# Biogeochemical Redistribution of Industry-Caused Sulphur in an Urban Ecosystem

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## Abstract

Sulphur content in the system woody plants–forest litter–soil (the profile-genetic level) was investigated within the boundaries of the urban territory. High accumulation of total sulphur in the needles and leaves of tree species was demonstrated. A substantial depositing ability of the forest litter, upper organics-accumulating and texture horizons of urban soil with respect to the industry-caused sulphur compounds was discovered. A close connection between the sulphur content of tree needles (leaves) and forest litter, as well as the soil profile, was revealed. It was demonstrated that sulphur accumulation in assimilation organs of plants and its physicochemical redistribution in the soil profile might serve as the ground for the adequate evaluation of the degree of industrial load on an urban ecosystem.

## INTRODUCTION

Industry-caused fluxes formed in urban agglomerations are extremely diverse and contain high concentrations of toxic compounds. The main media depositing the pollutants in an urban ecosystem are the soil cover, woody vegetation and ground water, which bring about substantial danger for the living conditions of the population. Pollutants participating in natural biogeochemical cycles undergo chemical transformations and get redistributed over the components of the urban environment. As a consequence, they may affect humans directly or indirectly. Investigation of the routes of pollutant migration and transformation is necessary not only for an optimal evaluation of the ecological situation over urban territories [1] but also for the development of measures aimed at a reduction of hazardous industry-caused consequences and at the improvement of the quality of environment.

Under the conditions of the atmospheric industrial pollution of cities, sulphur compounds hold one of the leading positions among the toxic compounds. Under industry-caused pollution, sulphur-containing compounds are emitted into the atmospheric air in the form of gases, aerosol and dust particles. Among them, the most dangerous agents are gaseous compounds: sulphur oxides and hydrogen sulphide. In this situation, sulphur dioxide accounts for about 95 % of the total amount of sulphur-containing compounds in industrial emissions [2, 3]. Sulphur dioxide is not only the most widespread but also one of the most toxic pollutants. For example, it is known that this compound affects plants as a strong photosynthetic poison [4]. The mechanism of the toxic action of $SO_2$ on plants has been investigated in detail. Its key phenomena are acidation of the cytoplasm, disturbance of the performance of many enzymes, change of the ion balance of a cell, appearance of autocatalytic chain reactions of free radical oxidation, destruction of photosynthetic structures, suppression of photosynthesis, changes of the nitrogen and carbohydrate exchange, distortion of the synthesis of proteins and lipids [4, 5].

Accumulation of total sulphur in the assimilation organs of trees often serves as one of the main diagnostic indices of the industrial pollution of environment [2, 4]. Under the action of aerial industrial emissions, the sulphur content of pine needles may be several times higher than the background level [6, 7]. The data on the accumulation of industry-caused sulphur in the soil of polluted territories are rather scarce [3]. In addition, we did not find any publications concerning redistribution of sulphur-containing compounds in soil at the profile-genetic level within the boundaries of the urban ecosystem. At the same time, soil plays an important part in transformation and neutralization of pollutants (including industry-caused sulphur) under the conditions of industrial pollution [1, 3].

Soil stability towards pollutants is determined by its buffer capacity. The threshold of soil stability may be exceeded as a consequence of long-term aerial arrival of industry-caused sulphur into the ecosystem. As a result, soil becomes a toxic medium for plant growth and development, and a source of additional pollution of the ecosystem.

Thus, the results of the investigation of sulphur accumulation and redistribution in the most important components of the ecosystem – plants and soil – allow one to evaluate the degree of its pollution adequately.

The goal of the investigation was to examine biogeochemical accumulation of industry-caused sulphur over the urban territory in the system woody plants–forest litter–soil, to determine the specific character of the geochemical redistribution of sulphur in the soil profile, to reveal a connection between the concentrations of this toxic agent in soil and in woody plants.

## REGION, OBJECTS AND METHODS OF INVESTIGATION

Investigations were carried out at the territory of Irkutsk, a large industrial centre in East Siberia. The index of pollution of the atmospheric air in the city is 19.5, so the city is included into the list of ecologically unfavourable territories of he country [8]. Every year, industrial enterprises and motor transport emit up to 140 thousand tons of hazardous substances into the atmosphere of Irkutsk, which is 12–14 % of total emissions over the Irkutsk Region [9]. The total number of plants in the city is 147; among them 21 ones belong to the category of potentially dangerous plants for population. Five large industrial junctions are distinguished in the peripheral parts of the urban territory: Novo-Lenino, Irkutsk-2, Sinyushina Gora, Zhilkino, airport). One of the main sources of pollution of the atmosphere in Irkutsk is the emission from motor transport; they account for about 52 % of the total amount. Emission from industrial enterprises and motor transport contain large amounts of acidic components, especially sulphur oxides. Their annual emission into the atmosphere of the city reaches 58 thousand tons [8]. The high level of pollution of the urban territory is also due to the complicated orographic and natural climatic features. Frequent calm episodes, weak wind, mist and high air humidity serve as the reasons of stagnation of polluted air masses and the formation of smog over the orographically closed territory of Irkutsk.

The city is situated in the zone of sub-taiga pine forest. The soil cover of the adjacent territories is represented mainly by grey forest and soddy podzolic and sandy loam soil. The parent rocks are Jurassic sandstones which are characterized by the horizontal formation, low strength and are coated with modern quaternary alluvial sediments [10].

Investigations were carried out during the years 2002–2004 in the park and forest-park zones of Irkutsk. The anthropogenic transformation of the soil cover and vegetation over these territories is minimal; the soil conserves its natural biogeochemical characteristics, while plants better adapt themselves to pollution. So, the evaluation of functioning in the system soil–plant was expected to be the most adequate.

The objects of investigation were natural grey forest soils of different disturbance degree, and wood species: pine (*Pinus sylvestris* L.), siberian larch (*Larix sibirica* Ledeb.), poplar (*Populus* sp.), birch (*Betula pendula* Roth.). Test grounds (TG) were laid in the sites of soil and woody plant examination in agreement with the accepted procedures [11, 12]. Total number of TG laid during the investigation was 13: TG No. 1 was in the forest park zone in Akademgorodok; TG No. 2 in the Central Park for recreation; TG No. 3 in a green zone in Baikalskiy micriodistrict; TG No. 4 in a park zone in Lisikha microdistrict; TG No. 5 in Parizhskoy Kommuny park); TG No. 6 in a forest park on Kayskaya Gora; TG No. 7 in a forest park near the airport; TG No. 8 in a forest zone near the Regional hospital; TG No. 9 in a park near Irkutsk-Sortirovochnaya station; TG No. 10 in the forest park in Novo-Lenino; TG No. 11 in a park in Irkutsk-2; TG No. 12 in a forest massif at the territory of the Regional Station of Young Naturalists; TG No. 13 in a forest zone in Iskra village.

Examination of soil was carried out by means of soil profiles and with the help of averaged (joint) samples taken using the square envelope method in the forest litter (O), as well as in humus-accumulating horizons (Ad, A). The type names of soil were given according to the generally accepted classifications [13, 14] taking into account also modern nomenclature and classification of urban soil [5]. The multi-aspect examination of soil samples was carried out in parallel with the examination of woody plants. The state of trees was evaluated on the basis of a number of morphostructural parameters: crown defoliation level, degree of decolouration of needles and leaves, for coniferous trees on the basis of the length of the two-years-old sprouts, lifetime of needles, their size, mass and number on sprouts. Complete geobotanical description was made for each TG (according to Yaroshenko), the percentage of road and path network was estimated, the information about the occurrence of fire events and the presence of pests was also collected. Needles (leaves) and soil were sampled on all the TG to analyse sulphur content. The background TG were laid on the territories with identical soil types, situated at a long distance from the city (50–120 km) and unaffected by industrial emissions.

Sulphur concentration in plant samples and in forest litter was determined using spectrophotometry after preliminary dry ashing of the material [16]. In soil samples, mobile sulphur in the salt extract (1 M KCl) was determined using this method [17]. The data were processed statistically using standard procedures including correlation and regression analyses. Data processing and graphic representation were carried out using Microsoft Excel, Mathcad 12 software.

## RESULTS AND DISCUSSION

Determination of sulphur content in the needles and leaves of trees in Irkutsk revealed its substantial increase on all the TG (Fig. 1). The amount of sulphur is especially large on TG No. 11 in the north-west part of the city, which is likely due to the fact that large plants and the power station of the northern industrial junction are situated there. The four in-

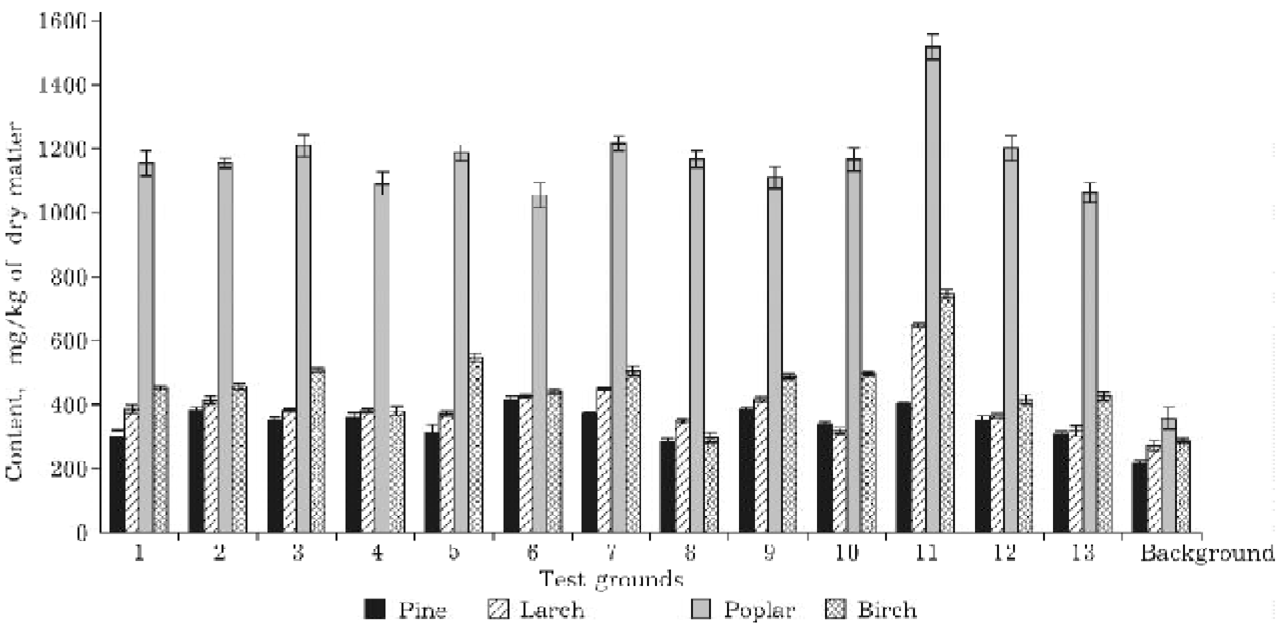


Fig. 1. Sulphur content in needles and leaves of trees at the territory of Irkutsk.

vestigated tree species possess different accumulating capacities with respect to industry-related sulphur. For instance, sulphur content in pine needles increases by a factor of 1.4–1.9, in larch needles by a factor of 1.2–1.4, in poplar leaves 3–4.3, birch leaves 1.2–1.9 times in comparison with the background level. So, poplar exhibits the highest sulphur-accumulating capacity. Results of calculations provide evidence that poplar leaves can accumulate up to 1500 mg of sulphur per 1 kg of the dry matter. The maximal accumulation of sulphur in pine and larch needles, as well as in birch leaves is lower and equals 300–700 mg/kg of the dry matter. Analysing the level of sulphur accumulation in different parts of the city we discovered that the highest sulphur concentrations in needles and leaves are detected in the central part of the city (TG Nos. 2, 3, 5), near roads (TG No. 6), in the north-west direction (TG Nos. 9, 11) and near the airport (TG No. 7). The results obtained by us agree with the data on atmospheric pollution with sulphur dioxide in Irkutsk [18, 19].

In the opinion or a number of researchers [6, 20, 21], an increase in sulphur content of pine needles even by 20–30 % of the background level is an evidence of air pollution with sulphur-containing compounds and degradation of tree stands. On the basis of the data obtained by us, we may speak of the substantial pollution of atmospheric air in Irkutsk and worsening of the life state of trees within the boundaries of the city. This is confirmed also by the analysis of the morphostructural parameters of trees. For instance, the level of defoliation of the crowns of coniferous trees is 60 %, deciduous trees 50 %; necrosis area in needles and leaves may reach 20 %, the lifetime of pine needles decreases to 1–3 years. In addition, the following parameters decrease: the length of needles of pine and larch (by a factor of 1.5–2), the number of needle pairs on sprouts (1.5–3 times), length of the two-year-old sprouts (3–5 times). Industry-caused changes of the morphostructural parameters of trees in the city are also evidenced by the high correlation coefficients ($r$ = 0.60–0.75, $P$ = 0.05, $n$ = 13) between these parameters and sulphur content in needles.

No reliable correlations between the morphostructural parameters of tree crowns and sulphur content in assimilating organs were detected at the background territories. Therefore, under the conditions of pollution of urban environment, sulphur-containing compounds of industrial emissions serve as a significant factor determining the life state of woody plants.

Investigating the concentration of the mobile forms of sulphur in urban soil we discovered that the high accumulation of sulphur-containing compounds is characteristic of all the genetic profiles in comparison with the background analogs (Table 1). Soil horizons, both at urban and background territories, are characterised by different depositing abilities with respect to this element. In the upper part of the soil profile, the main fraction of sulphur is concentrated in the humus-accumulating layer (Ad and A horizons), that is, sulphur is incorporat-

TABLE 1

Concentration of mobile sulphur in the genetic horizons of grey forest soil of the urban and background territories, mg/kg

| Statistical index | Indices of the horizons of soil profile | | | | | | |
|---|---|---|---|---|---|---|---|
| | Ad | A | AB | B | Bt,f | BC | C |
| | | | *Urban TG* | | | | |
| $\bar{X}$, lim | 20.8 / 10.7–36.6 | 14.2 / 5.2–29.5 | 8.8 / 3.6–20.6 | 8.0 / 2.3–14.6 | 12.6 / 3.3–17.5 | 8.9 / 3.7–15.7 | 6.5 / 2.6–11.2 |
| $V$, % | 37.5 | 47.6 | 66.6 | 56.1 | 46.4 | 35.0 | 51.5 |
| | | | *Background TG* | | | | |
| $\bar{X}$, lim | 6.3 / 4.2–10.1 | 1.8 / 1.1–2.9 | 0.7 / 0.3–1.2 | 1.2 / 0.2–1.7 | 3.3 / 2.3–4.3 | 1.3 / 0.3–1.9 | 0.3 / 0.2–0.3 |
| $V$, % | 52.8 | 55.9 | 41.1 | 57.2 | 27.1 | 69.3 | 5.8 |

ed in the organic matter. The obtained high concentrations of mobile sulphur in the upper horizons of urban soil are not typical for non-polluted grey forest soil and are due to the accumulation of industry-related sulphate ions. A gradual decrease in the level of sulphur is observed in the lower-lying soil horizons AB and B. However, sulphur content somewhat increases in horizons Bt,f and BC. This may be explained by the stronger adsorption of sulphate ions by mineral colloids which are prevailing in these horizons. An increase in sulphate ion binding in horizons Bt,f and BC of the soil profile may be also due to the presence of the oxides of iron and aluminium which are the major absorbents for sulphur [22, 23].

Estimating the limits of the variation of mobile sulphur concentration in soil horizons on the basis of variation coefficients $V$ (see Table 1) we may state that the grey forest soils of Irkutsk differ from each other substantially in the profile distribution of mobile sulphur. The results obtained provide evidence that in addition to the organic accumulation in horizons Ad and A, urban soils exhibit also pronounced granulometric concentrating of sulphur in horizons Bt,f and BC. This fact is due to the good water migration of sulphur and to the presence of adsorption barriers against sulphur in the profile of these soils. Such regularity in the redistribution of mobile sulphur over the soil profile of grey forest soil was discovered by us also at the background territories. However, natural soil exhibits smaller accumulation of sulphur in the upper horizons Ad (3.5 times lower than the level in urban soil), especially in horizons A (8 times lower). In the AB horizons of urban soil, high concentrations of sulphur-containing compounds are observed (exceeding the background level by a factor of 12.5), along with pronounced redistribution of sulphate ion into the texture horizons Bt,f. In addition, the high sulphur content in the soil-forming horizons C of urban soil (exceeding the background level by a factor of 24) can be an evidence of its active filtration from the upper part of soil profile into the lower-lying horizons.

An integral part of the biogeochemical redistribution of sulphur in the ecosystem is its return on the surface of soil with leaf litter. At the territories under industrial pollution, an increase in sulphur content in needles and leaves leads to its accumulation in dead leaves [3]. Therefore, an increase in sulphur concentration in dead leaves and then in forest litter can serve as a substantial additional source of this element entering the soil. Studying the sulphur content of forest litter in the soil in Irkutsk we detected an increase in its concentration by a factor of 2–6 over the background level. The maximal levels were detected in the north-western (TG 10, TG 11) and central (TG Nos. 2–4) parts of the city: sulphur concentration was 1100–1500 and up to 800–900 mg/kg of dry matter, respectively.

A highly reliable correlation ($r = 0.91$, $P = 0.05$, $n = 15$) was revealed between sulphur content in forest litter and in the upper organics-accumulating soil horizons Ad (Fig. 2). This dependence provides evidence of the substantial income of industry-related sulphur with the emissions from plants and motor transport on soil surface, and also an evidence of further fixation of sulphur-containing compounds by the organic substance. In addition, a correlation between sulphur content in forest litter and in texture horizons Bt,f was revealed ($r = 0.93$, $P = 0.05$, $n = 15$). The results obtained point to the active redistribution of mobile sulphur from forest litter and upper horizons downward the soil profile and subsequent accumulation in the texture sorption barrier (Bt,f) where sulphur compounds are fixed by the uliginous fraction of soil.

One of the determinative factors of sulphur accumulation and migration in the soil profile system is the acidity of the soil solution [3, 23]. The negative effect of sulphur on soil is exhibited mainly as the acidification of soil solution. Quite contrary, soil alkalisation creates favourable conditions for neutralization of sulphate ions [22]. So, positive correlations should exist between the acidity of soil solution (within the range pH $< 7$) and the concentration of mobile sulphur in soil profile. However, it was detected in out experiments that soil acidification causes a substantial decrease in sulphur content, while an increase in its concentration in forest litter and along the whole depth of the soil profile is observed when the soil solution pH shifts to the neutral and weakly alkaline side. No regularities of this kind were revealed under the background conditions: for the case

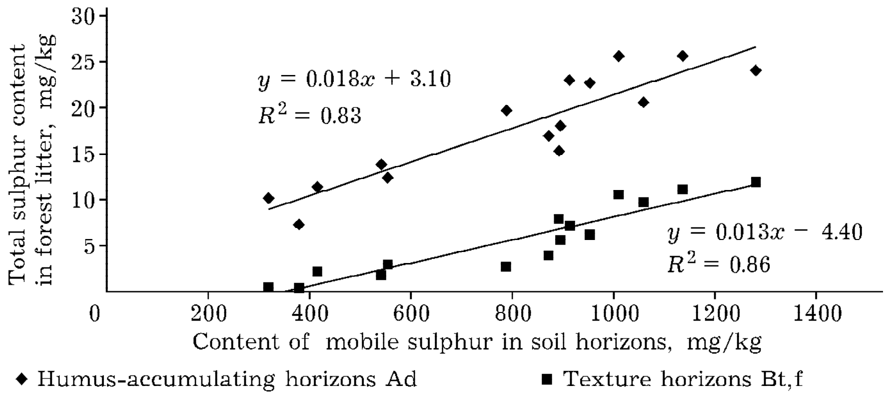


Fig. 2. Dependence between total sulphur content in forest litter and the mobile form of sulphur in soil horizons.

of weakly acidic pH of soil solution in the upper humus-accumulating horizons (Ad and A) the concentration of sulphate ion here decreases by a factor of 8–27 in comparison with its level in urban soil.

A trend to alkalisation of soil and forest litter in Irkutsk is most probably due to the action of emissions of heat and power plants and boiler houses using coal of the lowest quality. It is known that unscreened coal is used in the city heating systems; much smoke is formed in the combustion of this coal; large amounts of soot and ash are also formed [10]. Because of this, alkaline components of emissions including also various calcium forms are likely to promote the formation of complex organic and mineral compounds with sulphur in soil. In addition, it is known that calcium carbonates serve as a geochemical barrier for the accumulation of mobile sulphur forms in soil binding them with the formation of poorly soluble gypsum [22].

Since calcium plays a part of the dominant cation of the soil-absorbing complex determining the alkaline reaction of soil [23], we considered a connection between the accumulation of this element and sulphur in the soil profile (Fig. 3). The maximal content of sulphur and calcium was detected in the organics-accumulating and texture horizons, which is due to the presence of humic acids and a fine uliginous fraction with the high adsorption ability. No reliable correlation between the concentrations of mobile sulphur and calcium was revealed for background soil. Therefore, accumulation of these elements in the exchangeable and complex forms in natural soil is less clearly expressed than in the soil of an urban ecosystem.

The high content of industry-related sulphur in soil causes more active intake of its oxidized forms into plants [24]. It was established for herbaceous plants that for the case of simultaneous pollution of air and soil cover sulphur is more actively absorbed by plants from the soil but not from the air [3]. A comparison of our data on sulphur content in soil with that in needles and leaves of trees at the territory of Irkutsk demonstrated the occurrence of positive reliable correlations between these parameters for different soil horizons (Table 2).

The closest correlations were revealed for forest litter, humus-accumulating horizons Ad and texture horizons Bt,f. Less close correlations were revealed for the soil-forming horizons (BC and C), which is an evidence of a decrease in the arrival of mobile sulphur into plants from the lower soil horizons. A lower conjugation of correlation is characteristic of horizons A, AB and B. In a number of cases, the level of correlation is uncertain, which is likely to be due to the active migration of mobile sulphur from

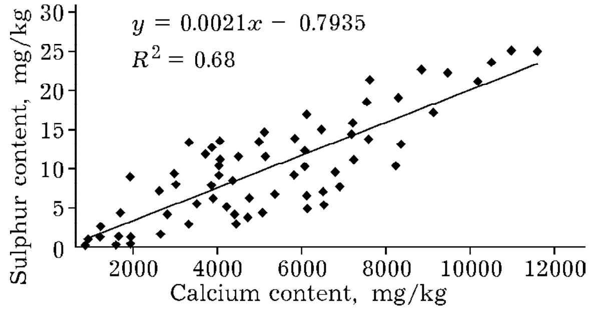


Fig. 3. Dependence between the concentration of mobile sulphur and calcium forms in the genetic profile of grey forest soil of Irkutsk.

TABLE 2

Significant coefficients of correlation between sulphur content in soil horizons and needles (leaves) of woody plants ($P$ = 0.05, $n$ = 16)

| Object | Indices of soil profile horizons | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | O | Ad | A | AB | B | Bt,f | BC | C |
| Pine needles | 0.86 | 0.82 | 0.58 | – | 0.50 | 0.75 | 0.65 | 0.55 |
| Larch needles | 0.68 | 0.70 | 0.48 | – | – | 0.65 | 0.50 | 0.45 |
| Poplar leaves | 0.77 | 0.65 | 0.60 | 0.45 | 0.55 | 0.70 | 0.60 | 0.50 |
| Birch leaves | 0.87 | 0.60 | 0.55 | 0.50 | 0.60 | 0.80 | 0.70 | 0.60 |

these horizons into lower-lying ones. In general, the data obtained on the profile-genetic redistribution of sulphur depict the possibility of its active absorption and accumulation by woody plants from the polluted soil of urban territories.

## CONCLUSIONS

As a result of the investigation of soil and woody plants, we demonstrated a substantial pollution of the urban ecosystem of Irkutsk with sulphur-containing compounds of industrial origin. The accumulation of sulphur in needles and leaves of woody plants at the urban territory exceeds the background level by a factor of 1.7–3.2. Accumulation of industry-related sulphur differs substantially for different tree species. The highest level of accumulation is characteristic of the leaves of poplar, a lower one is observed for pine and larch needles, as well as for birch leaves. A dependence between sulphur concentration in needles and the changes of a number of morphostructural characteristics of trees was revealed (with the correlation coefficients at a level of 0.60–0.75), which is an evidence of the anthropogenic nature of the disturbance of the living status of woody plants in the urban environments. Investigation of the mobile sulphur content in the soil profile revealed a high level of sulphate ion accumulation for all the soil horizons. The clearly pronounced industry-related pollution of urban soil is evidenced by the high sulphur concentration in forest litter (O), upper soil horizons (Ad and A), and at the depth of texture horizons (Bt,f). An increase in the level of mobile sulphur with a shift of the acidity of soil solution towards alkalisation was detected, which is untypical for grey forest soil; this is expressed most clearly near roads and in the central part of the city which is the most heavily polluted one. It is shown that sulphur accumulation at different depth of the soil profile is affected by such soil characteristics as the content of organic matter, uliginous fraction, mobile calcium, and the acidity of soil solution. It may be concluded on the basis of the data obtained that the soil cover under urban conditions acts as the main accumulating barrier for sulphur compounds and to a large extent determines the degree of their accumulation in the assimilation organs of plants. This is confirmed by the occurrence of positive correlation between the accumulation of sulphur in forest litter, humus-accumulating, texture, soil-forming horizons of soil and the level of sulphur in the assimilation organs of plants. The results of investigations allow us to state that sulphur content in needles and leaves of trees and in the urban soil cover serves as an adequate bioindicator criterion fort he integral evaluation of the pollution of urban ecosystems.

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