Influence of filling stations on the ecological condition of the atmosphere of urban agglomeration (on the example of Arkhangelsk)

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Abstract. The article assesses the effect of filling stations on the content of polyaromatic hydrocarbons (PAHs) in the snow cover of urban agglomerations. Snow cover is a good depositing matrix of organic pollutants and serves as an indicator of the state of the atmosphere. During the work, snow samples were taken at about two filling stations - within the city limits and outside the city limits. The samples carried out a quantitative chemical analysis of 14 PAHs and their total concentration. It was determined that the total concentration of PAHs in the snow cover does not depend on the distance from the filling station, the pollution of the atmosphere of PAH occurs regardless of the economic activity of the filling station. The maximum total concentration of PAHs is 283 ng / kg at one of the points near the filling station located in the city. According to the results of the research, it can be concluded that the pollution of the atmosphere of Arkhangelsk occurs in places of traffic congestion and is significantly lower than other cities of Russia (13 times lower than in Moscow). 83% of polyaromatic hydrocarbons in the snow of Arkhangelsk account for phenanthrene, fluoranthene, pyrene, chrysene, benz(b)fluoranthene and naphthalene. Benz(a)pyrene is 4% of the total PAH content. In order to study the ecological status of areas, it is necessary to investigate a complex of 14 PAHs. The content of benz(a)pyrene and naphthalene in snow samples taken in the city of Arkhangelsk does not exceed the MPC.

1. Introduction

Filling stations can cause environmental damage, polluting the atmosphere, soil, surface and groundwater with polyaromatic hydrocarbons. This has a negative impact on the environment and human health. It is shown that polyaromatic hydrocarbons can disrupt the normal functioning of the human endocrine system, have a detrimental effect on the epithelial cells of the respiratory tract and have an immunosuppressive effect [1-3]. PAHs are one of the most dangerous substances released during the operation of internal combustion engines. In the exhaust gases of internal combustion engines detects more than 20 different representatives of PAHs. The molecules of these hydrocarbons contain two, three and four ring structures and 12–20 carbon atoms. The most representative of them are pyrene, fluoranthene (not carcinogenic), chrysene (slightly toxic), phenanthrene, benz(b)fluoranthene, benz(j)fluoranthene, benz(k)fluoranthene, dibenz(a,h)anthracene, dibenz(a,j,a)anthracene, dibenz(a,h)pyrene, dibenz(a,i)pyrene, benz(a)pyrene (the most dangerous) [4-7]. In addition, PAHs and can be emitted into the environment when using tanks, as they are contained in certain quantities in different types of fuel. [8]. Thus, the pollution of the PAH atmosphere can be caused by two reasons: the operation of internal combustion engines and the evaporation of fuel.
vapors. The quality of atmospheric air reflects the snow cover in which pollutants can accumulate and remain. Due to the low temperature of the snow, even unstable compounds persist for a considerable period compared to the summer period when it settles on the soil. In the study of snow there is no need to make additional cleaning from interfering components, since there are no compounds in the snow that can mask the presence of the components being detected [8-11]. Snow cover can be considered an indicator of air pollution by toxic substances. Snow is an excellent depositing matrix and allows you to save the vast majority of organic compounds. The study of toxic substances in the snow can show the degree of air pollution in the immediate vicinity of various sources of man-made pollution, including filling stations [11-14].

Thus, the aim of the work is to study the snow cover in the zone of influence of filling stations on the content of various PAHs; assessment of the contribution of various PAHs to total pollution; comparing the levels of pollution of the snow cover of PAHs with the values of pollution of Arkhangelsk as a whole and other cities.

2. Methods
To study the effect of the filling station on the air environment, were selected two filling stations. The first filling station (filling station-1), with coordinates 64,52716° N, 40,56945° E.. There is a filling station in the city, close to the center of the city of Arkhangelsk, the motorway near the filling station is characterized by a significant traffic load. Nearby are large transport interchanges (Leningradsky Prospect - Naberezhnaya of Northern Dvina), which are subject to significant traffic loads.
The second filling station (filling station-2), with coordinates 64.62799° N, 40.67004° E. It is located on the margin of the city of Arkhangelsk, the motorways near the filling station are characterized by low density of traffic. The samples of snow were taken along the secondary road, which is in the forest, where there is no significant traffic flow (about 5 cars per hour). Due to the high surface area of trees, pollutants in the form of solid particles can settle on the leaves and fir-needles, as a result of which not all pollutants reach the surface of the earth. Snow is a good indicator of the accumulation of polycyclic aromatic hydrocarbons and other organic pollutants. In this regard, we decided to use the snow selected in the surface layer as the object of study. PAHs are deposited from the atmosphere to the snow surface during the entire winter period [15]. Sampling of snow was carried out in areas of both filling stations near highways of the city, with an interval of about 100 m. Sampling of snow was made on December 14. A small precipitation intensity was observed the day before, which allowed organic toxicants in the air to settle on the snow matrix. At the same time, the snow layer was already significant - more than 30 cm.

Sampling places are presented in Figures (1, 2). Snow was taken in specially prepared three-liter glass jars. Jars are pre-washed with running and distilled water several times and treated with hexane (HC). For the experiment, was selected surface layer of snow. To do this, opened the lid of the jar. Samples snow was scooped up the throats of jar, avoiding contact with uncharacteristic objects: sticks, household garbage, etc. After the jar was filled, the snow was compacted, without contact with the snow, by shaking the jar. Whereupon was continued to fill the jar with snow. After the jar was filled, it was closed with a special lid and marked. Samples were melted at room temperature. The volume of the sample after the converse of snow in the liquid phase ranged from 1.0 to 1.3 liters. For the extraction, 1 liter of melted snow water was used.

For sample preparation, the microextraction technique was used in the analysis. Before the extraction, the pH value was checked. If the pH value is outside the range (6-7) units pH was added dropwise sodium hydroxide solution and the pH was adjusted to the desired value. Then water was transferred into a 1000 ml calibrated flask, added 5 ml of hexane (grade 1, Cryochrome, Russia) by dosing pipetted and intensively mixed by magnetic stirrer for 30 minutes. After complete separation of the layers, the hexane extract was collected in a separate tube. The collected extract was kept in the freezer for at least 2 hours at a temperature of - 25 ° C. This is a method of the transfer of water remaining in the extract to ice. After that, the extract was quickly poured into clean tubes of the same volume. Then the extract was evaporated in a sand-bath with a temperature of (60 ± 5) ° C in a stream of air to a
volume of 1 ml. The residue of the extract was quantitatively transferred to a 1.5 ml vial and evaporated until the hexane was completely volatilized. At the end of the evaporation, 0.5 ml of acetonitrile was added to the vial and sealed. The concentration factor for sample preparation was 2000.

For the preparation of the eluent for gradient chromatography was used acetonitrile (grade 0, Cryochrome, Russia) and deionized water with a specific resistance of 18.2 MΩ cm, obtained using the Simplicity UV system (Millipore, France). The eluent was filtered using an Agilent (USA) eluent filtration system with a 45 mm diameter nylon membrane filter and 0.45 μm pore size. Samples were filtered before entering the chromatographic system using syringe membrane filters (Supelco, USA) with a diameter of 13 mm and a pore size of 0.2 μm.

To determine the PAHs, an Agilent 1260 chromatographic system (Agilent, USA) was used, equipped with a fluorescence detector. Separation was performed on a LiChrospher PAH chromatographic column (Agilent, USA), 250 × 3 mm, particle size 5 μm. The temperature of the column thermostat is 20 °C. The flow rate of the mobile phase is 0.56 ml/min. Fluorescence registration was carried out with programming of excitation and emission wavelengths in time. A spectrophotometric detector with a wavelength of 254 nm served as confirmation for the identification of peaks. The determined PAHs were identified in accordance with the calibration dependence. Graded solutions of naphthalene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene and benzo(g,h,i)pyrene, with concentrations from 0.5 mg/l to 0.001 mg/l were prepared from base solutions of SRS. Analysis of the calibration dependence of the peak area on the concentration showed compliance with linearity in the analyzed concentration range of analytes, and the correlation coefficient was more than 0.999. The concentration of each of PAHs was determined by the peak area.

3. Results and discussion

The results of the quantitative chemical analysis of snow samples taken in the area of filling stations are presenting in Tables 1, 2, Figures 1–2. They show that at each point some polycyclic aromatic hydrocarbons are found. The presence of PAHs in the snow we associate primarily with the influence of wheeled transport in these areas. The maximum content of PAHs was found in the sample taken 100 meters from the filling station - 1. The highest content of PAHs was observed at point L1 - phenanthrene 67.5 ng/kg. Also, markedly high concentrations of phenanthrene were observed in samples L0 and L1', L2'. A significantly high content of fluoranthene (62.5 ng/kg) was observed at point L1. The concentration of benzo(a)pyrene varies from 1 to 10.5 ng/kg. Minimum concentrations are observed for benzo(g,h,i)pyrene and benzo(k)fluoranthene (from 0.5 to 7.5 ng/kg). Dibenzo(a,h)anthracene was not detected in any samples in the filling station area - 1 and in the filling station area - 2. Acenaphthene was found only in one samples of L1 in an extremely low value, at the limit of determination, only trace numbers of contaminants were detected. Anthracene and benzo(a)anthracene are also recorded only in trace amounts around the filling station – 1.

The total content of PAH was also calculated for all samples. The average total PAH concentration in the filling station area - 1 was 135 ng/kg of snow, which is not much less than the average data for the city observed in 2017 (167 ng/kg) [16]. At the same time, around the filling station - 2, the average concentration of PAHs was 9.3 ng/kg, which indicates that no effect on the PAH content in the snow cover of the filling station (Fig. 1, 2). In our opinion, insignificant amounts of PAHs found around the filling station - 2 are associated with the operation of internal combustion engines of motor vehicles [17]. At the same time, samples around the filling station - 2 were taken near the highway, where there is observed, albeit not significant, but the movement of road transport.

Comparing the average values of total PAH concentrations around filling stations - 1 and filling stations - 2, it can be seen that these concentrations are significantly lower (14.5 times) in the filling station area - 2. Based on this, it can be concluded that the pollution of the PAH atmosphere occurs regardless of activity of the filling station.
When comparing the total concentrations of PAHs in the snow collected in the filling station area - 1 (Fig. 1, 3, Table 1) located in the city, it can be observed that in all snow samples, there are high concentrations of PAH, regardless of distance from the filling station. These values range from 49.5 ng/kg to 283 ng/kg.

Analyzing the data obtained, it can be seen that the total concentration of PAHs does not depend on the distance from the filling station and no effect is exerted on the PAH content in the snow cover.

When comparing the total concentrations of PAHs in the snow, collected around the filling station-2 (Fig. 2.4, Table 2) located outside the city limits, it can be observed that in all snow samples, extremely low concentrations of PAH are observed, regardless of distance from filling station. These values range from 6 ng/kg to 17.5 ng/kg.

![Figure 1](image1.png)

**Figure 1** - Place of sampling in the area of the filling station-1 and the concentration of the total content of polycyclic aromatic hydrocarbons in the snow cover, ng/kg.

![Figure 2](image2.png)

**Figure 2** - Place of sampling in the area of the filling station-2 and the concentration of the total content of polycyclic aromatic hydrocarbons in the snow cover, ng/kg.

In samples collected at the filling station-2 were not found: benz(a)anthracene, anthracene, phenanthrene, fluorene, acenaphthene, naphthalene and dibenz(a,h)anthracene. An anomalously high value of the total PAH content for this area can be observed at point P4. This may be due to the conditions of sampling, in particular in this place is the expansion of the road, with the help of which the oncoming transport was carried out.

According to the presented data, it can be observed that the total concentration of PAHs does not depend on the distance from the filling station and is observed to a significant degree only in the area of the filling station-1. The average total concentration of PAHs around filling station-1 is 135 ng/kg, which is not much less than the average total concentration of PAHs in 2017 in Arkhangelsk city of 167 ng/kg [26]. At the same time, the average total PAH concentration in the area of the filling station-1 is 13 times less than the average PAH concentration in Moscow city [15], 16 times less than in Novosibirsk city [17] and 17 times less than the average concentration in Blagoveshchensk city [18].
Based on the data, it can be concluded that the atmospheric pollution of Arkhangelsk, even in places where motor vehicles are crowded, is significantly lower than other cities in Russia. Probably, this fact is due to the fact that in comparison with the larger cities in Arkhangelsk there are less traffic flows.

To study the component composition of PAHs in the snow, we have constructed diagrams of component composition of samples taken around the filling station-1 (Fig. 5). Sample analysis showed that 83% of these pollutants accounted for phenanthrene, fluoranthene, pyrene, chrysene, benz(b)fluoranthene and naphthalene, the rest occupy 17%, among them benz(a)pyrene, which is 4%.

Table 1 - The content of polycyclic aromatic hydrocarbons in snow samples taken in the area of the filling station-1, ng/kg.

| №  | PAHs         | Place of sampling, ng/kg |
|----|-------------|--------------------------|
|    |             | L0          | L1           | L2           | L3           | L1'          | L2'          |
| 1  | Naphthalene | 24±10       | 21±3         | 2±0,9        | 1,5±0,7      | 11±4,8       | 18,5±8,1    |
| 2  | Acenaphthene| -           | 0,5±0,2      | -            | -            | -            | -            |
| 3  | Fluorene    | 6,5±2,7     | 9,5±4,0      | 0,5±0,2      | -            | 4,0±1,7      | 6,5±2,7     |
| 4  | Phenanthrene| 65,5±26,9   | 67,5±27,7    | 10±4,1       | 1±0,4        | 57,5±23,6    | 59,5±24,4   |
| 5  | Anthracene  | -           | 3,0±1,3      | 1,0±0,4      | -            | 1,0±0,4      | 3,0±1,3     |
| 6  | Fluoranthene| -           | 62,5±28,2    | 15±6,8       | 13±5,9       | 23,5±10,6    | -            |
| 7  | Pyrene      | 1,0±0,4     | 44±18        | 11±4,5       | 10,5±4,3     | 21±8,6       | 0,5±0,2     |
| 8  | Benz(a)anthracene | 3,0±1,3 | 2±0,9 | 2±0,9 | 2,5±1,1     |
| 9  | Chrysene    | 21,5±9,7    | 25±11        | 5±2,3        | 7±3,2        | 12±5,4       | 15,5±6,9    |
| 10 | Benz(b)fluoranthene | 6,5±2,9 | 22,5±9,9    | 3,5±1,5      | 5±2,2        | 7,5±3,3      | 9±3,9       |
| 11 | Benz(k)fluoranthene | 3,5±1,5 | 6,5±2,8    | 2,0±0,9      | 2,5±1,1      | 4±1,7        | 4,5±1,9     |
| 12 | Benz(a)pyrene| 4,5±1,9     | 10,5±4,5     | 3,5±1,5      | 4±1,7        | 4,5±1,9      | 7,0±3,1     |
| 13 | Dibenzo(a,h)anthracene | - | - | - | - |
| 14 | Benz(g,h,i)perylene | 4,5±1,8 | 7,5±3,1 | 2±0,8 | 3±1,2 | 4±1,6 | 6,5±2,7 |

Total concentration of PAHs, ng/kg

| №  | PAHs         | Place of sampling, ng/kg |
|----|-------------|--------------------------|
|    |             | P0          | P1           | P2           | P3           | P4           | P5           |
| 1  | Naphthalene | -           | -            | -            | -            | -            | -            |
| 2  | Acenaphthene| -           | -            | -            | -            | -            | -            |
| 3  | Fluorene    | -           | -            | -            | -            | -            | -            |
| 4  | Phenanthrene| -           | -            | -            | -            | -            | -            |
| 5  | Anthracene  | -           | -            | -            | -            | -            | -            |
| 6  | Fluoranthene| 2,5±1,1     | -            | -            | 1,5±0,7      | 3±1,4        | 1±0,5        |
| 7  | Pyrene      | 2,5±1,0     | 2,5±1,0      | 1,5±0,6      | 3,5±1,4      | 10,4         |
| 8  | Benz(a)anthracene | 1,5±0,7 | 2±0,9       | 3,0±1,4      | 1,5±0,7      | 3,0±1,4      | 2,0±0,9      |
| 9  | Chrysene    | 0,5±0,2     | 1,5±0,7      | 0,5±0,2      | 0,5±0,2      | 0,5±0,2      | 0,5±0,2      |
| 10 | Benz(b)fluoranthene | 0,5±0,2 | 0,5±0,2     | 0,5±0,2      | 0,5±0,2      | 1±0,4        | 0,5±0,2      |
| 11 | Benz(k)fluoranthene | 0,5±0,2 | 0,5±0,2     | 0,5±0,2      | 0,5±0,2      | 1±0,4        | 0,5±0,2      |
| 12 | Benz(a)pyrene| 1±0,4       | 1±0,4        | 1±0,4        | 1±0,4        | 2,5±1,1      | 1,5±0,6      |
| 13 | Dibenzo(a,h)anthracene | - | - | - | - |
| 14 | Benz(g,h,i)perylene | 0,5±0,2 | 1±0,4       | 0,5±0,2      | 0,5±0,2      | 2±0,8       | 0,5±0,2      |

Total concentration of PAHs, ng/kg
The composition of PAHs in the snow of the city of Arkhangelsk observed in 2017 is comparable to what we see in the snow selected in the area of the filling station-1. The percentage of phenanthrene around filling station-1 (32%) practically corresponds to its average percentage in Arkhangelsk (34%) in 2017. Also, the percentage of pyrene, fluoranthene and chrysene are approximately at the same level. In this regard, we can conclude that the activity of the filling station does not introduce any additional PAHs. The predominant PAHs in total concentrations are phenanthrene, fluoranthene, pyrene. At the same time, the component composition of PAH in Moscow shows a similar picture when analyzing snow samples taken in Arkhangelsk. Thus, we can conclude that in addition to benz(a)pyrene, traditionally defined for the purpose of studying the ecological status of areas, it is necessary to investigate all 14 PAHs. This conclusion is confirmed by the works [6, 11]. In particular, in the cities of Blagoveshchensk and Moscow, benz(a)pyrene accounts for only 7% of the total PAH content in urban snow [15, 17]. In Russia, MPCs only have naphthalene and benz(a)pyrene. MPCs naphthalene is 4000 ng/kg. The naphthalene contents determined by us do not exceed the MPCs at any of the sampling points. MPCs for benz(a)pyrene are established as the most resistant and potent carcinogen among PAHs, which is an indicator of the presence of carcinogenic PAHs in the environment. MPC benz(a)pyrene in water is 10 ng/kg. A concentration level comparable to the MPC was observed only at point L1. According to hygienic standards, drinking water should contain up to
0.005 mg/l of benz(a)pyrene and up to 0.01 mg/l of naphthalene, our results do not exceed the norm at any sampling point.

4. Conclusion

The total concentration of PAHs in the snow cover does not depend on the distance from the filling station, and the pollution of the atmosphere of PAH occurs regardless of the activity of the filling station. The maximum total concentration of PAH is 283 ng/kg at the sampling point L1. Atmospheric pollution in Arkhangelsk city occurs in places where traffic is concentrated and is much lower than other cities in Russia (13 times lower than in Moscow).

83% of polyaromatic hydrocarbons in the snow of Arkhangelsk account for phenanthrene, fluaranten, pyrene, chrysene, benz(b)fluaranten and naphthalene. Benz(a)pyrene is 4% of the total PAH content. In order to study the ecological status of areas, it is necessary to investigate a complex of 14 PAHs.

The content of benz(a)pyrene and naphthalene in snow samples taken in the city of Arkhangelsk is not higher than the MPC and is safe for the life of citizens.

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