20–30	64.19 ± 3.53	61.99 ± 2.06	18.31 ± 0.57	30.83 ± 1.98	31.61 ± 0.19	34.03 ± 0.65	18.85 ± 0.99	18.89 ± 0.95	9.66 ± 0.20	11.21 ± 0.39
30 g/kg											
June	0–10	119.68 ± 9.32	139.73 ± 1.68	33.21 ± 5.09	< LLQQ	27.91 ± 1.60	28.56 ± 1.29	20.19 ± 2.33	15.49 ± 3.56	10.94 ± 0.36	10.33 ± 0.28
	10–20	75.24 ± 3.21	73.17 ± 8.38	74.19 ± 3.02	< LLQQ	35.57 ± 1.36	30.58 ± 1.44	22.13 ± 1.59	17.90 ± 2.28	13.02 ± 0.60	10.72 ± 0.53
July	0–10	88.16 ± 4.37	158.67 ± 3.49	20.12 ± 5.57	17.35 ± 6.58	28.91 ± 2.04	28.79 ± 2.00	21.72 ± 0.88	15.47 ± 2.01	10.79 ± 0.77	11.60 ± 0.73
	10–20	64.29 ± 5.70	75.95 ± 8.53	51.60 ± 11.41	17.80 ± 4.52	33.43 ± 1.07	30.60 ± 1.06	25.12 ± 0.31	13.88 ± 2.95	12.11 ± 0.49	11.46 ± 0.53
August	0–10	113.73 ± 1.71	158.50 ± 3.15	< LLQQ	< LLQQ	29.37 ± 0.04	31.52 ± 2.23	21.03 ± 0.83	19.23 ± 1.45	12.17 ± 0.35	9.63 ± 0.28
	10–20	60.50 ± 10.65	75.48 ± 5.86	25.88 ± 9.06	30.91 ± 6.42	29.34 ± 2.82	30.76 ± 1.73	16.33 ± 1.73	17.50 ± 2.98	9.57 ± 0.68	10.99 ± 0.83
September	0–10	135.04 ± 0.42	177.21 ± 6.62	< LLQQ	< LLQQ	29.94 ± 0.92	32.07 ± 1.29	17.89 ± 0.22	16.30 ± 1.83	11.04 ± 0.44	11.12 ± 0.55
	10–20	70.49 ± 5.87	68.64 ± 4.50	59.06 ± 3.73	14.19 ± 2.10	36.27 ± 1.03	30.70 ± 1.09	17.98 ± 1.82	21.05 ± 1.31	12.78 ± 0.25	11.13 ± 0.90
	20–30	66.55 ± 1.94	62.17 ± 1.31	20.89 ± 1.95	20.58 ± 3.74	33.12 ± 0.15	30.46 ± 1.06	17.45 ± 1.10	17.47 ± 1.53	10.53 ± 1.54	12.04 ± 0.89

in zinc content were detected in any of the horizons considered. At a low contamination level (10 of PP per kg), zinc accumulated in the 0–10 cm layer only in August 2019. A medium level of contamination (20 g/kg) resulted in minor zinc accumulation in the topsoil in July 2019 and September 2020. However, the most pronounced geochemical barrier for Zn is created by a high pollution dose (30 g/kg). Barriers of low and medium contrast for this metal occur in the upper humus horizon; their effect can be seen throughout almost the entire observation period (except for the first 2 months after the contamination).

Based on the radial contrast coefficients, copper is highly mobile in soils contaminated with PPs. Already in the control samples, there is a tendency for leaching of this element from the upper to the underlying soil layers. Thus, for Cu at a depth of 10–20 cm in July and August 2020, the presence of a low-contrast barrier was detected. In September 2019, the upper layer accumulated this metal (R = 1.38); in other months of the two-year study period, the element content in both layers remained quite stable. With the presence of petroleum products and an increase in their concentrations the tendency for Cu migration only intensifies. For example, at a dose of 10 g PP/kg, a low-contrast barrier at depths of 10–20 cm was detected in September 2019 and in August 2020. However, the R value of 1.32, typical for a depth of 20–30 cm, indicates the ability of copper to further penetrate into lower soil horizons. At a medium level of pollution (20 g PP/kg), the mobility of copper significantly increases, the maximum rate of migration of this metal is observed in the topsoil, and its accumulation occurs at a depth of 10–20 cm. Low- and medium-contrast barriers in the second layer under consideration are detected

Table 6. Radial contrast coefficients of heavy metals in soil at sampling depths of 0–10, 10–20 and 20–30 cm. The values of low-contrast (R = 1.3–1.5) and high-contrast barriers (R = 1.5–3.0) are highlighted in bold.

Month	Sampling depth, cm	Radial contrast coefficient									
		Zn		Cu		Ni		Pb		Co	
		2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
Control (0 g/kg)											
June	0–10	0.97	0.99	0.85	1.01	0.97	0.96	1.04	0.97	0.96	0.98
	10–20	1.03	1.01	1.15	0.99	1.03	1.04	0.96	1.03	1.04	1.02
July	0–10	1.01	0.96	0.99	0.56	1.01	0.92	1.18	1.07	1.03	0.97
	10–20	0.99	1.04	1.01	**1.44**	0.99	1.08	0.82	0.93	0.97	1.03
August	0–10	0.97	1.00	0.71	0.61	0.95	0.97	1.03	0.97	1.00	0.95
	10–20	1.03	1.00	1.29	**1.39**	1.05	1.03	0.97	1.03	1.00	1.05
September	0–10	1.07	1.02	**1.38**	1.03	1.03	1.01	1.14	1.02	1.00	1.01
	10–20	0.95	0.97	0.90	0.75	0.97	0.91	0.90	1.16	1.04	0.99
	20–30	0.98	1.01	0.72	1.22	1.00	1.07	0.96	0.82	0.96	0.99
10 g/kg											
June	0–10	1.17	1.06	0.81	0.95	0.93	1.01	0.83	0.82	0.92	0.98
	10–20	0.83	0.94	1.19	1.05	1.07	0.99	1.17	1.18	1.08	1.02
July	0–10	1.09	1.26	0.87	0.82	1.00	0.99	1.25	1.28	1.00	0.97
	10–20	0.91	0.74	1.13	1.18	1.00	1.01	0.75	0.72	1.00	1.03
August	0–10	**1.35**	1.23	1.01	0.67	0.96	1.00	1.04	1.09	0.95	0.93
	10–20	0.65	0.77	0.99	**1.33**	1.04	1.00	0.96	0.91	1.05	1.07
September	0–10	1.02	1.24	0.93	0.75	0.95	0.99	0.87	1.05	0.99	0.94
	10–20	0.98	0.93	**1.44**	0.93	1.06	0.97	1.08	0.85	1.04	1.04
	20–30	1.00	0.83	0.64	**1.32**	0.99	1.04	1.06	1.10	0.98	1.03

Month	Sampling depth, cm	Radial contrast coefficient									
		Zn		Cu		Ni		Pb		Co	
		2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
					20 g/kg						
June	0–10	1.11	1.12	0.46	0.61	0.92	0.93	0.92	0.99	0.90	0.97
	10–20	0.89	0.88	**1.54**	**1.39**	1.08	1.07	1.08	1.01	1.10	1.03
July	0–10	**1.33**	1.14	0.75	0.60	0.99	0.97	1.03	0.94	0.99	0.92
	10–20	0.67	0.86	1.25	**1.40**	1.01	1.03	0.97	1.06	1.01	1.08
August	0–10	0.99	1.28	0.45	0.26	0.89	0.96	1.08	1.06	0.96	0.99
	10–20	1.01	0.72	**1.55**	**1.74**	1.11	1.04	0.92	0.94	1.04	1.01
September	0–10	1.25	**1.52**	0.90	1.05	1.05	1.07	1.04	1.22	1.07	0.96
	10–20	0.84	0.78	**1.65**	0.81	1.03	0.95	0.97	0.83	1.05	1.04
	20–30	0.91	0.71	0.45	1.14	0.92	0.99	0.98	0.95	0.87	1.00
					30 g/kg						
June	0–10	1.23	**1.31**	0.62	0.84	0.88	0.97	0.95	0.93	0.91	0.98
	10–20	0.77	0.69	**1.38**	1.16	1.12	1.03	1.05	1.07	1.09	1.02
July	0–10	1.16	**1.35**	0.56	0.99	0.93	0.97	0.93	1.05	0.94	1.01
	10–20	0.84	0.65	**1.44**	1.01	1.07	1.03	1.07	0.95	1.06	0.99
August	0–10	**1.31**	**1.35**	0.09	0.00	1.00	1.01	1.13	1.05	1.12	0.93
	10–20	0.69	0.65	**1.91**	**2.00**	1.00	0.99	0.87	0.95	0.88	1.07
September	0–10	**1.49**	**1.73**	0.33	0.21	0.90	1.03	1.01	0.89	0.96	0.97
	10–20	0.78	0.67	**1.97**	1.14	1.10	0.99	1.01	1.15	1.12	0.97
	20–30	0.73	0.61	0.70	**1.65**	1.00	0.98	0.98	0.96	0.92	1.05

throughout almost the entire observation period. High concentrations of the pollutant in the soil (30 g PP/kg) provoke an even more intensive migration of copper from the humus horizon to the underlying layers. Thus, in August of both years and in September 2019, contrast barriers were typical for a depth of 10–20 cm (the R coefficient ranged from 1.91 to 2.00). In September 2020, copper accumulations occurred at a depth of 20–30 cm; a contrasting barrier was detected (R = 1.65).

According to data obtained in laboratory experiments with soil columns, the accumulating capacity of sandy horizons is higher than that of loamy horizons (Gruzdkov et al., 2009). The authors point out the impulse nature of HM migration: pollutants accumulate in the humus layer to a certain limit, then their “impulse” of maximum concentration is generated, which eventually shifts to the underlying horizons. There is a consistent alternation of maxima and minima of pollutant concentrations down the soil profile. In our case, sandy loam soils are located down to a depth of 60 cm, followed by a textural layer represented by heavy loam (Table 1). This may explain the observed tendency for periodic accumulation of copper at depths of 10–30 cm during several months under the conditions of our experiment – heavy loam acts as a powerful geochemical barrier for copper compounds. Rainwater is trapped by this layer and can contribute to secondary contamination of the overlying soil layers with HMs, which is clearly seen in the case of copper in samples taken from a depth of 10–20 cm over the entire observation period. According to some authors (Mikhalchuk et al., 2022; Sindireva et al., 2022), in comparison with zinc, copper has a higher intra-profile migration activity due to less stable fixation of the element by soil organic matter. In our study, a similar pattern is observed: at a depth of 10–20 cm, copper accumulates most intensively; in the case of low (10 g PP/kg) and high (30 g PP/kg) contamination levels, accumulation was also detected in deeper soil layers. As a result, concentrations of copper in the topsoil are low, often (for example, in August–September) only very low concentrations of this metal are recorded, but they cannot be reliably detected by an instrument. It is known that copper is a fairly mobile element in terms of acidity (Zubarev and Mishchuk, 2019). The soil we studied is slightly acidic in the upper layer (pH = 5.59), then its acidity increases with increasing depth (Table 1), which also contributes to the vertical migration of water-soluble compounds of this element.

Conclusion

Thus, the factor of soil contamination with PPs mainly affects the migration and accumulation properties of copper and zinc. The tendency for zinc to accumulate in the upper soil horizon is due to its high ability for biogenic accumulation. This is facilitated by the presence of petroleum products in the soil which stimulate the activity of hydrocarbon-oxidizing bacteria. In addition, a lower migration capacity of this metal under increased air temperature and precipitation conditions contributes to its accumulation in the topsoil.

It is found that copper exhibits high mobility, which intensifies proportionally to the increase in the dose of PPs applied to the soil. This pattern is explained by a low degree of biogenic accumulation of this metal and its less stable fixation by soil organic matter, increased air temperature and precipitation during the observation period, as well as an increase in the acidity index down the soil profile. At the same time, the observed tendency towards an increase in copper concentration at a depth of 10–20 cm may be due to secondary contamination of this soil layer as a result of rainwater retention by a thick layer of heavy loam at a depth of 60 cm and below, which obviously acts as a geochemical barrier for Cu. In addition, sandy loam soil located at a depth of 0–60 cm along the profile has a greater capacity to accumulate HMs compared to loams, which also explains the observed instability in the processes of Cu migration.

Such metals as cobalt, lead and nickel are distributed evenly within the 0–30 cm layer; no correlation between their content in the soil and the level of its contamination with PPs is found.

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