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Main factors in polycyclic aromatic hydrocarbons accumulations in the long-term technogenic contaminated soil

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Abstract

The PAHs transformation in the soils of the coal mining enterprises impact zones and thermal power plants remains poorly studied. In turn, coal mining can be considered as a primary cycle in the production of electricity. One of the main sources of negative environmental impact is the coal mining industry located on the territory of the upland in the south of the East European Plain. The features of PAHs accumulation in the soils of fuel and energy enterprises have been studied on the example of mines impact zones with different service life and the current coal-fired power plant. It was established that, regardless of the period and intensity of the emission source, as well as its current status, the polycyclic aromatic hydrocarbons (PAHs) content in the soils of the impact zones was significantly higher than in the soils of the background territory. The content of low molecular and high molecular weight PAHs in the impact zones soils differed depending on the land use type, as well as the period and intensity of an industrial effect type. The pollutants content of in the soils of all considered impact zones significantly exceeded the background values and according to the low molecular weight PAHs content in the soils, they formed the following decreasing series: Mayskiy ≥ Ayutinsky > Novoshahtinsk > Power station > Background. According the high molecular weight PAHs content, the series changed to: Novoshahtinsk >Mayskiy ≥ Ayutinsky > Power station > Background. Soil pollution markers for enterprises of the fuel and energy complex were identified as pyrene and chrysene, which are part of coal, formed from the hydrocarbon sources. The influence of the power plant was accompanied by the benzo(g,h,i)perylene concentration increase.

Keywords: Priority PAHs, thermal power station, coal mining, anthracite, soil pollution.

Introduction

Industrial production is a key link in techno genesis for the whole environment and for individual components of the landscape. Diverse in intensity, duration and direction, anthropogenic impacts transform the initial parameters of the functioning of natural landscapes and create conditions for the ecological situation formation. The soil cover is a product of the natural and technogenic factors interaction, and, as the most stable component of the natural environment, reflects the level of long-term technogenic impact. The result of this impacts complex is manifested in a decrease of the soils quality up to their complete degradation, including those caused by chemical pollution. PAHs are considered as priority pollutants with carcinogenic activity (ATSDR, 1995; Adriano, 2001; Antoniadis et al., 2019). Soil pollution with PAHs is not only a direct environmental hazard for living organisms, adjacent components of natural and technogenic landscapes, but also acts as a risk factor for human's health (Asante-Duah, 2017; US EPA, 2020; Bezberdaya et al., 2022).

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Conventionally, all PAHs are classified into low molecular weight, which include 2- and 3-ringed compounds, and high-molecular, which include 4, 5, 6 or more ringed compounds. With an increase in the number of benzene rings in the PAH molecule, the stability of the pollutant in the medium increases, its lipophilicity increases, as well as its toxicity (ATSDR, 1995; Rengarajan et al., 2015; Abdel-Shafy and Mansour, 2016; IARC, 2020). At the same time, pollutants are amenable to microbiological and photochemical destruction, which allows the soil to self-purify over time (Haritash and Kaushik, 2009; Premnath et al., 2021).

Among the key anthropogenic sources of soil pollution in industrialized regions, FAO identifies mining, manufacturing, energy production, construction and transport (FAO and UNEP, 2021). Pyrolytic processes are an integral part of various branches of industrial production, which poses a threat to the continuous supply of PAHs to soils. The main sources of pollutants are emissions from thermal power plants and coal mining (Tsibart and Gennadiev, 2013; Sushkova et al., 2020).

There is not enough information about the PAHs transformation in the soils of the coal mining enterprises impact zones and thermal power plants. The coal mining industry can be considered as a primary sourse in the production of electricity. Therefore, the purpose of this study was to determine the main factors in polycyclic aromatic hydrocarbons accumulations in the long-term technogenic contaminated soil of the fuel and energy complex enterprises.

Material and Methods

The coal mining industry located on the territory of the East European Plain is one of the priority sources of negative environmental impact in the south of the above-mentioned area. Coal reserves in the south of the East European Plain amount up to 9.6 billion tons, while 7.2 billion tons of them are reserves of especially valuable anthracite, one of the best in the world in terms of calorific value and properties. Coal production volume reaches 2.3 million tons and is carried out at 7 mines (State Report, 2001). In the mining areas of the old development, significant areas are occupied by disturbed lands, quarries, dumps, mines. On this territory the special technogenic landscape-geochemical systems have been formed in the type of the mining landscapes (Perel'man, 2013). Mines not only occupy productive lands but serve as a secondary source of suspended particles and various pollutants due to dusting and combustion.

Coal reserves in the south of the East European Plain is mainly used by local industry, almost half is consumed by thermal power plants, one of which is Novocherkassk Power Station (NPS), with an installed electric capacity of 2258 MW. Annually, about 2.5 million tons of coal are consumed to produce the electricity at the station (Annual Report 2021, 2022). NPS is one of the five largest coal-fired power plants on the European territory, and the only one using anthracite pebbles. As a result of the station operation, more than 250 thousand tons of pollutants enter the atmosphere, including nitrogen oxides, carbon dioxide, sulfur oxides, greenhouse gases, PAHs, heavy metals, etc. (Korotkova et al., 2017; Sushkova et al., 2017).

The object of the study was the soils associated with mines of the coal mining industry and the impact zone of the NPS. Mines located in the western part of the region, near the rural settlements of Ayutinskaya and Mayskiy, as well as within the boundaries of the Novoshakhtinsk city. For mines located near Ayutinskaya and Mayskiy, the operation life was 58 and 48 years, and their closure took place in 2007 and 2002, respectively. The spoil mines, located on the territory of the Novoshakhtinsk city, ceased to function in 2002 after 71 years of its operation. The projected production capacity of the mines also varies. Thus, mines located near Ayutinskaya and Mayskiy are composed of coal mining waste from mines with a projected capacity of 400-600 thousand tons per year. The spoil mines, located directly within the boundaries of the Novoshakhtinsk city, was formed from an enterprise with a projected capacity of 1,500 thousand tons per year. Soils located far from industrial zones, including territories with a special protected status, were used as reference soil. The soil cover of the study area is predominantly represented by Haplic Chernozems, Gleyic Chernozems and Fluvisols (Table 1, Figure 1).

Soil sampling was carried out from each monitoring site to a depth of 0-20 cm in accordance with GOST 17.4.4.02-2017 (GOST 17.4.4.02-2017, 2019). Extraction of PAHs from soil samples was carried out with n-hexane. The pre-interfering lipid fraction was removed by saponification of 1 g of soil with a 2% KOH alkali solution. The PAHs in the extract were quantified by high performance liquid chromatography using HPLC Agilent 1260 (ISO 13877-2005, 2005). The content of individual PAHs in the analyzed samples was calculated using the external standard method.

Table 1. Soil properties of industrial and natural areas

Object type	Nearest city/village	Soil properties	Statistical parameters			
			Mean	Standart deviation	Minimum	Maximum
Power station	Novocherkassk	OM, %	2,0	0,5	1,2	2,9
		pH	7,5	0,5	6,2	8,5
		< 0,001,%	22,0	10,7	7,2	36,8
		< 0,01, %	45,4	12,8	22,4	64,4
Waste heap	Ayutinsky village	OM, %	2,6	1,2	0,3	3,5
		pH	7,6	0,3	7,3	7,9
		< 0,001,%	24,1	7,5	14,8	32,8
		< 0,01, %	47,3	11,7	33,6	58,8
	Novoshakhtinsk	OM, %	2,7	1,0	1,5	4,1
		pH	7,2	0,8	5,8	7,9
		< 0,001,%	8,3	6,5	2,2	20,4
		< 0,01, %	27,5	9,6	11,2	38,1
	Mayskiy village	OM, %	1,4	0,3	1,2	1,9
		pH	7,9	0,4	7,3	8,4
		< 0,001,%	23,9	9,3	8,0	34,0
		< 0,01, %	45,7	16,3	14,4	63,2
Background		OM, %	1,9	1,0	0,9	3,7
		pH	7,8	0,4	7,3	8,3
		< 0,001,%	21,7	11,6	1,6	31,7
		< 0,01, %	38,7	20,1	2,8	58,0

* OM - organic matter; < 0,001,% - soil particles less 0,001 mm, < 0,01,% - soil particles less 0,01 mm

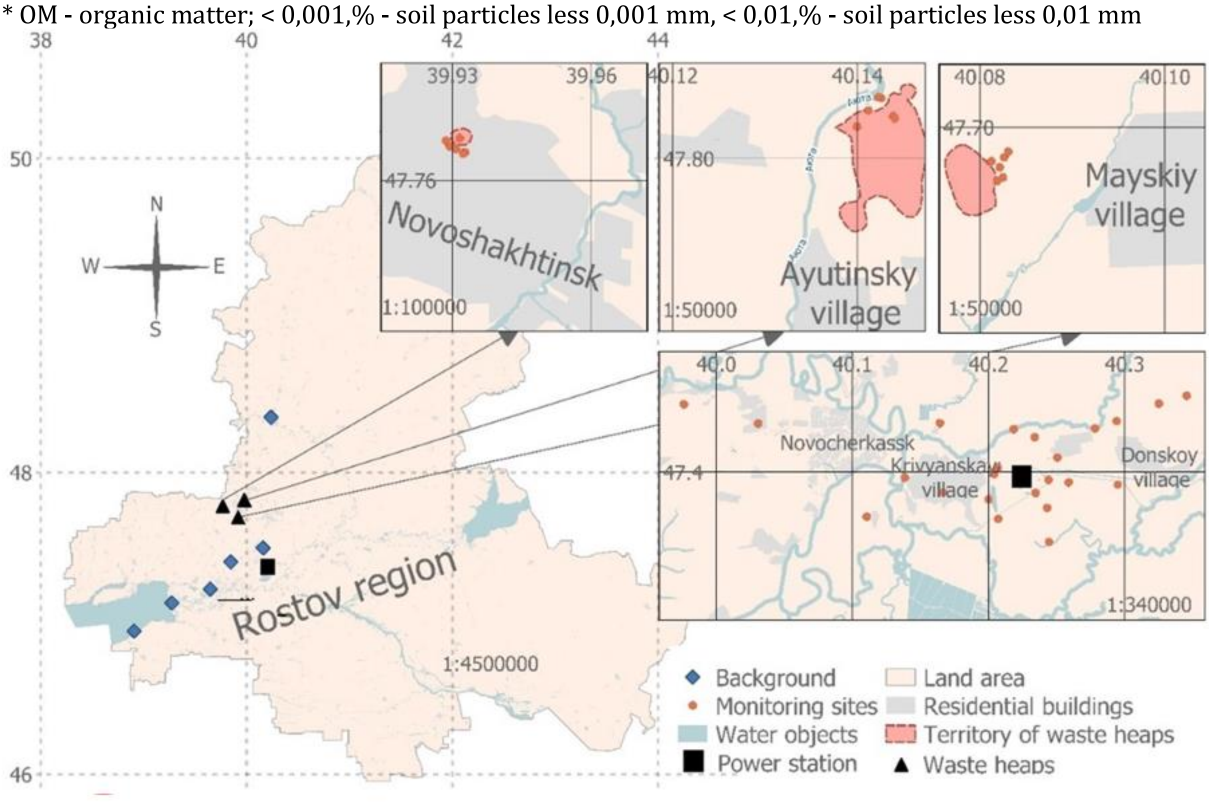

Figure 1. Soil sampling sites

During laboratory experiments, the concentrations of 15 priority PAHs were determined: low molecular weight: naphthalene, phenanthrene, anthracene, acenaphthene, acenaphthylene, fluorene; high molecular weight: pyrene, chrysene, benzo(a)anthracene, fluoranthene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(g,h,i)perylene (US EPA, 2020). All laboratory analyzes were performed in 3-fold analytical replication.

Statistical analysis of the obtained results was performed using STATISTICA 7 and OriginPro 2018 software. The significance of differences between samples was assessed using one-way analysis of variance followed by Tukey's post hoc test for unequal samples.

Results and Discussion

The content of PAHs in the soils of the background territory didn't exceed the norms established for agricultural territories (Maliszewska-Kordybach, 1996), average level of pollutants in soils of temperate latitudes and generally corresponds to the soils of natural areas (Wilcke, 2000; Sosa et al., 2017; Pikovskii et al., 2019; Hong et al,. 2020).

The content of low molecular weight (at $F=67$ and $p<0.0001$) and high molecular weight (at $F=47$ and $p<0.0001$) PAHs in the impact zones soils differed depending on the type of land use, the period and intensity of an industrial facility operation. At the same time, the content of pollutants in the soils of all considered impact zones significantly exceeds the background values. On average, in terms of the concentration of low molecular weight PAHs in the soils, they formed the following decreasing series: Mayskiy ≥ Ayutinsky > Novoshahtinsk > Power station > Background (Figure 2). For high molecular weight PAHs, the series changed to: Novoshahtinsk >Mayskiy ≥ Ayutinsky > Power station > Background. The higher values of low molecular weight PAHs in the soils of mines with a shorter service life (Mayskiy Ayutinsky), compared with the mine located within the Novoshahtinsk city, indicated ongoing processes of PAHs transformation from more nuclear compounds to less nuclear ones (Figure 2).

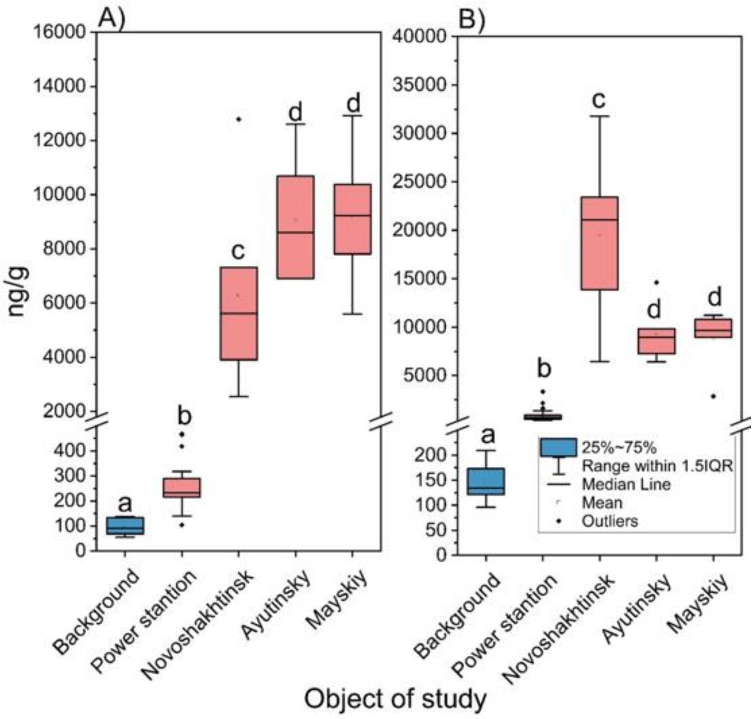

Figure 2. The content of low molecular weight (A) and high molecular weight (B) PAHs in soils of industrial and natural areas. Different letters indicate significant differences ($p < 0.05$) resulting from the post hoc Tukey's—honestly significant difference for unequal number test

The obtained values significantly exceeded the average pollution level in the soils of the coal mining enterprises and NPS impact zones. Often, the total content of PAHs in soils near coal mines does not exceed 6000 ng g, while in soils in the impact zones of power plants it reaches up to 3000 ng g (Wang et al., 2010; Kumar et al., 2014; Mukhopadhyay et al., 2017; Yakovleva et al., 2017, 2021; Amster and Lew levy, 2019; Gupta and Kumar, 2020). This may be due to the high affinity of the studied Haplic Chernozem for carbonaceous particles carriers of pollutants, as well as directly to PAHs themselves due to the high content of organic matter and finely dispersed fraction (Guo et al., 2010; Sushkova et al., 2019). On the one hand, this helps to fix pollutants and limit their migration into the tissues of living organisms (Guo et al., 2010; Cachada et al., 2014). On the other hand, the strong sorption of pollutants by the organic mineral part of the soil reduces the potential of the soil for self-purification through the microbiological destruction of PAHs due to their inaccessibility to microorganisms (Figure 2) (Liste and Alexander, 2002; Luo et al., 2012).

It is shown that the relative content of low- and high-molecular pollutants in soils of the background territory had the similar tendencies. Phenanthrene predominated (52%) in the composition of low molecular weight compounds while other PAHs had approximately equal shares. High molecular weight pollutants are characterized by an unexpressed dominance of the 4-ring association represented by fluoranthene, pyrene, and chrysene (14–16%), as well as benzo[b]fluoranthene (Figure 3).

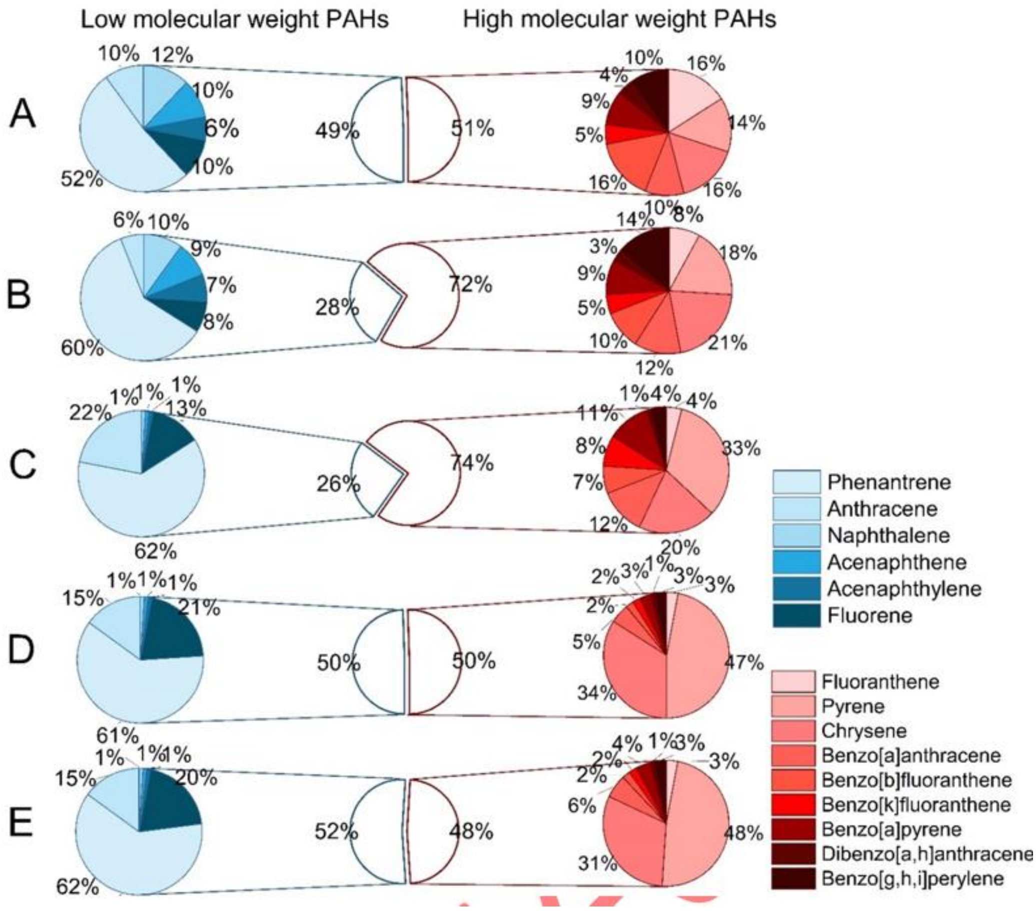

Figure 3. Relative content of individual PAH compounds in the composition of low and high molecular weight PAHs in the soils of the background area (A), the impact zone of the power plant (B), mines located near settlements Novoshahtinsk city (C), and on the territory Mayskiy (D) and Ayutinsky (E) mines

The main factors of the relative content of individual PAH compounds accumulation in the studied territories soils were established depending on the intensity and duration of an industrial facility operation and its current status (Figure 3). Under intense pressure from the power plant, there was a sharp shift in the relative content of pollutants towards the dominance of high molecular weight PAHs compounds, which indicates the lack of balance between the accumulation and destruction of pollutants in the soil. This kind of changes occurred mainly due to an increase in the pyrene, chrysene, and benzo(g,h,i)perylene proportion. Elevated concentrations of these compounds compared to other priority PAHs are markers of a pyrogenic-coal source, since pyrene, chrysene, and benzo(g,h,i)perylene are often found in soils of areas affected by fuel and energy complex enterprises and coal mining (Khaustov et al., 2021; Khaustov and Redina, 2022). At the same time, the proportion of individual low molecular weight compounds in this group is almost identical to the background territory.

In the soils of the impact zone of the mine located in the Novoshakhtinsk city, the proportion of low and high molecular weight PAHs was similar to the soils of the impact zone of the NPS. This indicates a strong fixation of 4-6 annular compounds by the organo-mineral part of the soil and the almost complete absence of pollutant transformation. However, differences were found in the dominant composition of the PAHs groups. The sum of low molecular weight compounds consists of 97% phenanthrene, anthracene, and fluoranthene, which is not typical for the soils of the NPS impact zone and indicates the presence of a large amount of these compounds in the composition of mined anthracites. Compared to the soils of the NPS impact zone, only pyrene and chrysene dominate in the composition of high molecular weight compounds. The share of pyrene (33%) is 1.5 times higher than the share of chrysene (20%). This indicates that the accumulation of pyrene in the soils of the impact zones under consideration is directly related to its presence in the composition of coal. The content of chrysene in soils is due not only to its presence in the composition of the fossil, but also to the formation of a pollutant, as in the process of coal pyrolysis at a thermal power plant.

In the technogenically disturbed soils of the impact zones of mines located near the settlements of Mayskiy and Ayutinsky, about half of the total content of pollutants falls on low molecular weight compounds, which is similar to the soil of the background site and indicates the stability of the ecosystem (Khaustov et al., 2021;

Khaustov and Redina, 2022). The composition of low molecular weight compounds is dominated by the same substances as in the soils of the more impact zone of the mines confined to the Novoshakhtinsk city (phenanthrene, anthracene, fluorene). In turn, with the PAHs active transformation in the soils of the impact zones of mines (Mayskiy and Ayutinsky), a decrease in the proportion of 5- and 6-ringed soils was observed due to an increase in 4-ringed soils. More than 70% of the total high molecular weight PAHs were pyrene and chrysene. This fact indicates that the transformation of 5- and 6-ringed pollutants proceeds with the formation of intermediate compounds of pyrene and chrysene, followed by the destruction of the PAH molecule to simple compounds. For example, pyrene in the process of microbiological or chemical oxidation according to the Fenton reaction type (Luo et al., 2014; Qin et al., 2017; Yan et al., 2017; Wang et al., 2018; Mazarji et al., 2021, 2022).

Conclusion

The study showed that in the soils of the impact zones of the largest thermal power plants in Russia, as well as mines located near the settlements of Mayskiy, Ayutinsky and Novoshahtinsk, the content of PAHs significantly exceeded the background levels, the average level for temperate latitudes, and the norms for agricultural soils. The content of pollutants in the soils of the impact zones varies depending on the type of land use, the period and intensity of operation of an industrial facility. In the soils of mines with a total service life of 48-58 years with a relatively low amount of coal mined per year (up to 600 thousand tons per year), the proportion of low-molecular and high-molecular PAHs is equalized, which is close to the values obtained for the soil of the background plots and is a consequence of the transformation of PAHs.

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