

Article

The Mercury Content in the Upper Layers of Soils in the Selected Area of the Masovian Landscape Park

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Abstract: Due to the high toxicity of the element itself and its compounds, mercury poses a significant threat to the human natural environment, in particular to the soil and water environment. An additional risk is the possibility of mercury accumulation in various elements of the environment. Soil is one of the most important elements of the natural environment and is the main link in the mercury cycle. The cycle of this element is related to both the natural processes taking place in the environment and human activity. The presence of mercury in the natural environment poses a particularly high risk of its re-accumulation as a result of its re-emission. The aim of the research was to determine the mercury content in the upper layers of soils collected from the area and buffer zone of the Mazowiecki Landscape Park as a potential source of this element's re-emission to the environment. The paper presents the results of research on mercury content in the upper soil layer from the areas adjacent to the communication routes running through the area of the Mazowiecki Landscape Park and its buffer zone. The mercury content in the analyzed soil samples collected from the area of the Mazowiecki Landscape Park ranged from 0.082 mgHg·kg⁻¹ dm to 0.183 mgHg·kg⁻¹ dm, and in the soils from the cover, these values were slightly higher and ranged from 0.125 mgHg·kg⁻¹ dm up to 0.362 mgHg·kg⁻¹ dm. In order to avoid uncontrolled re-emission and, consequently, possible accumulation of this element in the environment, it is necessary to continue monitoring the level of mercury contamination, among others, in soil.

Keywords: Hg; contamination; protected areas; soil; traffic



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1. Introduction

Masovian Landscape Park is a protected area in east-central Poland, established in 1986, on a part of the former Siedlce Province and enlarged in 1987 by a part of the former Warsaw Province [1,2]. Currently, the Park covers an area of 15,710 ha. A buffer zone covering an area of 7992 ha was created around the Park. The buffer zone of the park is not a continuous zone, and its border in some places overlaps with the Park border [3]. Mazovian Landscape Park is located in the sub-province of Central Poland Lowland, the macro-region—Central Mazovian Lowland and meso-regions: Middle Vistula Valley, Garwolińska Plain and Wołomińska Plain. The analyzed area is characterized by high forest cover, in which pine dominates [4,5].

It is a protected area located within the administrative boundaries of Warsaw—the capital of Poland. This park was established to protect forests and the most valuable natural areas of Mazovia on the right side of the Vistula River. One of the largest peat bogs in Mazovia—Bagno Calowanie—is located in the Landscape Park. In this area, there are protected areas of the European Union under the NATURA 2000 program, the Special Area of Habitats Protection (PLH140001)—“Ostoja Bagno Calowanie”—and the Special Protection Area of Birds (PLB140011)—“Bagno Calowanie”. You can meet there the black

stork (*Ciconia nigra*)—a bird that is under strict species protection in Poland—as well as the moose (*Alces alces*), which is under protection in Poland.

The area of the landscape park is crossed by two national roads No. 17 and No. 50. The national road No. 17 is a connection between the Warsaw and Lublin agglomerations, while the national road No. 50 is the southern bypass of Warsaw for transit truck traffic. Significant density of vehicle traffic on both roads can be a major source of mercury, which, due to the high toxicity of the element itself as well as its compounds, is a significant threat to the human environment. An additional threat is the possibility of mercury accumulation in various environmental elements.

Due to the high toxicity of the element itself and its compounds, mercury poses a significant threat to humans. According to the World Health Organization, the inhalation of mercury vapor can produce harmful effects on the nervous, digestive and immune systems, lungs and kidneys and may be fatal. The inorganic salts of mercury are corrosive to the skin, eyes and gastrointestinal tract and may induce kidney toxicity if ingested [6]. The most harmful to humans are organic forms of mercury, found, among others, in fish. Fish accumulate this element by taking mercury directly from the water or by consuming other organisms. Similarly harmful to human health can be frequent consumption of water bird meat, e.g., mallard. Due to the strong genotoxic and teratogenic effects of mercury and the risk of damage by toxic metal of the central nervous system, consumption of food contaminated with mercury by children and pregnant women as the most vulnerable to the negative effects of this element should be limited [7,8].

Therefore, in a number of law regulations in Poland include the limited values of this element. This applies to the permissible content of mercury in soils as well as the concentration of this element in surface and groundwater and that intended for human consumption. The permissible values are, respectively, in soils 2.0 mg kg^{-1} [9], in surface waters $0.07 \text{ } \mu\text{g L}^{-1}$ [10], in groundwater 0.001 mg L^{-1} [11] and in water intended for human consumption $1.0 \text{ } \mu\text{g L}^{-1}$ [12]. The cited law regulations were created on the basis of the legislation of the European Union, of which Poland is a member.

The circulation of mercury in the environment is related to both naturally occurring processes and human activity. The natural release of mercury to environment occurs as a result of weathering of rocks and volatilization from the surface of seas and oceans. The significant influence on the mercury content in the environment has a foundation in metal processing industries and chemical works as well as earlier use of fungicides containing Hg (which is, of course, forbidden today) [13,14]. The results of the research presented by Xu et al. [15] indicate that the combustion of fossil fuels as well as the long-range transport process should be considered as the main sources of environmental pollution with mercury. Human activity increases the mercury load in the surrounding areas over time. As a result, the higher rates of re-emission of Hg appear leading to the longer time of emissions decrease including, among others, the pathways of exposure to human [16]. As a result, the higher rates of re-emission of Hg appear to be, the shorter amount of time it takes leading for Hg to re-enter, among others, pathways of exposure to humans [16].

An additional risk is the possibility of mercury accumulation in various elements of the environment. One of the most important elements of the natural environment, which is the main link in the mercury cycle, is soil. The occurrence of this element in the soil environment poses a particularly high risk of its subsequent accumulation as a result of re-emission to other elements of the environment.

The behavior of elements such as mercury in the soil environment is accompanied by a number of different processes, such as sorption or desorption, bio-accumulation or mineralization of organic matter. Additionally, the fate of mercury in the soil is influenced by such physicochemical properties of soils as the granulometric composition, reaction and the content of organic matter [13]. Lowering the pH of the soil to acidic favors leaching of mercury, while binding by organic matter or clay minerals reduces its re-emission. All this makes it extremely difficult to assess the degree of environmental contamination with mercury. Moreover, this assessment is hampered by the diversity of environmental

conditions caused by diversified human activities. Therefore, our previous research was conducted in an urban area, and our current research was carried out in a rural area. Due to the fact that one of the main sources of mercury in the environment is road transport and combusted fuel, the research was carried out in areas in the vicinity of communication routes. Particular attention was paid to protected areas, where favorable conditions for the re-emission of mercury to the environment may occur. The soils of protected areas require special attention as habitats for birds and mammals under species protection.

The aim of the research was to determine the content of mercury in the upper layers of soils taken from the area and buffer zone of the Mazowiecki Landscape Park as a potential source of re-emission of this element to the environment.

2. Materials and Methods

Soil samples were collected in May 2018 from the surface layer of soil (0–20 cm) from areas adjacent to communication routes running through the Masovian Landscape Park and its buffer zone ($52^{\circ}02'27.6''$ N $21^{\circ}23'40.4''$ E—coordinates of the middle point of the studied area). Nineteen square-shaped surfaces and dimensions 1.5×1.5 m were selected in the study area (Figure 1). Five samples were taken from each square, which were homogenized, and an average 1 kg sample was taken to analyze soil properties and mercury content.

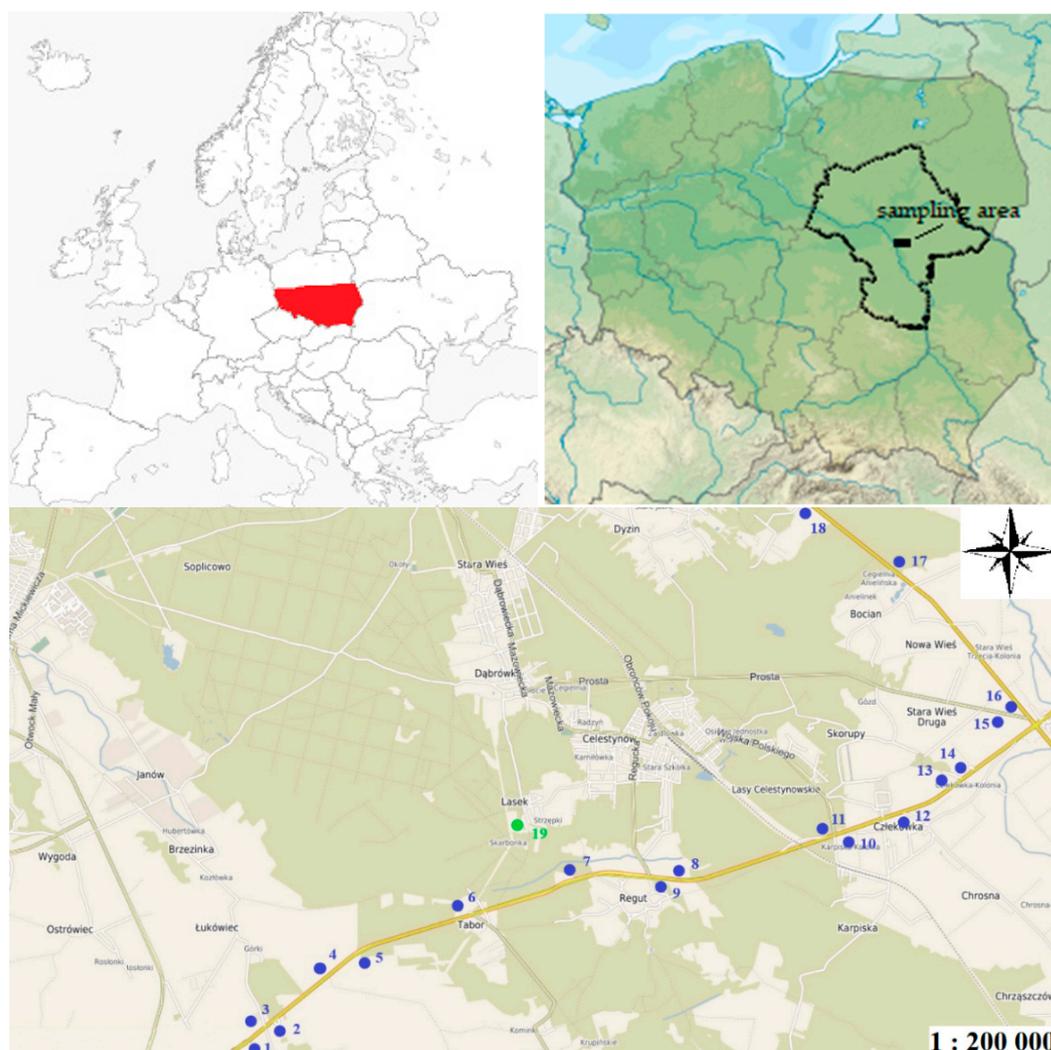


Figure 1. Locations of sampling points ($52^{\circ}02'27.6''$ N $21^{\circ}23'40.4''$ E).

The studied material was initially prepared by removing plants, stones and visible invertebrate parts for further analytical steps. Then, the samples were dried under the room temperature across 5 weeks in order to obtain an air-stable, dry mass. The dried samples were sieved through a sieve with a diameter of 2 mm. Afterward, the granulometric composition and other physicochemical soil properties were determined according to known soil procedures [17,18]: the soil granulometric composition was determined using the Bouyoucos erometric method modified by Casagrande and Prószyński; pH in 1 mol·L⁻¹ KCl was measured via the potentiometric method using a pH meter model inoLab pH 720 (WTW, Weilheim, Germany) with glass combine electrode; hydrolytic acidity (Hh) as well as the sum of alkalinity exchangeable cations (S) was measured using Kappen's method; Hh was measured after extraction of soil with 1 mol L⁻¹ (CH₃COO)₂Ca using a laboratory shaker WU-4 (PREMED, Marki, Poland) and determined using an automatic titrator, Jencons Digitrate (Leighton Buzzard, UK), to pH 8.2 with 0.1 mol L⁻¹ NaOH; S was measured after extraction of soil with 0.1 mol L⁻¹ HCl using a laboratory shaker WU-4 (PREMED, Marki, Poland) and determined using an automatic titrator, Jencons Digitrate (Leighton Buzzard, UK), to pH 8.2 with 0.1 mol L⁻¹ NaOH; cation exchange capacity of soil (CEC) was calculated by summing the values of Hh and S; organic matter content (OM) was measured by losing mass during anneal in 550 °C using the muffle furnace model SNOL 8.2/1100 (SNOL, Utena, Lithuania).

The concentration of the total mercury in this study was determined using the atomic absorption spectroscopy (ASA) method, and the analysis was performed on an AMA 254 mercury analyzer (Altec, Uherské Hradiště, Czech Republic).

For analysis, 100 to 300 mg of each sample were ground to a fine powder using a S100 model centrifugal ball mill (Retsch, Haan, Germany) to improve homogeneity. The sample prepared in this way was placed in a nickel cuvette, where it was automatically weighed and dried. The sample was then subjected to thermal decomposition in the oxygen stream to the gaseous form, and its decomposition products were transferred to the amalgamator for the selective capture of mercury. Mercury in the form of steam is released from the amalgamator by its short heating. The amount of mercury released was measured by the atomic absorption method (the detector in the AMA 254 analyzer is a silicon UV diode) at a wavelength of 254 nm in a system of two measuring cells. The samples were analyzed at specific cycle parameters: 60/160/60, which, in turn, means the times (in seconds) of drying, decomposition (at 550 °C) and waiting for the measurement. The mercury detection limit for this apparatus is 0.01 ng. For each sample, 2 replicates were made, and the obtained mean concentration result was given in mg·kg⁻¹ of dry matter.

During the determination of the mercury content in soils, to control accuracy and precision of the results, Certified Reference Standard EnviroMAT Contaminated Soil (SS-1) with a reference value of mercury 0.41 mg·kg⁻¹ was used.

Correlations between Hg concentrations in soil samples and the parameters characterizing soil properties were determined by a simple regression analysis. All the statistical calculations were performed using Statistica 6.0 software (StatSoft Polska, Krakow, Poland).

3. Results and Discussion

The results of granulometric composition of studied soil samples did not differ significantly. According to Polish Soil Society soil standards [19], soil samples were qualified to mechanical group sand and loamy sand (Table 1). The cited classification is based on the USDA (United States Department of Agriculture) soil texture classification system, widely used as an international standard. According to Gworek et al. [20], in sandy loams, the leaching of mercury and its compounds to the deeper soil layers occurs faster, which may result in the decrease of mercury concentration in the surface layer. The reduction in the mobility of elements such as mercury due to the high proportion of clay and silt in the studied soils is also indicated by Romeh [21] researching the soils near the Zagazig-Ismailia highway in Egypt.

Table 1. Granulometric composition of studied fractions (%).

| Sample No. | Granulometric Fractions Diameter in Millimeters (%) | | | | | | | | | | Mechanical Group |
|------------|---|-----|-------|----------|----------|----------|-----------|------------|-------------|--------|------------------|
| | >2 | 2–1 | 1–0.5 | 0.5–0.25 | 0.25–0.1 | 0.1–0.05 | 0.05–0.02 | 0.02–0.006 | 0.006–0.002 | <0.002 | |
| 1 | 15 | 6 | 14 | 45 | 2 | 6 | 3 | 3 | 3 | 3 | sand |
| 2 | 3 | 5 | 25 | 37 | 13 | 5 | 6 | 2 | 1 | 3 | sand |
| 3 | 8 | 5 | 13 | 47 | 7 | 5 | 7 | 3 | 1 | 4 | loamy sand |
| 4 | 1 | 1 | 3 | 28 | 47 | 6 | 8 | 3 | 0 | 4 | loamy sand |
| 5 | 1 | 1 | 13 | 56 | 26 | 2 | 1 | 2 | 0 | 0 | sand |
| 6 | 1 | 2 | 17 | 45 | 23 | 5 | 2 | 5 | 0 | 0 | sand |
| 7 | 6 | 5 | 19 | 35 | 23 | 0 | 7 | 2 | 0 | 3 | sand |
| 8 | 1 | 1 | 20 | 50 | 19 | 3 | 2 | 3 | 0 | 2 | sand |
| 9 | 2 | 4 | 11 | 49 | 17 | 6 | 5 | 2 | 1 | 4 | loamy sand |
| 10 | 2 | 1 | 12 | 53 | 22 | 5 | 2 | 3 | 0 | 0 | sand |
| 11 | 2 | 1 | 18 | 47 | 18 | 7 | 3 | 4 | 0 | 1 | sand |
| 12 | 1 | 1 | 13 | 53 | 24 | 3 | 3 | 2 | 0 | 0 | sand |
| 13 | 3 | 2 | 5 | 22 | 44 | 13 | 7 | 5 | 0 | 0 | sand |
| 14 | 2 | 2 | 7 | 24 | 41 | 10 | 5 | 5 | 1 | 4 | loamy sand |
| 15 | 12 | 11 | 8 | 24 | 13 | 6 | 9 | 7 | 4 | 6 | sandy loam |
| 16 | 12 | 6 | 11 | 42 | 10 | 7 | 5 | 3 | 0 | 5 | loamy sand |
| 17 | 5 | 5 | 15 | 29 | 16 | 8 | 11 | 5 | 3 | 3 | loamy sand |
| 18 | 4 | 3 | 10 | 38 | 23 | 8 | 7 | 4 | 0 | 3 | loamy sand |
| 19 | 1 | 0 | 11 | 61 | 22 | 2 | 3 | 0 | 0 | 0 | sand |

The results of physicochemical properties of studied soils are included in Table 2. The pH in the studied soil samples proved that soils originating from the research area are rather acidified, as pH was in the range of 3.35–4.75. In two investigated samples, pH in the range of 6.65–6.75 indicated the neutral soil. Similar pH values were determined by Kuziemska et al. [22] during research in areas adjacent to the national road No. 63. The authors indicated that, in their opinion, road dust may contribute to an increase in the pH of soils adjacent to the road.

In order to determine the values of cation capacity exchange (CEC), the values of hydrolytic acidity (Hh) and sum of alkalinity exchangeable cations (S) were calculated, and then the obtained values were added. The investigated soil samples were characterized by diversified Hh values. The highest value equal to $2.08 \text{ cmol}(+) \cdot \text{kg}^{-1}$ was found in Sample 4, whereas the lowest equal to $0.39 \text{ cmol}(+) \cdot \text{kg}^{-1}$ was obtained in Sample 16. Values of the S were also diversified. The highest value equal to $36.14 \text{ cmol}(+) \cdot \text{kg}^{-1}$ appeared in Sample 16, whereas the lowest value equal to $28.28 \text{ cmol}(+) \cdot \text{kg}^{-1}$ was found in Sample 9. Cation capacity exchange (CEC) was in the range of $29.63\text{--}36.88 \text{ cmol}(+) \cdot \text{kg}^{-1}$.

In the studied soil samples, the organic matter content was in the range of 1.00–4.75%. As reported by various researchers, organic matter can both mobilize and immobilize Hg, depending on soil pH and aerobic conditions in soils. According to Poulin et al. [23], mercury has affinity to the OM content in the soil that tends to cumulate this element in surface soil layers. This is also confirmed by the results of research conducted in Switzerland by Gilli et al. [24].

The highest mercury content of $0.362 \text{ mg Hg} \cdot \text{kg}^{-1} \text{ dm}$ was found in Sample 15. The lowest content of this element, $0.082 \text{ mg} \cdot \text{kg}^{-1} \text{ dm}$ was determined in Sample 10. For comparison, Sample 19 from the forest area away from the tested roads contained $0.117 \text{ mg Hg} \cdot \text{kg}^{-1} \text{ dm}$, which was a value similar to that determined in several samples collected along the communication routes covered by the research. According to the law regulations, the considered soils can be treated as unpolluted with mercury [9].

Table 2. Physicochemical properties and total Hg content of soil samples.

| Sample No. | Fractions (mm) | | | OM (%) | pH KCl | Hh | S (cmol(+)·kg ⁻¹) | CEC | Hg (mg·kg ⁻¹ dm) ± SD |
|------------|----------------|----------------|--------|--------|--------|------|-------------------------------|-------|----------------------------------|
| | 2–0.05 | 0.05–0.002 (%) | <0.002 | | | | | | |
| 1 | 88 | 9 | 3 | 2.75 | 6.65 | 0.51 | 32.52 | 33.03 | 0.114 ± 0.023 |
| 2 | 88 | 9 | 3 | 2.50 | 3.72 | 1.48 | 29.88 | 31.36 | 0.163 ± 0.039 |
| 3 | 85 | 11 | 4 | 1.50 | 4.60 | 0.61 | 30.40 | 31.00 | 0.104 ± 0.019 |
| 4 | 85 | 11 | 4 | 4.75 | 3.60 | 2.08 | 29.76 | 31.84 | 0.327 ± 0.065 |
| 5 | 97 | 3 | 0 | 2.00 | 3.68 | 1.22 | 29.39 | 30.61 | 0.118 ± 0.024 |
| 6 | 93 | 7 | 0 | 2.75 | 3.52 | 1.60 | 28.41 | 30.01 | 0.161 ± 0.039 |
| 7 | 88 | 9 | 3 | 4.25 | 4.75 | 1.38 | 31.00 | 32.37 | 0.183 ± 0.054 |
| 8 | 93 | 5 | 2 | 1.50 | 3.82 | 1.33 | 28.88 | 30.21 | 0.118 ± 0.019 |
| 9 | 88 | 8 | 4 | 2.75 | 3.78 | 1.35 | 28.28 | 29.63 | 0.161 ± 0.039 |
| 10 | 95 | 5 | 0 | 1.00 | 3.70 | 1.12 | 30.23 | 31.35 | 0.082 ± 0.018 |
| 11 | 92 | 7 | 1 | 4.75 | 3.35 | 2.00 | 31.02 | 33.02 | 0.160 ± 0.038 |
| 12 | 95 | 5 | 0 | 2.50 | 3.81 | 1.03 | 30.95 | 31.98 | 0.125 ± 0.026 |
| 13 | 88 | 12 | 0 | 2.00 | 3.46 | 1.48 | 31.11 | 32.59 | 0.178 ± 0.052 |
| 14 | 85 | 11 | 4 | 2.25 | 3.49 | 1.48 | 31.48 | 32.96 | 0.171 ± 0.051 |
| 15 | 74 | 20 | 6 | 2.25 | 4.02 | 1.08 | 35.81 | 36.88 | 0.362 ± 0.083 |
| 16 | 87 | 8 | 5 | 1.50 | 6.76 | 0.39 | 36.14 | 36.53 | 0.129 ± 0.026 |
| 17 | 78 | 19 | 3 | 2.50 | 5.10 | 0.79 | 33.16 | 33.95 | 0.160 ± 0.037 |
| 18 | 86 | 11 | 3 | 3.75 | 4.38 | 1.29 | 31.23 | 32.52 | 0.251 ± 0.045 |
| 19 | 97 | 3 | 0 | 1.75 | 3.47 | 0.96 | 32.57 | 33.53 | 0.117 ± 0.019 |

OM—organic matter content; H—hydrolytic acidity; S—sum of alkalinity exchangeable cations; CEC—cation exchange capacity.

The obtained results are comparable with the results of the research conducted by Małuszyński et al. [25] along communication routes in the Ursynów district in Warsaw (0.0334–0.4004 mg·kg⁻¹). Similar results were also obtained by Gruba et al. [26] in soils under pine and fir stands located near the S7 expressway between the cities of Skarzysko-Kamienna and Kielce, central Poland (average 0.225 mg·kg⁻¹). Lower mercury contents, ranging from 0.013 to 0.041 mg·kg⁻¹, were determined by Jaworska and Klimek [27] in surface layers of soils collected in the vicinity of the A1 motorway, along the international E75 route, central Poland. Much higher contents, ranging from 2.26 to 3.68 mg·kg⁻¹, were determined in studies conducted on canola soils along the Zagazig-Ismailia highway in Egypt [21], which the author explains with a high content of organic matter in the investigated arable soils.

Additionally, statistically significant correlation between the properties of the soil and the content of Hg was analyzed. It was found statistically significant a correlation between soil grain size, the content of organic matter and mercury content at the confidence level of 95%.

Figure 2 shows a negative correlation between the Hg concentration and soil grain size–sand (ø 2.0–0.05) content in the tested samples, whereas Figure 3 shows a positive correlation between the concentration of mercury and soil grain size–silt (ø 0.05–0.002) content in the studied soils. Figure 4 shows a positive correlation between the Hg concentration and soil grain size–clay (ø < 0.002)

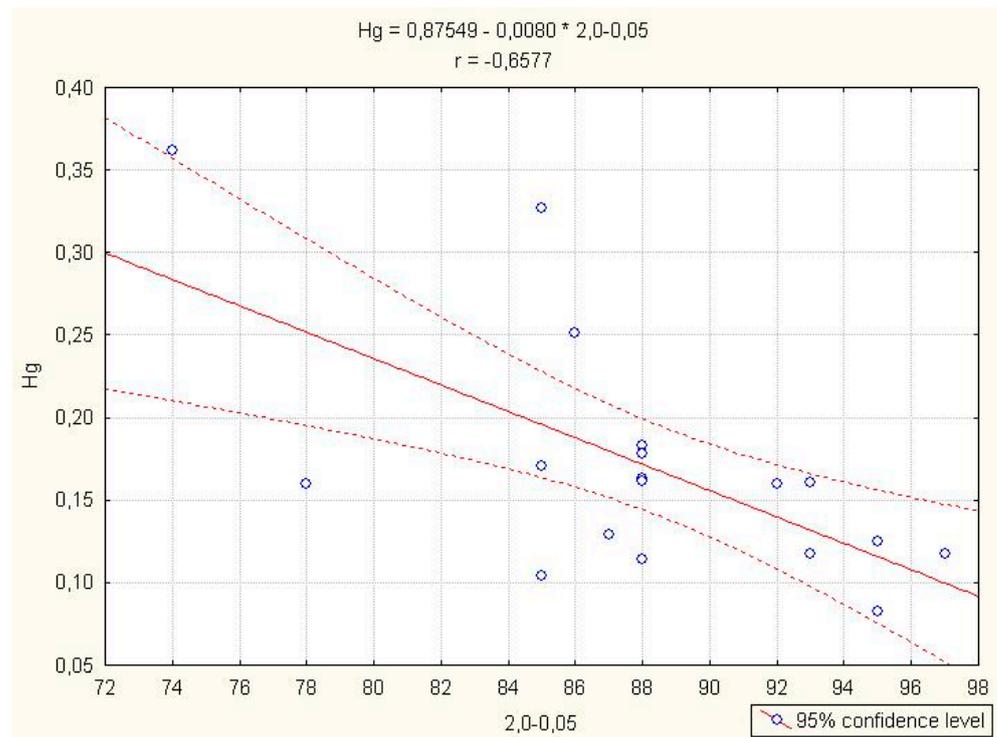


Figure 2. The correlation between Hg concentrations in the soil and soil grain size (2.0–0.05 mm) ($p < 0.05$, $n = 19$).

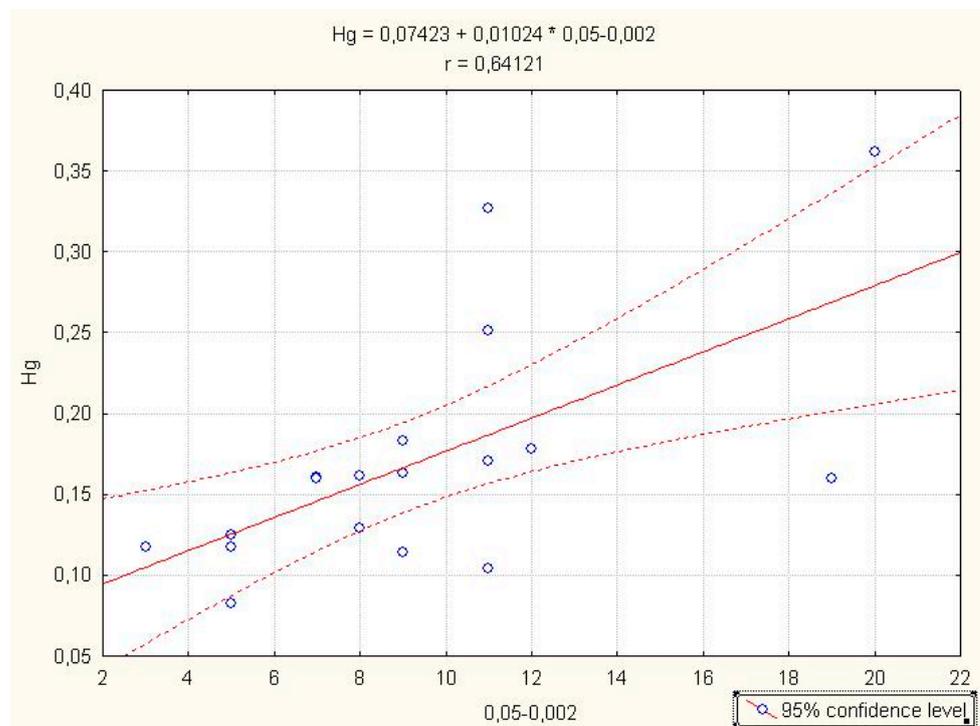


Figure 3. The correlation between Hg concentrations in the soil and soil grain size (0.05–0.002 mm) ($p < 0.05$, $n = 19$).

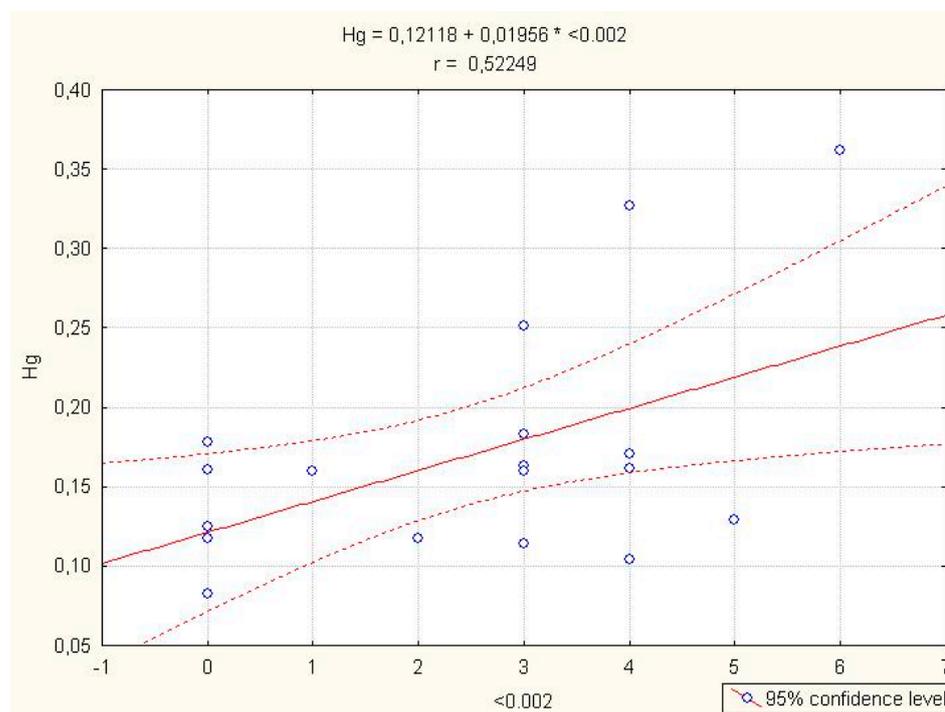


Figure 4. The correlation between Hg concentrations in the soil and soil grain size (<0.002 mm) ($p < 0.05$, $n = 19$).

The positive correlation between the Hg content and the content of organic matter is shown in Figure 5.

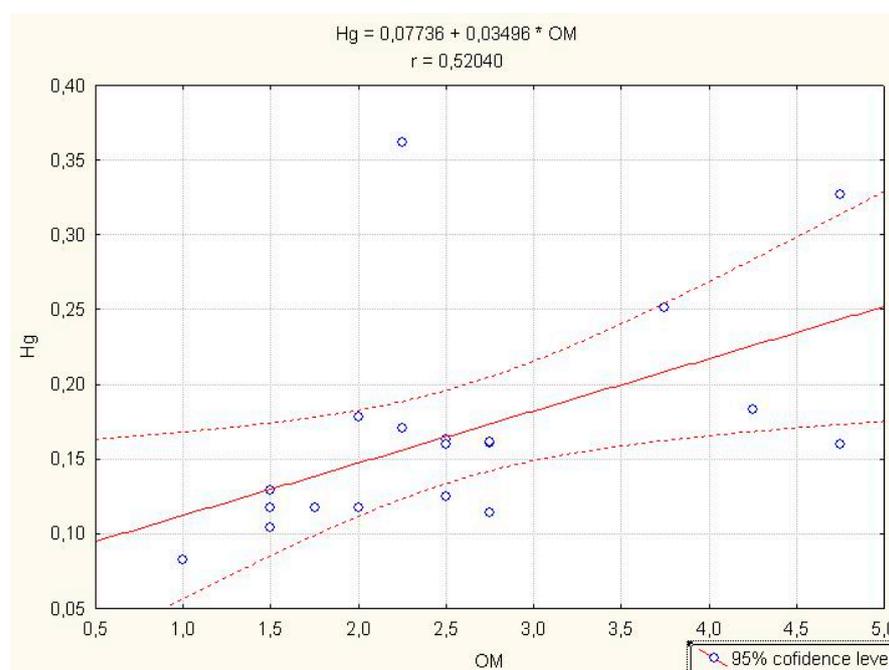


Figure 5. The correlation between Hg concentrations in the soil and organic matter content ($p < 0.05$, $n = 19$).

The significant correlation measured between soil grain size and mercury content indicates that soils containing higher amounts of silt may contain more mercury, while soils containing higher amounts of sand can be expected to contain lower levels of mercury. At

the same time, the found positive correlation between the content of organic matter and the content of Hg indicates that this factor may also significantly affect the content of this element in soils. Both Gworek et al. [20] examining the impact of road traffic in the vicinity of Warsaw on the mercury content in soils and plants, as well as Jaworska and Klimek [27] conducting research in the vicinity of the A1 motorway along the international E75 route, confirm the influence of soil texture on the content of this element in soil. It follows from these studies that the concentration of Hg in soils tends to increase with the decrease in grain size, which is caused by the tendency of this element to bind to finer particles.

4. Conclusions

Soil samples collected for research from the selected area of the Masovian Landscape Park were characterized by a significant share of sand and an acidic reaction, which may contribute to leaching of mercury from the upper layers. The predominance of alkaline ions over acidic ions in the soil, as well as the presence of organic matter in the range from 1.00 to 4.75%, may result in the accumulation of mercury in the surface layers of the soils under study.

The analysis of the relationship between the properties of the studied soils and the mercury content showed a significant correlation between the soil grain size and the organic matter content and mercury content. The value of the correlation coefficients indicates that as the proportion of fine particles in the soil increases, more mercury may be retained in the upper layers of the studied soils. Similarly, in the case of organic matter, its increasing content may increase the amount of this element in upper layers.

The total content of mercury determined in the studied soils varied from 0.082 to 0.362 mg/kg dm, which allows us to treat the studied soils as unpolluted according to the Polish regulation. Regardless of whether the soil is contaminated or not, it should be remembered that mercury is an element that changes easily and therefore can move quickly if there are conditions that mobilize it. Therefore, it is worth conducting constant observations of soil contamination with mercury in order to prevent uncontrolled re-emission and re-accumulation in the soil environment. More and more often, in order to avoid the negative effects of mercury pollution of the environment, remediation is proposed.

One of the methods of reducing soil contamination with this element is the immobilization of mercury with ground sulfur. The results obtained by Król et al. [28] 24 h after the remediation treatment prove that the emission volume was reduced by 26 ÷ 92%. Another way is bio-remediation, i.e., the use of living organisms, for example, bacteria, to bio-accumulate Hg. As reported by Nurfitriani et al. [29], *Fictibacillus nanhainensis* and *Bacillus toyonensis* were able to accumulate Hg with bio-accumulation of up to 82%.

It should be remembered at all times that the effectiveness of our actions to reduce mercury pollution of the environment requires taking into account the place and nature of the pollution.

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