Original Russian Text © 2020 V.V. Ershov published in Forest Science Issues Vol. 3, No. 2, pp. 1-34

DOI 10.31509/2658-607x-20214125

MONITORING OF ATMOSPHERIC AND SOIL WATER COMPOSITION IN FOREST ECOSYSTEMS: MILESTONES AND OUTLOOK

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Received 20.04.2020
Accepted 28.05.2020

The article provides an overview of Russian and foreign studies on assessment of the composition of atmospheric fallout and soil waters in forest ecosystems. The conclusion was made that little attention is given to the transformation of precipitation chemical composition by forest ecosystems, taking into account the influence of the species composition of the stand and the mosaic structure of the biogeocenosis (areas below the crowns, between the crowns, and open sites). European studies usually look at long-term development of the composition of atmospheric fallout and lysimetric waters, detecting long-term trends in composition changes of atmospheric and soil waters and identifying the factors driving these changes. In Russia, no such long-term (lasting for more than 10 years) continuous observations on the effect of man-made pollution on the composition and properties of atmospheric and soil waters were carried out. This task is very relevant for Russia and, especially, for its industrial regions.

Key words: *forest biogeocenoses, air pollution, atmospheric fallout, soil waters, monitoring, long-term dynamics, critical loads*

Environmental conditions have changed rapidly on a global scale since the beginning of the industrial era (Schöpp et. al., 2003; IPCC, 2007). Technical advances of mankind have led to a significant change in the chemical composition of the atmosphere, hydrosphere, pedosphere, as well as flora and fauna. Production activities cause the transformation of natural ecosystems, the degree of such transformation depending on the nature and intensity of anthropogenic impact and the stability of ecosystems. Assessment of the current state of ecosystems and forecasting of their development is an important scientific task and has great applied significance (Anan'eva et al., 2012). Forest ecosystems play an important role in maintaining ecological balance, sustainable functioning of the biosphere, as well as in providing for the material and spiritual needs of humans (Smit, 1985). Air pollution is one of important factors determining the current state of forest ecosystems. Forest biogeocenoses accumulate pollutants, acting as a kind of transformer of atmospheric technogenic fallout and, in fact, being a filter at a biosphere level in a large biogenic flow of matter (Rassejannye jelementy..., 2004). By absorbing pollutants of anthropogenic and natural origin, forests purify the atmosphere and thus perform an important sanitary function.

The **objective** of the work is to analyze the literature data on the impact of air pollution on forest biogeocenoses.

At the end of the last century, humanity already faced an acute problem of protecting the environment from anthropogenic impact, and there was an impending threat of forest ecosystem degradation from pollution. An objective analysis of this hazard requires assessments based on quantitative measurements and broad-scale generalizations. There are 3 classes of interactions between forest ecosystems and atmospheric pollutants. Interactions of class 1 are characterized by a low content of pollutants; their impact on the forest ecosystem, depending on their nature, is either insignificant (harmless) or stimulating. With interactions of class 2, concentrations of pollutants have a depressing effect on some components of the biota. With class 3 interactions, there is a high content of pollutants that cause disturbances of biogeochemical cycles and energy flows (Smit, 1985).

From the end of the 19th century to the mid-80s of the 20th century, large regions of Europe were being negatively affected by acid precipitation due to an increase in anthropogenic emissions of pollutants into the atmosphere. Sulfur (S) emissions in Europe increased sevenfold, and nitrogen (N) emissions increased approximately fivefold, which correlated with a similar increase in acidity of precipitation (Schöpp et. al., 2003; Engardt et al., 2017). In 1980s, the state and viability of forest ecosystems caused public concern due to extensive damage to the forests of Central Europe, which was caused by air pollution, including pollution due to acid-forming compounds that fall with atmospheric precipitation (Schütt et al., 1983; Lammel, 1984). Extensive research to gain a deeper understanding of the mechanisms of atmospheric pollution impact on forest ecosystems was made in the 1980s and 1990s, which included continuous monitoring of ecosystems and modelling. On the basis of these studies, the following effects of air pollution on the state of forest ecosystems were identified (De Vries et al., 2000): elevated concentrations of SO_2 in the atmosphere cause stomata disorders and premature leaf senescence, impaired distribution of assimilates resulting in the weakening of the root system, accelerated leaching of nutrients from plants; eutrophication due to deposition of nitrogen compounds (NO_3, NH_4), causes an imbalance between base cations and nitrogen, which can lead to increased stress from drought and sensitivity to frost and fungal diseases, as increased nitrogen content contributes to the growth of above-ground biomass, while the rate of root growth is not changed; soil acidification, i.e. the decrease in pH, leads to loss of base cations from the soil, causing nutrient deficiency (especially that of Mg), release of toxic aluminium compounds that negatively affect root growth, and increased migratory activity of heavy metals.

The effects of industrial air pollution caused public concern and, under the umbrella of the United Nations Economic Commission for Europe (UN-ECE) and the EU, the International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) was established in the mid-1980s (Lorenz, 1995). The main objective of Level I Monitoring Programme was to obtain data on spatial and temporal changes in the state of forest ecosystems, as well as on the impact of stress factors, including atmospheric pollution, on these ecosystems. Over time, it became obvious that large-scale monitoring was unable to clarify causal relationships, so in 1994, under the umbrella of UN-ECE/ICP Forest and the EU, a pan-European programme of intensive and continuous monitoring, so-called Level II Monitoring Programme, was launched. Later, both these monitoring networks became known as ICP Forests Level I and Level II (Lorenz and Becher, 2012). While Level I ICP Forests network is aimed at obtaining data on spatial and temporal changes affecting the state of the forest, Level II network is designed to clarify cause-and-effect relationships. Indicators measured as part of ICP Forests include: the state of tree crowns (degree of defoliation and discoloration); tree trunk growth; ground cover biodiversity; chemical composition of needles/leaves, soil, atmospheric fallout, and soil waters; meteorological data, ambient air quality, data on the biomass of ground vegetation. All these observations are carried out at specially selected sites of continuous observation (Ferretti et. al., 2013).

The global monitoring network created in Europe not only helped to assess the impact of air pollution on ecosystems, but also allowed for measures to reduce the content of acidifying and eutrophying element compounds through a number of protocols. Such protocols established acceptable limits for emissions of certain pollutants such as NH_3, NO_x, and SO_2 (Maas, Grennfelt, 2016). According to European scientists, long-term continuous observations are necessary for the successful implementation of measures aimed at reducing the impact of air pollution on ecosystems (Ferm et al., 2019).

Impact of man-made ecosystem pollution is a relevant problem for Russia, especially for the industrial regions of the north, such as Krasnoyarsk Krai and Sverdlovsk, Chelyabinsk, and Murmansk Oblasts, where large smelters operate (Evdokimova et al., 1984; Jarmishko, 1984; Karaban' et al., 1985; Jarmishko, Jarmishko, 2002; Derome, Lukina, 2010; Cvetkov, Cvetkov 2012; Derjabina, Peretykin, 2015; Zajceva, 2016; Kurpatov et al., 2016; Vorobejchik, Kajgorodova, 2017; Kljuev, Jakovenko, 2018; Shepel', 2019, etc.).

Chemical composition of atmospheric fallout in the form of snow and rain: the effect of air pollution and forest stand

Atmospheric precipitation is one of the components of the biogeochemical cycle. Atmospheric fallout is divided into wet (substances that reach the earth surface with actual precipitation: rain, snow, frost, fog, dew, etc.) and dry (particles and gases). The amount and quality of atmospheric precipitation has a huge impact on the state of forests. When atmospheric precipitation interacts with tree crowns, both the absorption of compounds of various elements and their leaching from the crowns occur. Substances leached from the crowns of woody plants can be absorbed by plant roots, and during the growing season this process can occur repeatedly (Lukina, Nikonov, 1996; Kislotnye osadki..., 1990).

Fallout in the form of snow

Winter atmospheric fallout is associated with the period of biological dormancy. In boreal forests, the duration of snow cover is 100–200 days per year, which determines the significant role of snow precipitation in biogeochemical cycles (Rassejannye jelementy..., 2004). During thaws, substances leach from tree crowns, significantly affecting the spatial distribution of elements in snow (Helmisaari, Malkonen, 1989; Lukina, Nikonov, 1996; Pomeroy et al, 1999; Lukina, Nikonov, 2003; Pristova, 2005; Gandois et al., 2010; Martynjuk et al., 2011; De Vries et al., 2014, etc.). Snow falling on the earth surface forms a snow cover, and during its formation it accumulates, in addition to aerosol fallout that reflects the natural composition of the atmosphere, the products of man-made emissions, which build up in the snow mass as part of solid hydrometeors (snow, silver thaw, frost) (Tentjukov, 2007). Snow cover acts as one of the indicators of pollution not only of atmospheric fallout, but also of atmospheric air, as well as subsequent pollution of waters and soils (Voevodova, 1979; Ratkin, 2000).

In Switzerland, a multi-year assessment of the composition of atmospheric throughfall and fallout at open sites was carried out in 1995–2001. It revealed the exceedance of nitrogen critical loads on ecosystems (Thimonier et. al., 2005; Waldner et. al., 2007). In Western Europe, 169 intensive monitoring sites were studied (in 1996–2001) looking at the composition of atmospheric fallout and the effect of precipitation on forest ecosystems. This revealed a significant decrease in the content of sulfates in atmospheric fallout, both at open sites and of the throughfall, while the nitrogen content decreased less significantly (Fisher et. al., 2007). Joint studies of the effect of atmospheric fallout on vegetation in monitoring networks in 1996–2000 revealed a statistically significant shift towards nitrophilic species at high content of nitrogen and towards acidophilic species at high content of sulfur compounds in atmospheric precipitation (van Dobben, de Vries, 2010). Studies of the long-term development (1995–2007) of changes in the composition of atmospheric fallout in France showed a decrease in the concentrations of sulfur compounds and increased pH. According to the authors, this may be due to a reduction in sulfur dioxide emissions in Europe, but despite the reduced emissions, concentrations of nitrogen compounds remained unchanged or even increased in some areas (Pascaud et. al., 2016).

In Poland, studies were conducted in coniferous and deciduous forests to assess the transformation of atmospheric throughfall, as well as to assess the influence of the species composition of stands on the chemical composition of atmospheric fallout. These studies were conducted from January to December 2010. The data about two periods was analyzed: winter-spring (from January to May and December) and summer-autumn (from June to November). The authors report that in winter there is usually an increase in the level of air pollution. During the low temperature season, the influence of pollutants, especially those of anthropogenic origin, on the composition of atmospheric throughfall and fallout at open sites was assessed. It was found that concentrations of Cl^-, SO_4^{2-}, Al, and Zn contained in winter atmospheric fallout increase when precipitation passes through the canopy of coniferous trees. This was accounted for the fact that the stationary area of research was located near the region with the highest population density in the country and with developed industry. Ion concentrations in throughfall of coniferous trees are usually higher than below the crowns of deciduous tree species in a similar habitat due to the higher filtration efficiency of dry sediments, especially acidic ions. It is also reported that the concentrations of such elements as Mn, K, Mg, and PO_4^{2-} in the throughfall of coniferous and deciduous forests are comparable, which is evidence of low biological activity of trees in winter (Kowalska et. al., 2016).

There are lots of publications made by Russian researchers on the study of chemical composition of snow cover. The role of snow cover in natural processes has been investigated since the 20th century (Voejkov, 1949). Data on its physical and mechanical properties, the processes of its formation and melting, etc. are analyzed in the works of I.D. Kopanev (Kopanev, 1982). Transboundary transport of sulfates and surface water pollution have been assessed since the late 20th century (Glazovskij et al., 1983; Vasilenko et al., 1985; Obolkin, 1991). A considerable part of the Russian and foreign works was carried out on the territory of Murmansk Oblast, which borders the Nordic countries.

On the Kola Peninsula, in the zone of influence of the Copper-Nickel Smelter, the composition of atmospheric fallout in coniferous forests was studied (Lukina, Nikonov, 1996). The authors observed that winter atmospheric fallout is characterized by reduced mineralization and carbon concentration as compared to summer fallout. This is due to the low activity of organisms in the winter. Differences in the composition of snow waters below and between the crowns are smaller than during the growing season. When approaching the source of pollution, there is a statistically significant increase in the concentrations of nickel, copper, iron, chlorine, nitrates, sulfates, ammonium and calcium in the composition of snow waters, as well as increase of snow water acidity.

Studying the composition of snow deposition as an indicator of environmental air pollution in the zone of influence of *Pechenganickel* smelter, allowed calculating the boundaries of the local high level of snow cover pollution. A method was developed for calculating total retrospective and prospective ("dry "and" wet" precipitation) air pollution loads (for the period of maximum snow accumulation) on any selected natural-territorial complexes (Ratkin et al., 1998). This method made it possible to determine the boundaries of the local high level of pollution for sulfates at 58–60 km, and for nickel and copper at 28–30 km. It was found that the leachability of nickel from the atmosphere is 10% higher with snow than with rain, while for copper this parameter is 30% higher than for nickel. Terrain, climatic conditions, as well as features of the pollutant and the source contribute to unequal air pollution load on ecosystems in the zone of local pollution (Ratkin, 2000).

The composition and properties of snow depositions at different stages of pine and spruce forest degradation on the Kola Peninsula in 1991–1997 were evaluated. In the background spruce forests, there is an enrichment of precipitation below the crowns in winter, while the total amount of substances entering the spaces between the crowns is higher than below the crowns, which is due to less precipitation entering the wood parcels. In pine forests, concentration of pollutants is less significant, and spatial differences are less pronounced than in spruce forests. In defoliating forests, concentrations of pollutants are significantly increased as compared to the background, and there is increased intake of nutrients (K, P, NH_4, Mn, and Zn) under the tree canopy, which is due to increased amount of snow throughfall because of their sparseness both in spruce and pine forests. In pollution-induced sparse forests, as well as in defoliating forests, concentrations of the major pollutants (copper, nickel and sulfur) are considerably increased as compared to the background, with a rather even distribution of fallout over the area, which is also due to the strong thinning of crowns and the death of most trees in pine and spruce forests (Rassejannye jelementy..., 2004).

On the Kola Peninsula, assessment of the chemical composition of atmospheric fallout at a distance of 11 km from the *Severonickel* smelter was carried out in 2005–2011. (Kashulina et al., 2014). The study showed that during the study period, concentrations of SO_4^{2-} in the snow waters were 4 times higher than the background levels, whereas the concentrations of Ca and Mg were 5–6 times higher than the background levels. The content of heavy metals in the snow near the smelter was 2,500 times (for Ni), 1,500 times (for Cu) and 400 times (for Co) higher than the background values. It is stated that the main source of snow cover pollution is highly concentrated technogenic dust from low-lying sources (windows, doors, ventilation exhausts, transportation of ore and ore concentrate). It was also found that the concentrations of copper and nickel in snow waters had remained virtually unchanged since 1994, which is due to a significant amount of technogenic dust, despite the gradual reduction of emissions by the smelter (Kashulina et al., 2014).

In the Komi Republic in 1996–1998 and 2005–2007, studies of the chemical composition of snow cover in forest ecosystems in the zone of influence of a pulp and paper plant (PPP) were conducted. The research showed that the snow deposition of the taiga landscapes under study had acidic and slightly acidic reaction. Forest stands usually have an acidifying effect. There are increased concentrations of potassium, sodium, manganese and sulfates, as well as decreased content of cadmium, nickel and nitrates in the snow cover below the crowns, as compared to the snow cover between the crowns. PPP emissions have a significant impact on the content of C_{org}, HCO_3^-, SO_4^{2-}, Na^+, Ca^{2+}, Mn, Ni, and Fe in snow deposition. Concentrations of the majority of the determined components of the snow cover of forest stands are higher than in the background area (Pristova, Vasilevich, 2010).

In the forest ecosystems of Moscow, Vologda and Kostroma Oblasts, studies on the composition and spatial distribution of atmospheric fallout of mineral nitrogen were conducted (2013–2014). The authors conclude that the concentration of mineral nitrogen in atmospheric fallout is mainly influenced by the nature of emissions and specialization of industrial enterprises and the increase in nitrate emissions is also associated with vehicle emissions. It was found that the concentration of mineral nitrogen in the forest snow cover is determined primarily by the intensity of anthropogenic emissions, and not by the type of stands, and that stands significantly increase the concentration of mineral nitrogen (ammonium and nitrate) in snow waters (Kudrevatyh, 2017).

Thus, the study of snow chemical composition and its impact on forest ecosystems is covered in many works of foreign authors and a number of Russian publications. Many foreign authors study snow deposition not separately, but in combination with rain, analyzing the total amount of precipitation per year or by season: December–February (winter), March–May (spring), June–August (summer) and September–November (autumn). However, foreign studies rarely pay attention to the influence of the micromosaic structure of the biogeocenosis (spaces below and between the crowns) on the composition of atmospheric fallout. These studies distinguish precipitation below the forest canopy and in treeless areas. At the same time, in foreign studies, the composition of snow waters is usually studied in its long-term development, whereas in Russian studies, such long-term (for more than 10 years) continuous observations are quite rare (Ershov et al., 2016). Using the example of Murmansk Oblast forests, analysis of the variability of the composition of snow waters in pine and spruce forests at different stages of pollution-induced degradation was carried out taking into account the long-term (for about 20 years) development and micromosaic structure of the forest cover. It was shown that the snow waters below the crowns of spruce and pine forests are richer in element compounds than that between the crowns, which is due to their washing out and leaching from the crowns of woody plants. Snow waters of spruce forests have high concentrations of element compounds as compared to pine forests, which is due to the sorption capacity of spruce crowns. Long-term development of the snow composition demonstrates decreased content of the major pollutants in the background setting and in defoliating forests, both below and between the crowns, which is associated with a decrease in emissions. It is shown that the main factors of the dynamics of the snow water composition in the Kola Peninsula forests are edificator woody plants as well as industrial air pollution.

Fallout in the form of rain

Fallout in the form of rain also plays an important role in biogeochemical cycles in forest ecosystems. The snowless period (rains) is divided into two fundamentally different parts: the growing season and the periods of relative biological dormancy (spring and autumn). Atmospheric fallout in the form of rain has the most pronounced interaction with the tree canopy, especially during the growing season. During the snowless period, the concentration of substances in atmospheric precipitation significantly increases as compared to the winter period. It is associated with the active functioning of the forest biogeocenosis (Rassejannye jelementy..., 2004). In the spaces between the crowns, atmospheric fallout is mainly of atmogenic nature, i.e., the composition of fallout is determined by atmospheric precipitation, whereas in wood parcels they are of autogenic (biogenic) character (Nikonov, Lukina, 2000; Lukina et al., 2008). When rain waters interact with the forest canopy, physical and chemical reactions occur that can change the acidity and concentration of most of the elements contained in these waters (Karpachevskij, 1981; Medvedev et al., 1986; Matzer, Meiwes, 1994; Shil'cova, 2006). The paper by A. Thimonier (Thimonier, 1998) reveals that the study of the composition and properties of atmospheric fallout in the form of rain, both at open sites and for the throughfall, provides valuable information about the content of chemicals in atmospheric precipitation that cause acidification or eutrophication.

In the north-west of Germany, studies were conducted on the chemical composition of throughfall and rain waters at open sites in oak, birch and pine forest types. It was shown that the concentration of chemical compounds in throughfall is significantly affected by both the species composition of the stand and the distance from the source of pollution. Besides, differences in the deposition rate of ammonium, nitrogen, potassium and iron were found, which are explained by different tree crown projections, and the deposition rate of calcium, magnesium, manganese and zinc depends mainly on the tree type, since the leaching of these cations is higher in deciduous trees as compared to coniferous trees. Leaching of elements from the crowns of deciduous trees and enrichment of atmospheric precipitation with these elements is particularly pronounced in such tree species as *Betula pubescens* and *Betula pendula*. The level of cation leaching is highest during the growing season, resulting in higher throughfall concentrations of elements in spring, summer, and early autumn (Herrmann et. al., 2006).

Studies of dry and wet atmospheric fallout in Italy have shown that forest cover sorbs SO_4^{2-}, NO_3^-, and Na^+ more effectively than treeless areas do. The amount of rainfall below the crowns was lower than at open sites. Additionally, it is noted that the interception of rain deposition by the forest canopy depends on the type of vegetation. The difference between throughfall and precipitation at open sites for deciduous trees is 10–15%, whereas in coniferous forests the difference reaches 23–24% (Balestrini et. al., 2007).

Studies of transformation of atmospheric precipitation by deciduous and coniferous stands in Poland showed that intensive processes of ion exchange between the crown and rain during the growing season lead to increased cation deposition (K, Ca, and Mg) both in deciduous and coniferous forests. Species composition of the stand significantly affects the deposition, the composition of precipitation and depends on the region of the country. Coniferous species (pine and spruce) acidify the precipitation, while deciduous species (oak and beech) increase the pH. Higher deposition of K, Mg, and Mn ions is observed in the throughfall of pine and oak stands as compared to beech and spruce (Kowalska et. al., 2016).

In the Mediterranean forest ecosystems of the eastern Adriatic coast, atmospheric precipitation composition and tree viability indicators were assessed from April 2017 to December 2018. The findings showed that nitrogen deposition with the throughfall of pine forests (*Pinus halepensis* and *Pinus nigra*) is higher compared to open sites, whereas in oak forests (*Quercus pubescens, Quercus ilex*) there is no such trend. The lowest nitrogen concentrations were found in pine forests and the highest – in oak forests (Jakovljevic et. al., 2019).

Transformation of the composition and properties of atmospheric fallout in the form of rain by a tree canopy was also studied by Russian scientists (P'javchenko, Sibereva, 1959; Pozdnjakov, 1956; Morozova, Kulikova, 1974; Uchvatov, Glazovskij, 1984; Yelpatyevsky, 1993; Hrustaleva, 2002, etc.). Throughfall not only washes the settled dust away from the leaves, but also is saturated with metabolic products of plants and other organisms, and leaches some elements from living cells, actively affecting the biological cycle of substances. Many researchers have reported the acidification of rain waters by organic and mineral acids through the contact with the tree canopy, which contributes both to the transfer of chemical compounds into an accessible to plant roots form, and an increase in the concentrations of many chemical elements in the deposition. The highest content of elements and element compounds such as nitrogen, hydrogen carbonate, sulfur, sulfates, potassium, sodium, zinc and copper is found specifically in below-the-crown precipitation (Medvedev et al., 1986; Nikonov, Lukina, 2000, Marunich et al., 2006; Archegova, Kuznecova, 2011; Robakidze et al., 2013, etc.).

The composition of atmospheric fallout in coniferous forests of the Kola Peninsula was studied, taking into account the influence of air pollution (Lukina, Nikonov, 1996). The authors note that it is during the growing season that the majority of heavy metals and acid-forming substances falls. The composition of atmospheric fallout in forest ecosystems is determined by the degree of defoliation and tree density, whereas acidity is determined by the intensity of element leaching from tree crowns. When approaching the smelter, there is a significant increase in the content of elements that are part of the emissions. More acidic than in the background setting, atmospheric precipitation in defoliating forests contributes to leaching of the base cations, which explains their higher concentrations in rain waters below the crowns. In pollution-induced sparse forests, there is an increase in the acidity of rain precipitation, as well as a decrease in the concentrations of Ca, Mg, K, Mn and NH_4^+ due to the tree canopy absence.

On the Kola Peninsula, the nature of transformation of atmospheric fallout by the two tree species dominant in boreal forests, i.e., spruce and pine, is similar, and the throughfall becomes acidic and is enriched with chemical elements. However, the degree of transformation is different: spruce performs a deeper transformation than pine, which is especially pronounced in precipitation in the form of rain. Deeper transformation is due to the presence of a dense and extended spruce crown as compared to that of pine (Kislotnye osadki..., 1999).

Assessment of the composition and properties of rain deposition at different stages of pine and spruce biogeocenoses degradation on the Kola Peninsula carried out in 1991–1997 showed that during the snowless period, the concentrations of substances in atmospheric fallout in the background area increase significantly as compared to the period of biological dormancy. The composition of rain deposition was dominated by carbon; by calcium and ammonium among cations, and by sulfates among anions. In defoliating pine and spruce forests there is a sharp increase in sulfate, nickel and copper concentrations in the rain due to the influence of air pollution on tree canopy; moreover, increased concentrations of calcium, manganese and magnesium are observed, which is due to leaching of these elements from crowns by acidic precipitation. In spruce and pine pollution-induced sparse forests, the changes in the composition of rain deposition detected in defoliating forests persist and get worse. In spruce forests, there are no parcel differences in the element intake due to the strong destruction of the tree layer; in pine forests, parcel differences remain, and the number of elements entering the spaces below the crowns is higher than that between the crowns (Rassejannye jelementy..., 2004).

In the zone of influence of the *Severonickel* Copper-Nickel Smelter, other, two-year (2001–2002), studies of the effect of mining and metallurgical industry emissions on the chemical composition of atmospheric fallout in the form of rain were conducted. For rain deposition sampling, 6 plots at different distances from the smelter (1–17 km) were selected, taking into account prevailing and dominant winds. The authors note that with the emissions of mining and metallurgical industry, predominantly sulfates, as well as copper and nickel, enter the ambient air of Murmansk Oblast; these pollutants resulted in the formation of extensive zones of soil and vegetation cover degradation in the vicinity of *Severonickel* smelter. Chemical composition of the rain deposition of Monchegorsk landfill is extremely variable in space and over time, and pH values vary in a wide range from 4 to 7. Almost all chemical elements that make up the atmospheric emissions of the smelter do not show maximum concentrations in the proximity thereto; maximum concentrations are found at a distance of 5–10 km. Average concentrations of Ni, Cu, Zn, Mo, V, and Ag in atmospheric fallout exceed the $MAC_{fishery\ waters}$ (Dauval'ter et al., 2009).

G. M. Kashulina (Kashulina et al., 2014) conducted studies of the atmospheric fallout composition on the Kola Peninsula, near *Severonickel* smelter from 2005 to 2011. In the rain waters near the smelting plant, SO_4^{2-} concentrations were 4 times higher, and the concentrations of the base cations (Ca and Mg) were 5–6 times higher than the background levels. The content of heavy metals in the rain was higher than in the background by an order of magnitude or more: for Ni it was 146 times, for Cu – 80 times and for Co – 50 times higher than in the background area. It was reported that the dominant source of rain waters pollution were filtered gas and dust emissions and chimneys, and the reduction of emissions by the smelter itself had significantly reduced the amount of pollutants entering the rain waters from the pipes.

During the restoration of middle-taiga forests at a distance of 17 km from the city of Syktyvkar performed in 2006–2008, an assessment was made regarding the influence of woody plants on the chemical composition of atmospheric precipitation in the form of rain. It showed that the transformation of rain deposition was determined by a woody edificator plant. Increased concentrations of biogene elements were observed from spring towards summer and autumn. Composition of throughfall of deciduous and coniferous woody plants differs, especially in terms of content of organic carbon, potassium and calcium. Between the crowns, as compared to the areas below the crowns, concentrations of elements of biogenic origin are lower, and their fluctuations are more pronounced during the growing season. Rain waters collected below herbaceous vegetation demonstrated a lower concentration of potassium, calcium and magnesium than the waters below under woody plants (Archegova, Kuznecova, 2011).

Studies conducted in 2011 in the area of influence of the Mari sand-lime brick plant and in 2012–2014 in the Bolshaya Kokshaga nature reserve consider the effect of the aerial intake of substances on their circulation in forest ecosystems. To assess the aerial intake of chemical elements and their transformation during interaction with tree crowns, a *fabric dressings method* was developed. It complements the existing methods and makes it possible to evaluate plant exometabolites based on their involvement in the cycle. The amount and composition of atmospheric fallout, both rain and snow, vary significantly in space and time. Interaction with the tree canopy significantly changes their composition, and the degree of precipitation transformation depends on the type of woody plants, growing conditions and the phase of seasonal development. It is noted that leaching of calcium and strontium from the canopy of birch and pine forests growing on poor sandy soils, where there is an acute deficit of mineral nutrition elements, is much stronger than in the floodplain stand, where there is no deficit. Therefore, it was concluded that trees are able to control their mineral nutrition and biological cycle in forest ecosystems by releasing through the crown and trunk the necessary exometabolites. The composition and concentration of such exometabolites depend on the species composition of the stand and environmental conditions (Demakov, Isaev, 2015).

Thus, many Russian and foreign studies have been devoted to chemical composition of rain waters and its impact on forest ecosystems. Special attention is paid to the transformation of the precipitation chemical composition by forest ecosystems, taking into account the influence of the species composition of the stand. Foreign studies pay little attention to the influence of the micromosaic structure of the biogeocenosis on the composition of rain deposition. In European studies, the composition of rain waters has been usually studied in its long-term development, both as part of the ICP Forests program from 1995 to the present, and in other studies, for example, in a Swedish study of precipitation of sulfates, nitrates, ammonium, and other compounds, conducted from 1955 to 2017 (Ferm et al., 2019).

Consequently, long-term studies make it possible to detect long-term trends in changes in rain composition and identify the factors of these changes. However, foreign studies do not focus on the influence of forest micromosaic structure on the composition of rain deposit, and in domestic studies, long-term (lasting for more than 10 years) continuous observations of the influence of man-made pollution on the composition and properties of rain waters are anecdotal (Ershov et al., 2020). Using the example of the Murmansk Oblast forests, it was demonstrated that the chemical composition of rain waters shows significant intra- and interbiogeocenotic variation. On the basis of long-term (lasting for 18 years) data it was confirmed that the content of elements in the rain below the crowns is higher than between the crowns, and in spruce as compared to pine forests the concentration and fallout of elements was significantly higher which can be attributed to different sorption capacity of tree crowns. Long-term development of element concentrations in the rain waters of coniferous forests is highly variable. An increase in nickel concentrations was detected in the background setting during the period 2013–2017, which is explained by increased content of pollutants in aerosols that propagate over large distances.

Influence of forest stand and air pollution on the composition of soil waters

Soil is the most important component of the forest ecosystem; it accumulates most of the elements involved in biogeochemical cycles. Soil is porous and includes three main components: liquid, solid, and gaseous. Atmospheric precipitation is the most important source of moisture in soil. Water coming from the atmosphere contains dissolved gases and other substances, so it is not pure water that reaches the soil surface, but a certain solution of various gases, salts and other substances. Therefore, soil moisture is a certain solution, which is commonly called soil solution or soil water (Rode, 1955). V.I. Vernadskij believed soil moisture to be one of the most important categories of natural waters in the biosphere, "the main substrate of life" and "the main element of the biosphere mechanism" (Vernadskij, 1960). It is responsible for the mobility of elements in the soil profile, determining the redistribution of substances along the soil genetic profile and their removal to adjacent environments

(Raudina, 2015). The soil solution is the most active phase of the soil. It is where the majority of all chemical reactions of the soil occur. Water is a kind of a link in the organisms – soil – rock – atmosphere system. Metabolism is carried out mainly through the liquid phase, i. e. soil solution, ground and surface waters (Kovda, 1985). Soil water composition is the result of a complex of different processes, such as weathering, neutralization and buffering reactions in soils, precipitation from the atmosphere, absorption by plants, lateral and vertical water flows, and leaching (Motuzova et al., 2009). Chemical composition of soil waters provides information on both the availability of nutrients and the negative impact of pollutants on forest ecosystems.

Many foreign authors widely use the chemical composition of soil waters as a diagnostic tool for monitoring biogeochemical cycles in forest ecosystems. In order to better understand the mechanisms of impact of air pollution and other stress factors on the state of forest ecosystems, the second, intensive level of monitoring was established under the ICP Forests programme. The results of this intensive pan-European programme showed that in EU, NO_3 concentrations in the soil solution were beyond the levels allowed by water quality criterion in 9% of plots, and the ratio of concentrations of base cations and aluminium exceeded the critical limits in 30–39% of sites depending on the layer in 1997. The data obtained also led to the conclusion that the release of aluminium ions is the dominant process of buffering of acidic soil, whereas in less acidic soils, buffering occurs due to base cations. Changes in the base ion concentrations in soil waters can be explained by differences in atmospheric fallout and meteorological conditions (de Vries et al., 2003).

A study of soil water composition in European deciduous and coniferous forests (data for the period 1995–1998) demonstrated that the concentrations of nitrates in the soil solution have pronounced seasonal variability, summer concentrations being 25% higher than winter ones. It was also found that coniferous and deciduous forests react differently to the deposition of N compounds, so they need to be analyzed separately. Nitrate concentrations in the soil solution react to changes in nitrogen content in atmospheric fallout, and this is more noticeable in deciduous than in coniferous forests, mainly because deciduous forests grow on more fertile soils than coniferous forests (Kristensen et al., 2004).

In the UK, a long-term (lasting for 12 years) composition analysis of the atmospheric fallout and soil waters was performed at intensive monitoring sites. Assessment of the long-term dynamics of atmospheric precipitation composition confirmed the success of the UK's emission reduction policy. Long-term trends in the development of chemical composition of the soil solution revealed a gradual decrease in the content of sulfates and aluminium, as well as an increase in the pH. Concentration of dissolved organic nitrogen increased in atmospheric precipitation at open sites and in contact with the forest canopy, as well as in soil waters on most of the studied plots. There was a decrease in nitrates in soil waters on plots with high nitrogen deposition from the atmosphere. It was found that increased content of dissolved organic carbon in the soil can be explained by a noticeable decrease in pollution, a change in soil temperature, or an increase in microbiological activity (Vanguelova et al., 2010).

In the northern part of Belgium, studies of the chemical composition in atmospheric fallout and soil solution were conducted at five monitoring stations with coniferous and deciduous biogeocenoses over a seventeen-year period (1994–2010). The study revealed a decreased deposition of sulfate and ammonium in all the studied areas, as well as of nitrates in deciduous forests. The decrease in nitrogen and sulfur deposition was accompanied by a decrease in the deposition of the base cations (BC = $Ca^{2+} + K^{+} + Mg^{2+}$), and in general, there was a decrease in the flux of acidifying substances with the deposition. The content of ammonium, nitrates, sulfates, and base cations in the soil solution decreased with a decrease in atmospheric deposition of these elements. Deposition of acid-forming compounds N and

S in coniferous and deciduous forests in Belgium decreased significantly between 1994 and 2010, but forest soils are still in poor condition. Critical loads were exceeded, and soil acidification due to anthropogenic impact continued, which can be explained by a simultaneous decrease in the content of base cations, which affected the cation exchange processes (Verstraeten et al., 2012).

In the UK, a long-time (from 1993 to 2011) analysis of the composition of soil waters in forest and treeless areas was conducted to identify the impact of reduced air pollution and climate changes on the dynamics of dissolved organic carbon (DOC) concentrations. This study showed that the observed grassland and forest soils had significantly recovered from anthropogenic acidification; however, it is unlikely that these improvements can be associated with climate changes. Time models of long-term trends in DOC concentration vary between plots for reasons that may be related to soil properties, vegetation cover, the amount and source of acid-forming substances (anthropogenic or natural). DOC trends were mainly associated with temporary changes in acid-containing deposition. Changes in the composition of surface waters DOC largely correspond to changes in the DOC of the top layer of soil. If the deposition of acid-forming substances continues to decrease, an increase of DOC concentrations in surface horizons and adjacent surface waters may be observed (Sawicka et al., 2016).

Changes in the composition of soil waters of mineral horizons in European forests due to decreased deposition of acid-forming substances were being studied in 1995-2012. The data obtained revealed a significant decrease in the concentrations of sulfates in the soil solution by 52% at a depth of 10–20 cm and by 40% at a depth of 40–80 cm, whereas the concentrations of nitrates decreased only at a depth of 40–80 cm. The decrease in the concentrations of acid anions was accompanied by a significant decrease in the concentrations of the base cations (calcium, magnesium, and potassium) and Al_{tot}, and the changes in the soil solution acidity were not unidirectional. The results obtained revealed a non-linear relationship between reduced emissions and changes in the soil solution acidity and highlighted the importance of long-term monitoring for assessment of the response of ecosystems to reduced atmospheric pollution (Johnson et al., 2018).

Soviet and Russian scientists also studied the soil water composition, which can be used for early diagnosis of soil degradation under the influence of man-made pollution, as well as for soil monitoring and assessment of critical loads (Tjazhelye metally..., 1980; Karpuhin et al., 1993; Lukina, Nikonov, 1996; 1998; Motuzova, 2001; Kopcik, 2004; Motuzova et al., 2009; Lukina et al., 2018; etc.).

On the Kola Peninsula, studies are being conducted on the composition and properties of soil waters in forests at the northern range, both under natural conditions and in the setting of pollution by emissions from the most powerful sources of sulfur dioxide and heavy metal in Northern Europe – *Pechenganickel* and *Severonickel* mining and metallurgical smelters. According to the results obtained, significant intra-profile variability may be observed in the soil waters of boreal forests, which is expressed in decreasing concentrations of all elements with depth and is explained by the presence of biogeochemical barriers. Moreover, a pronounced intrabiogeocenotic variability is seen, soil waters below the crowns being more concentrated and acidic than between the crowns due to the formation of intense fluxes of acid-forming substances with crown and trunk waters. Variability of soil water composition depending on the type of biogeocenosis was reported: it was demonstrated that in the soil waters of spruce forests, the concentrations of elements are usually higher than in pine forests. The soil waters of boreal forests show pronounced seasonal variability: as a rule, carbon concentration and the acid-neutralizing capacity (ANC), which has a positive correlation with the carbon concentration, increase from spring to autumn. When approaching the source of pollution, functioning of all biogeocenosis components is disrupted, and therefore there is a decrease in the nutrient concentrations in soil waters and the ANC, as well as an increase in the concentrations of element compounds that make up the emissions. These changes are most noticeable in woody, especially spruce, parcels due to the high sorption capacity of coniferous crowns (Lukina, Nikonov, 1996, 1998; Kislotnye osadki..., 1999, Lukina et al., 2008).

Soil waters of the podzols in the zones of influence on the Kola Peninsula have an acidic reaction and a high content of organic matter; Ca and K are predominant cations, while anions are dominated by anions of organic acids and sulfates. Soil solutions in spruce forests are more acidic and concentrated in comparison with pine forests. When approaching the source of pollution, the concentrations of heavy metals (Ni, Cu, and Cd) sharply increase, copper being more strongly fixed by organic matter, and nickel being washed out into the underlying soil layers. Concentrations of organic matter, potassium, sodium, and under the spruce forests – also of calcium, manganese and zinc – decrease during the technogenic transformation. With an increase in air pollution, concentrations of soluble organic acids in litter waters decrease, and the concentration of sulfates increases. When comparing the soil waters of organogenic and mineral horizons, an increase in pollution leads to levelling of differences, which indicates a partial loss of the biogeochemical barrier function by the litter (Kopcik et al., 2007).

In the soil waters of pine and spruce forests of the Kola Peninsula, qualitative and quantitative composition of low-molecular-weight organic acids (LMWOAs) at different stages of technogenic degradation was studied. Studies of water composition of the background areas revealed that citric acid dominates among the LMWOAs. Leaching from tree crowns is an important source of LMWOAs for soil waters; however, in the middle of the growing season, the crown can perform barrier functions. When water migrates along the organogenic horizon of soils, it is enriched with organic acids. The content of LMWOAs in the soil waters of the background areas depends on the type of biogeocenosis, its parcel structure and the season of the year (Artemkina et al., 2008). When approaching the source of pollution, there is a decrease in the concentrations of citric acid in the soil waters of spruce forests below and between crowns. With increasing technogenic load, there are no differences in the concentrations of citric acid at the intrabiogeocenotic level. Atmospheric pollution affects LMWOA concentrations through changes in the composition of vegetation and litter (Artemkina et al., 2011).

A four-year monitoring of natural waters in deciduous and coniferous forests was conducted in Moscow Oblast. Its aim was to study the concentrations and flows of dissolved organic carbon (DOC) in the atmospheric precipitation – subsurface waters – soil waters system. The study revealed that atmospheric precipitation has low and relatively constant concentrations of organic carbon, and when in contact with the tree canopy, the precipitation is enriched with carbon, to the greatest extent in the pine-spruce forest. Soil waters have a high concentration of DOC, which varies widely depending on the type of biogeocenosis, soil properties and depth. Significant variability of carbon concentrations in below-the-crowns- and soil waters was observed, both from season to season and from year to year. It was found that soluble organic compounds of atmospheric and crown origin have a negligible effect on the carbon of soil waters. In the setting of the leaching water regime in podzolic soils, below the crowns of coniferous and mixed forests, the removal of organic carbon with soil waters during the growing season prevails over its intake with precipitation (Sultanbaeva et al., 2015).

In the *Kivach* reserve of Kondopoga district, the Republic of Karelia, the chemical composition of soil waters in coniferous forests of the middle taiga was studied in 2009–2011. It was noted that the ionic composition of soil waters was dominated by potassium, calcium, sulfates and hydrogen carbonates, and the pH corresponded to slightly acidic waters. Organic nitrogen dominated among the nitrogen compounds; there was a high content of organic substances and lithophylic elements; zinc and copper had the highest concentration among heavy metals. With depth of the soil profile in the spruce and pine forests, reduced concentrations of potassium and calcium were observed, as well as an increase in sulfates, sodium, hydrogen carbonates and the pH, and a decrease in the content of total nitrogen (and in the pine forest also of phosphorus) and organic matter in the soil waters. In both soil profiles, the content of aluminium and silicon decreased with depth. The profile of pine forests showed an increase in cadmium and copper content and a decrease in zinc content, whereas in spruce forests increased concentrations of cadmium, lead and zinc and decreased concentrations of cobalt, nickel and copper were found (Bahmet et al., 2011; Kravchenko, 2016).

Thus, the study of soil water composition is still a relevant problem. As in the case of research on atmospheric fallout, foreign studies of soil water composition do not pay attention to the influence of the micromosaic structure of biogeocenosis on the composition of soil waters, whereas Russian publications are rarely based on long-term studies (Ershov et al., 2019). Using the example of forests in Murmansk Oblast, significant intra- and interbiogeocenotic variability of soil water was revealed. As a rule, element concentrations in lysimetric waters of all soil horizons are higher below the crowns as compared to that between the crowns, while in the soil waters of spruce forests concentrations of element compounds are significantly higher than in the waters of pine forests. Long-term dynamics of the concentrations of heavy metals and sulfates in soil waters is highly variable and shows a downward trend, which may indicate a gradual decrease in the anthropogenic load. However, the analysis of the obtained data and their comparison with the background values gives evidence of a significant impact of industrial air pollution on forests.

Assessment of critical levels of impact on forests based on the composition of atmospheric fallout and soil waters

Assessment of soil resistance to pollutants is based on soil-ecological principles (Glazovskaja, 1989, 1990, 1999). Soils are considered contaminated if pollutants are accumulated in quantities that are hazardous for living organisms. Expert assessment of soil contamination is based on their intrinsic properties: horizon thickness; humus content, composition, and properties; grain size distribution, cation exchange capacity, soil biological activity, content of soluble forms of pollutants, etc. For more than a quarter of a century, another approach has been developed – the establishment of critical loads of pollutants. It is related to the systematization of soils and ecosystems according to the degree of their resistance to pollutants. The ideas of environmental load rationing at the international level were embodied in the development of the concept of critical loads as part of the Convention on Long-range Transboundary Air Pollution (LRTAP). In accordance with the Convention, emissions of pollutants should be reduced to acceptable (critical) deposition or concentrations (Kopcik, 2004).

The concept of critical load implies an indicator that characterizes the maximum value of impact of one or more pollutants per unit area of the landscape, below which, at the current level of knowledge, there are no significant negative processes for specific sensitive elements of the environment (Nilsson, Greenfelt, 1988). Critical loads are calculated using chemical indicators, or critical limits, which determine the harmful effects and thresholds. In atmospheric fallout (rain and snow), the critical load is usually calculated for a particular element, for example, for sulfate sulfur (Korhola et al., 1999), nitrogen (Waldner et al., 2007), or heavy metals (Reinds et al., 2006). To assess soil acidification that can negatively affect the growth of trees, the molar ratio of the sum of base cations (calcium, magnesium and potassium) to aluminium in soil solutions was proposed (Sverdrup, 1993). At elevated acid loads, the soil is gradually depleted of the base cations, mainly calcium (Ca^{2+}) and magnesium (Mg^{2+}). Toxic forms of aluminium are generated at soil acidification. An increased concentration of such Al forms in the soil solution can lead to inhibition of root growth, damage to small roots and mycorrhiza, and thus reduce the intake of nutrients and water (Foy, 1988; Ulrich, 1983; Boudot et al., 1994; Godbold, Hüttermann, 1988; Godbold, Kettner, 1991; Godbold et al., 1988). To assess eutrophication and nitrogen saturation, mineral nitrogen concentrations in soil solutions are used. An increase in N deposition can cause soil eutrophication, as well as intensify the leaching of nitrates, which is often referred to as N saturation (Aber et al., 1998; Gundersen, 1991). In general, the result may be a lack of nutrients and a decrease in the growth rate of forest plants. Increased nitrate leaching is associated with soil acidification, which causes a gradual decrease in the concentration of base cations in soil solutions, followed by an increase in Al concentrations.

The composition of soil solutions on six forest plots with acidic mineral soils was studied in Switzerland in 1999-2002 to determine the critical load exceedance. The molar ratio of base cations to the total dissolved aluminium (BC/Al_{tot}) was analyzed. The average BC/Al_{tot} ratio in the soil solution never reached a critical value in the root zone on all plots during the entire observation period. The fundamental BC/Al ratio in soils significantly correlated with the BC/Al_{tot} ratio in the soil solution. Soil solutions with the lowest BC/Al_{tot} ratios ($\leq$ 2) were usually found in mineral soils with a BC/Al ratio below 0.2. Chemical composition of the soil solution depended on the composition of the underlying soil layers. At 80 cm, the pH values and the BC/Al_{tot} ratio of soil solutions were much higher than expected, so it is recommended to take this into account when calculating critical loads of acidity (Graf Pannatier et al., 2004).

In an area with pronounced marine impulverization and high nitrogen deposition in the south-western part of Jutland, Denmark, studies were conducted in 1989–1999 to assess the suitability of the BC/Al indicator and the Ca/Al ratio in the soil solution as chemical criteria for calculating the critical load on forest ecosystems. It was found that the levels of atmospheric fallout in coniferous forests on acidic soils exceeded the critical load for N in Denmark. Intensive leaching of N from the soils of forests formed by Sitka spruce (*Picea sitchensis*) indicated a high level of N saturation. Although in spruce forests N deposition was higher than the level of critical loads, the study area was not yet saturated with N, since the leaching of nitrates with soil waters was low. Beech forests (*Fagus sylvatica*) had broader BC/Al_{tot} ratios in soil waters than forests formed by Sitka spruce and Norway spruce (*Picea abies*) in 1989, but these ratios were gradually approaching 1 for forests formed by all three species by 1999. Very narrow Ca/Al_{tot} and BC/Al_{tot} ratios observed in this study are not consistent with the improved or non-changing condition of the stands. This has called into question the applicability of these ratios as chemical criteria in areas with high levels of sea salt deposition. Proximity to the sea and significant supply of base cations to the upper soil layers can contribute to the resilience of forests and help them overcome nutrient deficiency in the deeper soil layers (Hansen et al., 2007).

In Europe, large-scale studies were conducted to study the variation of sulfur and nitrogen deposition and the exceedance of their critical loads at ICP Forests Level II intensive monitoring sites. Data at more than 150 monitoring sites were obtained from 2000 to 2005. Deposition of ammonium, nitrates and sulfates under the forest canopy was higher than at open sites, and there was also a reduction in sulfur emissions and a less significant reduction in nitrogen emissions in Europe. In the studied areas, the exceedance of critical loads for sulfur was less than for nitrogen. The exceedance of the critical nitrogen level was found in the throughfall of about 2/3 of the studied sites. The highest exceedances were found in the Netherlands, Belgium and some states of Germany, whereas the lowest – in the United Kingdom, in Fennoscandia, Greece and in the Alps. Critical loads were exceeded in less than a quarter of the sites located mainly in the Netherlands, southern Sweden, some states of Germany and in Hungary. It is noted that the exceedance of critical loads makes it possible to determine the threshold for certain pollutants that can cause damage to the ecosystem in some time. Therefore, to assess the ecosystem response, it is important to determine whether critical limits are reached (Lorenz, Granke, 2009).

In the northern part of Belgium, the chemical composition of atmospheric fallout and soil solutions over a seventeen-year period (1994–2010) was studied at five monitoring stations with coniferous and deciduous biogeocenoses. The study revealed that the BC/Al ratios were below the critical level at three sites on sandy soils with a low degree of soil saturation with bases. The acid-neutralizing capacity in the soil increased, but remained negative, indicating that the acidification of the soil continued, since the beginning of recovery was slowed down by a simultaneous decrease in the deposition of base cations and short-term processes of soil buffering. The critical limits of N concentrations in the soil solution were largely exceeded for most of the year at all depths at five sites (Verstraeten et al., 2012).

Another large-scale study was conducted in Europe in 2006–2009 at more than 200 monitoring sites. It was aimed to look into the relationship between critical load exceedance and the concentration of inorganic nitrogen, the ratio of base cations to aluminium in soil solutions, as well as the nutritional status of trees. This study demonstrated differences in the frequency of critical limit exceedance for soil solution between groups of sites classified according to the current excess of critical loads, and these exceedances are likely to persist for several decades. A similar differentiation was found when assessing the nutritional status of trees for groups identified in accordance with the critical limit exceedance for the soil solution. The results support the hypothesis that eutrophication or acidifying effects of the deposition of inorganic compounds N and S can lead to an imbalance in the nutrition of trees (Waldner et. al., 2015).

Assessments of critical loads on forests were also carried out by Russian scientists. For example, the state of atmospheric air and precipitation of the Russian Plain was studied in 1982-2003. On its vast territory, there are large pulp and paper, metallurgical, chemical, oil and gas production, and processing enterprises. Their emissions and discharges have a negative impact on the environment. The study demonstrated that critical limits for sulfur deposition were exceeded at local sites in Leningrad, Moscow, and Ryazan Oblasts. In terms of total nitrogen deposition, the level of critical loads was exceeded in about half of the European part of Russia: in the North-Western, Northern and Central regions. In general, at the studied area of the European part of Russia (3.2 million km^2), the levels of critical loads for nitrogen were exceeded on an area of 2.3 million km^2, and in terms of sulfur – on an area of 1.7 million km^2 (Trubicina, 2008).

Critical loads by acid-forming sulfur compounds and the risk of their excessive entry into ecosystems were assessed on the Kola Peninsula. The study revealed that 58% of the area of the Kola Peninsula is occupied by the most sensitive ecosystems (northern taiga forests and crooked forests on podzols, as well as tundra and forest-tundra communities), for which the critical sulfur loads do not exceed 400 eq ha^{-1} year^{-1}. Stable systems (from 500 to 600 eq ha^{-1} year^{-1}) include mountain tundra and forests, birch crooked forests and sparse forests on podzolized brown soils and illuvial-humus podzols that occupy up to 8% of the peninsula area. The most stable systems (more than 700 eq ha^{-1} year^{-1}) occupy 7% of the area and, along with swamp communities, include pine and spruce forests on illuvial-humus podzols. More than 20% of the Kola Peninsula are high-risk zones – these are vast territories in the north-west and in the centre, which are located near *Severonickel* and *Pechenganickel* smelters (Kopcik et al., 2008).

As part of one of the international projects of OOO Gazprom-VNIIGAZ, an assessment of critical loads of acidifying and eutrophying compounds for the territory of Venezuela was carried out in light of the planned gas industry intensification in this country. According to calculations, ecosystems of more than 40% of the territory of Venezuela have a high potential for resistance to acid deposition, while the ecosystems formed on 10% of the area are characterized by reduced resistance to acidifying compounds. The least stable are the mountain forest ecosystems on shallow soils along the eastern borders of Venezuela and in the mountainous region of the Andes in the west of the country. Increased and high risks of ecosystem eutrophication were detected in a limited area (approximately 0.2 and 0.8%, respectively) and are typical of territories located in the zones of impact of industrial centres of Venezuela (in the west and north-east of the country). This study of the joint use of risk and critical load strategies showed good perspectives of this approach for solving practical problems of environmental and nature protection activities of enterprises (Priputina, Bashkin, 2012).

In the review of approaches to assessing both the ecological state and the standardization of the quality of soils (Konovalov et al., 2017), an attempt was made to highlight Russian and foreign approaches to this problem. It is noted that when assessing the ecological state of soils, it is very important to distinguish between different types of soil degradation: physical, chemical, microbiological and complex. In Russia, the sanitary and hygienic approach is mainly used. It implies the concept of maximum allowable concentration (MAC), that is, such a content of chemical elements in the environment that does not cause a negative (direct or indirect) impact on human health during a long time. Other than that, the MAC characterizes only the degree of anthropogenic disturbances in ecosystems; if the existing level of pollutant concentrations is lower than the MAC, it is considered that further anthropogenic impact on this area is permissible, and if the MAC is exceeded, the impact should be stopped. However, this approach does not provide answers to the questions what level of pollutants entering the ecosystem is acceptable in order not to disrupt the functioning of natural systems and to which extent the impact should be reduced to stop negative changes in the environment (Bashkin et al., 2005). Therefore, this approach has significant drawbacks and other approaches need to be found for the development of the environmental regulation system. Possible options are biogeochemical and statistical approaches, as well as ecosystem standardization. An approach based on the assessment of environmental risks, which includes the concept of critical loads, is gaining popularity abroad. In Russia, its use can also be effective, as demonstrated in the works of V.N. Bashkin (2004), G.N. Kopcik and S.V. Kopcik (2008) etc.

Thus, foreign and domestic authors have developed various methods of assessment and measures to reduce the impact of industrial air pollution on forest ecosystems. It should be noted that when assessing critical loads on forests, the influence of the micromosaic structure of the biogeocenosis is rarely taken into account (Ershov et al., 2020). Using the example of Murmansk Oblast forests, it was revealed that the deposition of heavy metals and sulfates in the atmospheric fallout exceeded annual critical levels (up to 4 times) already in the background setting, but only below the crowns. The highest exceedances are observed in defoliating forests and pollution-induced sparse forests, which is especially marked below the crowns of spruce forests, where the maximal exceedances of the level of critical loads made up up to 7 times for sulfates deposition and up to 600 times for heavy metals deposition. The exceedance of critical levels in soil waters was estimated using international indicators: the ratio of base cations to aluminium (BC/Al) and the concentration of mineral nitrogen in the soil solution (Nmin) (Ershov et al., 2019). The ratio of base cations to aluminium at all stages of degradation in spruce and pine forests significantly exceeds the critical values, which can be explained by the fact that the soil-forming rocks and soils of the study region are rich in base cations. The critical level of mineral nitrogen is exceeded at all stages of degradation, and below the crowns the exceedance is usually higher than between the crowns.

The analysis of publications allows us to conclude that the assessment of the composition of atmospheric fallout and soil waters requires taking into account the intra- and interbiogeocenotic mosaic structure of the forest cover. It will allow for early diagnosis of technogenic pollution of forests. To assess and forecast the dynamics of biogeochemical cycles in forest ecosystems, it is necessary to assess long-term changes in the composition and properties of atmospheric fallout and soil waters. To identify the exceedance of critical loads on forest ecosystems, it is advisable to use the critical loads concept.

ACKNOWLEDGEMENTS

The study was carried out within the topic of the state assignment of Kola Scientific Center RAS No. AAAA-A18-118021490070-5

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