Direct and Indirect Effects of Regional Air Pollution on Tree Crown Defoliation

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Abstract

Changes in tree crown condition, in most cases, may be related to the integrated impact of natural and anthropogenic factors, where S and N deposition, through the combination of direct, above-ground, impacts of SO_2, NO_x and NH_4^+ on foliage and indirect, soil-mediated, impacts on roots play a predisposing, accompanying and locally, even a triggering role. This study was designed to check which effect is more significant on changes in tree crown defoliation. The findings revealed that the direct effect of air pollutants and acid deposition should be a more significant on Scots pine crown defoliation than the indirect effects of acidifying compounds through soil, ground and runoff water. Needles, which are present on trees all year round, seem to be more efficient aerosol collectors than leaves. In contrast, the direct effect of the considered contaminators, especially N compounds, on birch defoliation, was less pronounced than their indirect effect. In most cases, the considered contaminators had a negative effect on crown condition; meanwhile nitrate, its deposition and concentration in soil and groundwater, had a positive effect. *Ips typographus* L., which caused the dying of spruce trees, did not allow fulfilling the task.

Key words: nitrogen and sulphur deposition, soil and ground water contamination, tree defoliation

Introduction

Changes in forest ecosystems, in most cases, may be related to the integrated impact of natural and anthropogenic factors, where air concentration of surface ozone, and S and N deposition play a predisposing, accompanying and locally, even a triggering role (Chappelka and Freer-Smith 1995, Cronan and Grigal 1995, Manion and Lachance 1992, Schulze 1989). These rather different effects of air pollution could be explained by the combination of direct, above-ground, impacts of O_3, SO_2, NO_x, NH_4^+ and H^+ on foliage and indirect, soil-mediated impacts of acid deposition on roots, which may cause nutrient deficiencies and aggravate natural stress, such as physiological drought and the occurrence of pests and diseases (De Vries et al. 2000a). These indirect, soil-mediated effects of air pollutants on the vitality of forests are frequently more important than the direct above ground effects in local polluted areas (Roberts et al. 1989, De Vries et al. 2000b). However, on a regional scale, an indirect effect of pollutants could only be detected when data of repeated surveys for a relatively long period, especially for variables that change slowly, become available (De Vries et al. 2003a). The Integrated Monitoring Programme, which has been performed for 15 years in Lithuania, provides all the necessary data to detect which effects – direct, through the impact on leaves and needles or indirect, through the changes in soil mineralization and eutrophication processes – might be more significant to changes in crown defoliation of the prevailing tree species in Lithuania, and which of them constitutes a greater threat in their integrated impact.

To answer this question the following research was performed:

- an analysis of temporal and spatial variation of data on defoliation, air pollutant concentrations and acid deposition;
- an analysis of variation of data on quality of soil, ground and stream waters in relation to air pollutants and their deposition;
- a correlative analysis of data on crown defoliation of prevailing tree species in Lithuania versus air pollutants and acid deposition, as well as their concentrations in soil, ground and stream water;

• an estimate of the contributions of effect character to integrated impact of air and soil mediated pollutants on tree defoliation.

Method and materials

The following are considered in this study: concentrations of sulphur (S) and nitrogen (N) compounds on the air, precipitation and their deposition; concentration of N and S compounds in the ground water, soil water at the depth of 20 cm, and stream water; and defoliation of the prevailing tree species. The study was based on the 1994-2007 data available from 3 Lithuanian Integrated Monitoring Stations (IMS) established in Aukštaitija, Dzūkija (1993), Žemaitija (1994) National Parks (NP) (Fig. 1). In 2000, the Dzūkija IM Station was closed due to underfinancing and, therefore, only the data on pollution and crown condition over the period from 1994 to 1999 were used from this IM station.

Crown defoliation of the three tree species prevalent in the catchments – Scots pine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* Karst.) and Birch (*Betula pendula* 'Crispa' and *B. pubescens* Ehrh.) were assessed on vegetation plots established for intensive monitoring at IMS catchments. The crown condition of about 600 trees (pine trees – 30%, spruce – 50% and birch – 20%) was assessed annually, employing the methodology of the ICP Forest monitoring programme over the period 1994-2007 (UN-ECE 1994) on 3 intensive plots (IP's): A, B and C at LT-01, of about 100 trees (pine trees – 90%, spruce and birch – 10%) on 1 IP at LT-02 (A), and of about 100 trees (pine trees – 20%, spruce – 70% and birch – 10%) on 1 IP at LT-03 (A). The main characteristics of the considered stands are presented in Table 1.

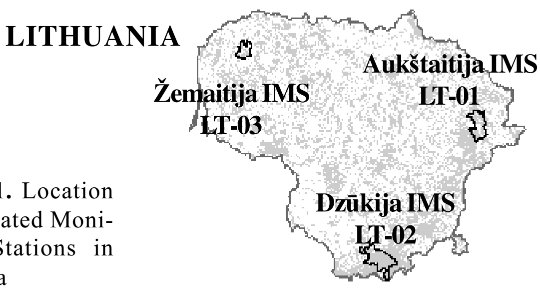

Figure 1. Location of Integrated Monitoring Stations in Lithuania

Table 1. Main dendrometric characteristics of the intensive monitoring plots

IMS	VG plot	Age class	Mean diameter cm	Mean height M	Stocking	Volume (m^3/ha)	Forest type
			Stand parameters				
LT-01	A	15	38	26	0,5	300	*vacciniosum*
LT-01	B	17	46	31	0,7	440	*oxalidosum*
LT-01	C	8	38	28	0,6	260	*caricosum*
LT-02	A	8	24	26	0,8	370	*myrtilius – oxalidosum*
LT-03	A	9	34	26	0,8	230	*cladoniosum*

Glacioaquatic accumulation forms with sand, gravel and stones are typical of the LT-01 catchment. In this area, IPs LT-01A and LT-01B, bores No 1 and 2 for ground water studies and lysimeters for soil water collection are arranged. Soil water samples were collected at 20 cm (main root zone), with a sampling period of 3–4 times per vegetation period. The surface area of the plate type lysimeter was 1000 cm^2. With a decrease in altitude, these glacioaquatic accumulation forms transfer into marsh accumulation forms with organic sediments where IP LT-01C and bore No 3 are located.

The geomorphologic structure of the LT-02 catchment is formed by more intensive glacioaquatic and eolian processes, than in LT-01. The relief represents a re-modified fluvioglacial plain, with clearly defined continental dunes of complicated shapes, where fine-grain sand dominates. All 3 bores for GW collection are arranged there. Soils are formed on quartz sands of eolian origin and contain no carbonates. Premature and mature pure pine stands on the haplic arenosol where lysimeters for soil water collection are arranged, dominate in the catchment.

The geomorphologic structure of LT-03 catchment is different from that of the other locations. The marsh accumulation forms with organic sediments transfer into limnoglacial accumulative forms and glacioaquatic accumulative sandy and hilly formations, with typical limnoglacial sand. Bores are situated on the slope of a glacioaquatic hill covered by sand stratified to a depth of more than 1.5 m with thin layers of clay loam.

A spruce forest with two or more age classes and with up to a 20-30% pine mixture on albic arenosol where IP LT-03A, bore No 1 and lysimeters for soil water collection are arranged, dominates in the Žemaitija IMS.

At IM stations, SO_2, SO_4^{2-}, the sum of nitrate ($\Sigma NO_3^- = NO_3^- + HNO_3$), and the sum of ammonium ($\Sigma NH_4^+ = NH_4^+ + NH_3$) concentrations in the air and H+, SO_4^{2-}, NO_3^- and NH_4^+ concentrations in precipitation, as well as their wet deposition, were established. The air sampling was carried out at weekly intervals. The sampling equipment for SO_2 and particulate sulphate consisted of a two-stage filter pack sampler with a cellulose filter (Whatman 40). SO_2 was collected by retention of particles using a Whatman 40 filter impregnated with potassium hydroxide (KOH). ΣNO_3^- and ΣNH_4^+ were collected using an open-face separate sampler with alkaline (KOH) and oxalic acid impregnated Whatman 40 filters, respectively.

Precipitation samples were collected in a polyethylene bulk-collector from December to March and in an automatic wet-only sampler during the remaining months. All samples were stored at 4°C until laboratory analysis. Ion chromatography using Dionex 2010i with conductivity detection was used for the chemical analysis of anions in precipitation and in water extracts from the impregnated Whatman 40 filters. NH_4^+ concentration in precipitation as well as in the extracted solutions from oxalic acid impregnated Whatman 40 filters was analysed spectrophotometrically using the indophenol blue method. The overall measurement and analytical procedures were based on a quality assurance/quality control (QA/QC) programme as described in the EMEP CCC manual for sampling, chemical analysis and quality assessment (EMEP 1977). Analytical methods were controlled through the international (EMEP and GAW) analytical intercomparisons.

The Fisher test was employed for estimating the significance of spatial and temporal differences in changes in pollution level and tree defoliation; multiple regression analysis was used to establish the significance of direct and indirect effects of acidifying compounds on tree defoliation. Statistical analysis was performed employing the techniques of "Statistica 6.0" software.

Results

Concentration of air acidifying compounds and their deposition fluxes

IMS data showed a significant decrease in pollutants until the year 2000 (Sopauskiene et al. 2001, Augustaitis et al. 2005, Sopauskiene and Jasineviciene 2006, Augustaitis et al. 2007a). The air concentration of SO_2 at Aukštaitija IMS decreased by 82% (from 2.73 to 0.49 µgS/m³), at Žemaitija IMS by 79% (from 2.22 to 0.47 µgS/m³), and at Dzūkija IMS by 57% (from 3.0 to 1.3 µgS/m³) (Fig. 2). Thereafter, the concentration was stable at the level of 0.5 – 1.0 µgS/m³. Air con-

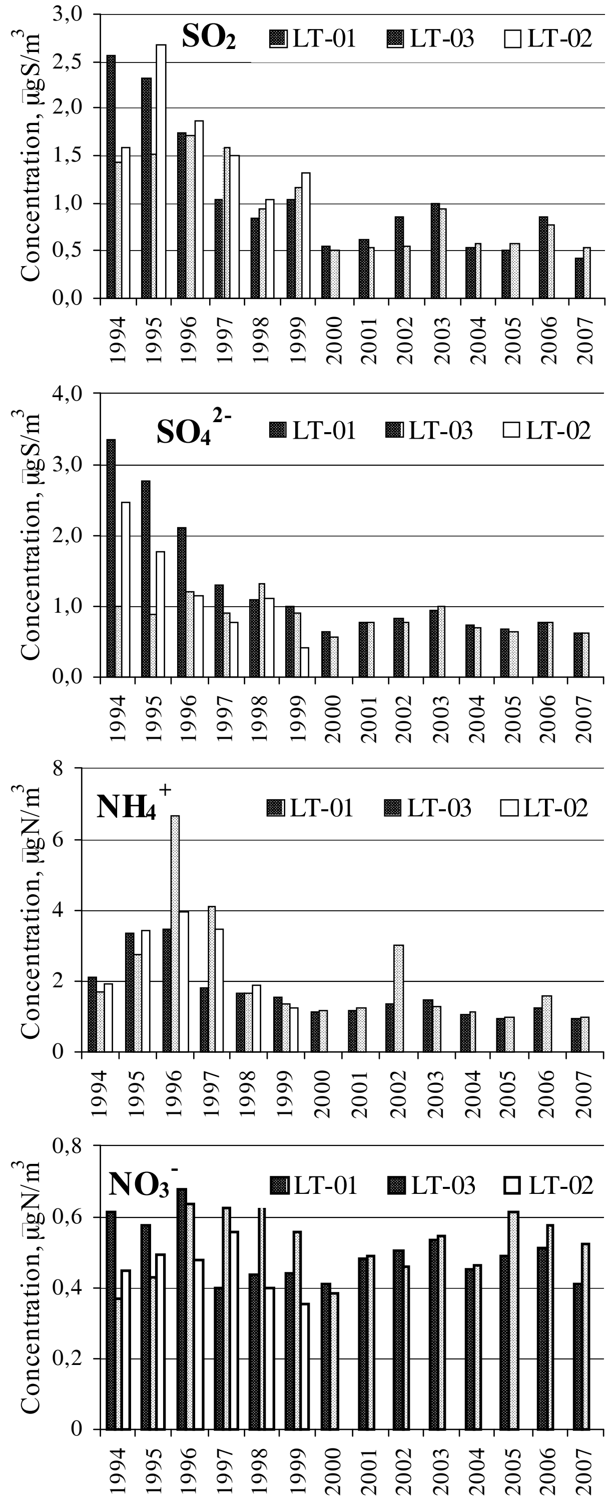

Figure 2. Changes in the mean air concentration of the considered contaminators from 1994-2007 (LT-01 – Aukštaitija IMS; LT-02 – Dzūkija IMS; LT-03 – Žemaitija IMS)

centration of aerosolic SO_4^{2-} changed in a similar pattern to SO_2 air concentration.

The most significant decrease in ΣNH_4^+ air concentration lasted until 2001: 86% in LT-03 (from 8.55 to 1.15 μgN/m^3), 77% in LT-01 (from 4.44 to 1.02 μgN/m^3), and 65% in LT-02 (from 3.91 to 1.37 μgN/m^3). Afterwards, a stabilization of air ΣNH_4^+ concentration at the level of 1.1 – 1.3 μgN/m^3 in both LT-01 and LT-03 was observed. Annual means of ΣNO_3^- concentration in the air were stable at the level of 0.5-0.7 μgN/m^3 in all stations over the entire considered period.

Changes in annual wet deposition had a very similar pattern to that of the air. The wet deposition of sulphur for the period 1994-2000 at the Aukštaitija NP decreased by 58% (from 600 to 250 mgS/m^2), at Žemaitija by 60% (from 750 to 300 mgS/m^2), and at Dzūkija by 52% (from 660 to 320 mgS/m^2) (Fig. 3). This marked decrease was most likely the result of a reduction in SO_2 emissions in Europe including Lithuania (CLRTAP 2005). Since 2001 at LT-01, sulphur deposition further decreased gradually to 200 mgS/m^2, while at LT-03, it drastically increased again in 2002, 2003 and 2007, reaching around 600 mgS/m^2.

Until 2001, a more than tenfold decrease in H^+ deposition and its concentration in precipitation were observed in the stations as well. However, since 2001, an increase in H^+ deposition and its concentration in precipitation has been observed, especially at LT-03.

Decreases in annual wet deposition of NH_4^+ - from 492 to 198 mgN/m^2 at LT-01 and from 537 to 303 mgN/m^2 at LT-03, have occurred. No significant change in wet deposition of NO_3^- was observed (Fig. 3). Annual wet deposition values for NO_3^- ranged from 241 to 211 mgN/m^2 at LT-01, from 241 to 270 mgN/m^2 at LT-02 and from 414 to 342 mgN/m^2 at LT-03, with the exception of 2002, when it reached the peak.

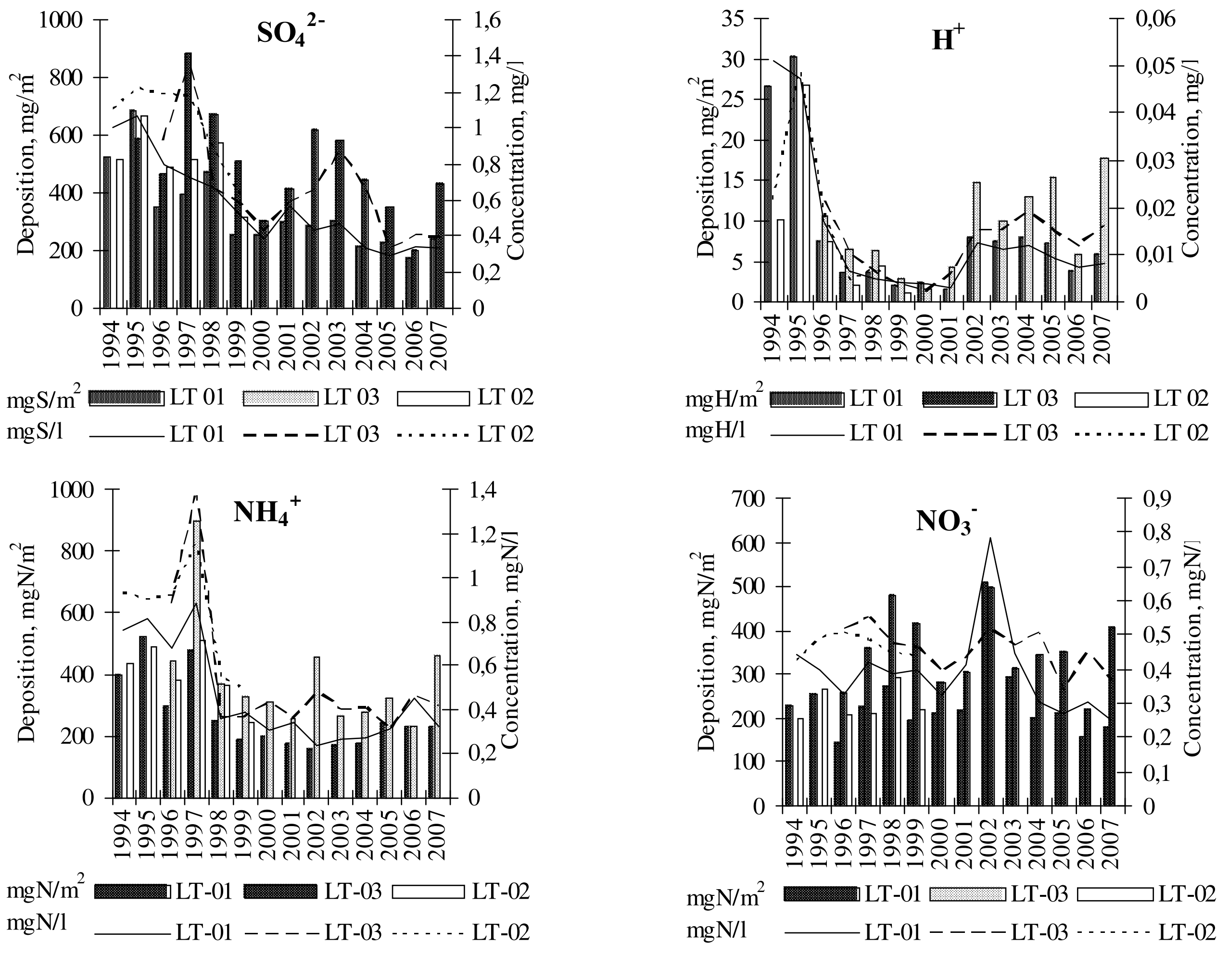

Figure 3. Changes in acid deposition and their concentrations in precipitation (LT-01 – Aukštaitija IMS; LT-02 – Dzūkija IMS; LT-03 – Žemaitija IMS)

An analysis of the spatial pattern of regional pollution levels reveals that the Western and South-western parts of Lithuania (LT-02 and LT-03 sites) were more polluted by the considered pollutants. This was most likely related to the proximity of these regions to the major pollutant sources in Central Europe as well as to the differences in the meteorology.

S and N concentrations in soil, ground and run-off water and their flows

Soil water quality

In the period from 1994 to 1999 soil water at LT-02 was more contaminated with SO_4^{2-} and NH_4^+ than in the other sites and demonstrated significant downward trends (Fig. 4): SO_4^{2-} concentration decreased by 0.79 mgS/l per year, and NH_4^+ by 0.15 mgN/l per year. At LT-03 the decrease in SO_4^{2-} concentration was not so evident at 0.24 mgS/l per year. Meanwhile at LT-01, SO_4^{2-} concentration in soil water was stable at the level of 4-5 mgS/l. The decrease in NH_4^+ concentration at LT-01 was at about 0.03 mgN/l per year, and at LT-03 the concentration remained stable at the level of 0.20 mgN/l. At LT-03 a downward trend in NO_3^- concentration in soil water was detected (0.013 mgN/l per year), while at LT-01 and at LT-02 the concentration increased by 0.021 mgN/l and 0.075 mgN/ l per year, respectively.

The detected changes in NO3- concentration resulted in a significant increase in soil water pH at LT-01, by 0.1 unit per year and a decrease at LT-03, by 0.01 unit per year (Augustaitis et al., 2005, 2008). This means the acidity of soil water at LT-01 decreased while at LT-03 it increased.

Ground water quality

Nitrate concentrations in the ground water of LT-01 had no statistically significant trends, whereas at LT-03 NO_3^- concentrations in the ground water of the

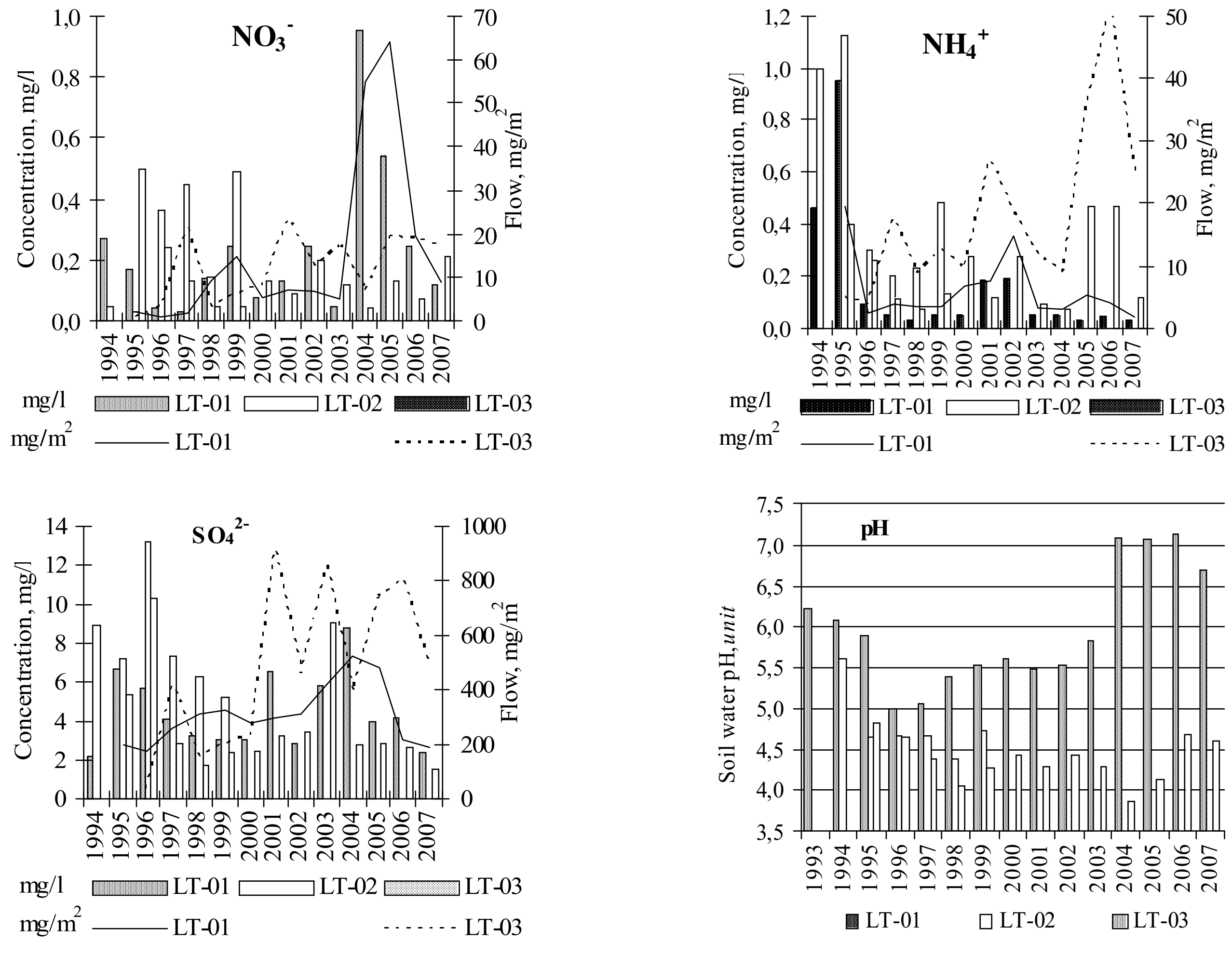

Figure 4. Concentrations of the acidifying compounds in soil water and their flows at 20 cm depth (LT-01 – Aukštaitija IMS; LT-02 – Dzūkija IMS; LT-03 – Žemaitija IMS)

shallow bores showed a decreasing trend, and in the water of the deeper bores an increasing trend. The tendency of NH_4^+ concentration in ground water was to decrease in all bores of all IM stations.

SO_4^{2-} concentration changes had no regular patterns at the LT-01 station. The exception was the year 1996 and the last period from 2005 to 2007, when SO_4^{2-} concentration increased drastically in the third bore (near the vegetation plot LT-01 B). At LT-03 SO_4^{2-} concentration changes had a tendency to decrease. The detected changes resulted in a gradual decrease of the ground water acidity at all considered depths. An exception, however, was the change in water acidity of the shallow bore at LT-03, which showed a tendency to increase.

A comparison of the means of concentrations of separate chemical components in soil and ground water of all three stations over the considered period, revealed higher concentrations of the most parameters in Dzukija (LT02), when this station was under operation. It is highly probable that this can be attributed to good infiltrational features of the continental dune sand. Since 2002, higher concentrations of the considered contaminators were also observed at LT03, mainly due to higher air concentration of the considered pollutants and their deposition.

Runoff water quality

Stream water quality and runoff of the main chemical compounds from ecosystems reliably reflect a common tendency of chemical processes occurring in forest ecosystems. Therefore, this parameter was used in the analysis to detect the indirect effect of deposition on tree conditions.

The concentrations of NO_3^- and SO_4^{2-} in the runoff water had no statistically significant trends over the considered period in all stations (Fig. 5), however, since 1998 at LT-01 a significant decrease in SO_4^{2-} concentration was observed. The concentration of NH_4^+ in stream water had a tendency to decrease in all stations over the whole observation period as well. The year 2007 was an exception, when the concentration of this contaminator at LT-03 increased drastically. These detected changes resulted in a gradual decrease of stream water acidity at all sites.

Despite the quite similar character of changes in NO_3^- and NH_4^+ concentrations in runoff water, there was evident difference in their output. Over the considered period, the output of both N compounds at LT-01 had a tendency to decrease, while at LT-03 the tendency was to increase (Fig. 6). Similarly, the output of sulphur at LT-03 increased significantly while at LT-01 and LT-02, it decreased only until 2000, after which some increase was observed.

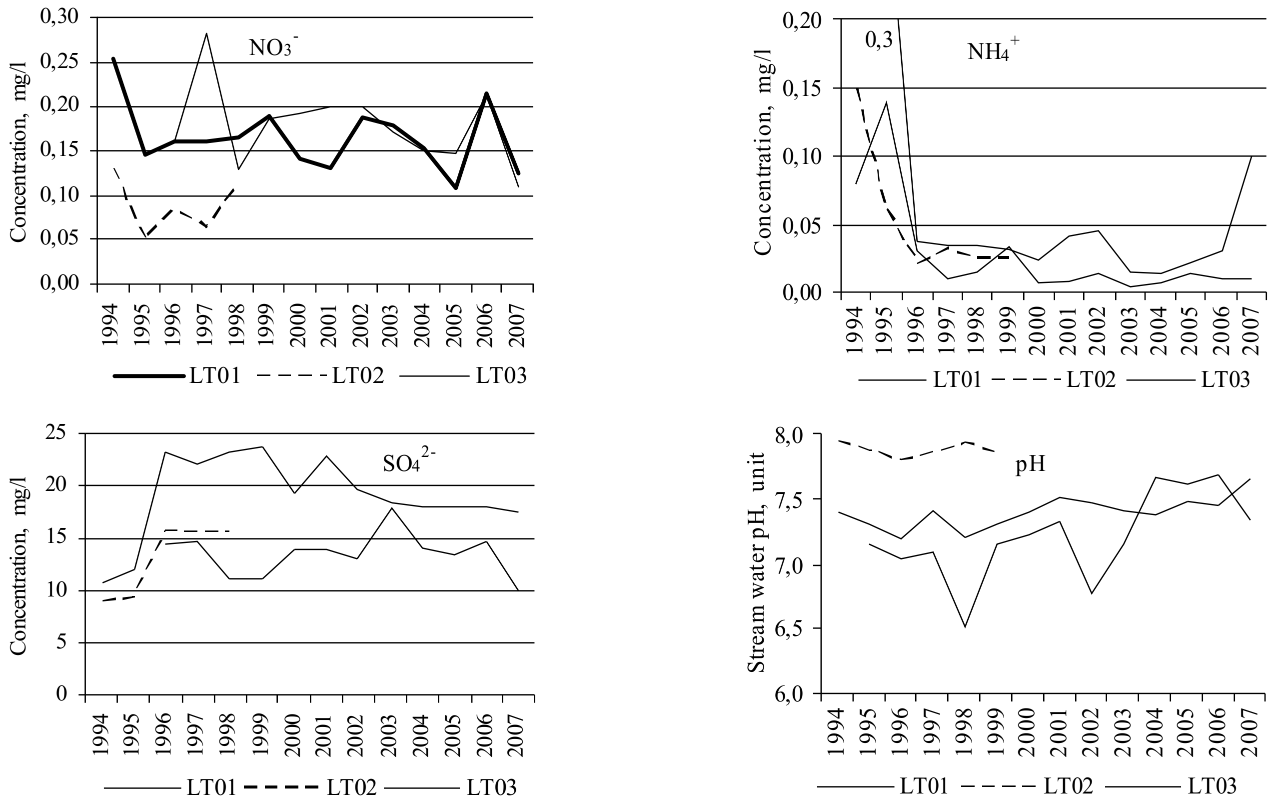

Figure 5. Concentrations of the acidifying compounds in stream water of IM sites

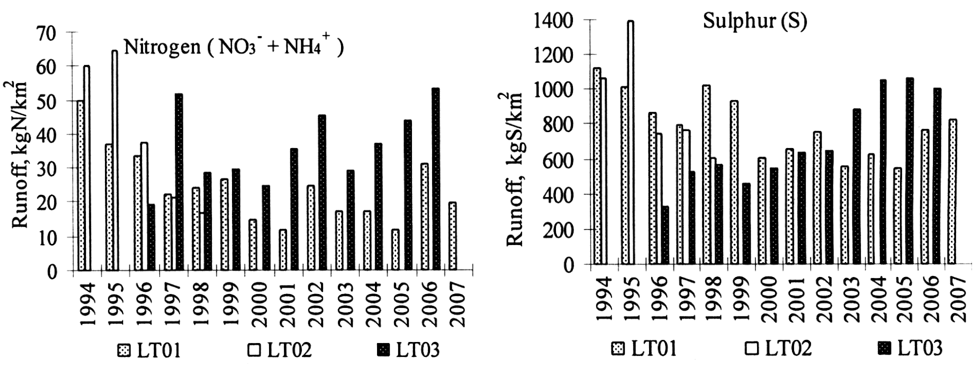

Figure 6. Runoff of sulphur and nitrogen from the considered forest ecosystems

Changes in crown defoliation of prevailing tree species

F test statistics indicated significant differences in mean defoliation of the prevailing tree species ($p<0.05$) among and within the intensive plots at Aukštaitija IMS (LT-01). It follows from figure 7 that from 1994 to 2004, mean defoliation of the considered tree species at the first plot, LT-01A, decreased from 30.4% to 22.1%, or an average of 0.61% per year. At LT-01B over the same period, the decrease was from 36.2% to 22.8%, or 1.02% per year, and at LT-01C, from 30.1% to 23.2% or 0.59% per year. Over the last period (from 2005 to 2007), mean defoliation of the monitored trees increased in all plots at LT-01.

The changes in stand defoliation at LT-03 had no regular pattern. Peaks in defoliation were observed in 1997 and 2002, when they exceeded 30% (Fig. 7). During the rest of the period, defoliation fluctuated between 22% and 27%.

The changes in mean defoliation at LT-02 demonstrated a significant trend towards decreasing ($p<0.05$). Between 1994 and 2001 it decreased from 26.7% to 21.6%.

Direct and indirect effects of considered pollutants on tree defoliation

Changes detected in mean defoliation were directly related to changes in air pollutant concentrations and deposition of the acidifying compounds. An analysis of the relationships between pollution and crown defoliation of considered tree species revealed the highest susceptibility of the Scots pines to the impact of air pollution by sulphur compounds (SO_2 and aerosolic SO_4^{2-}) and SO_4^{2-} and NH_4^+ deposition (Fig. 8).

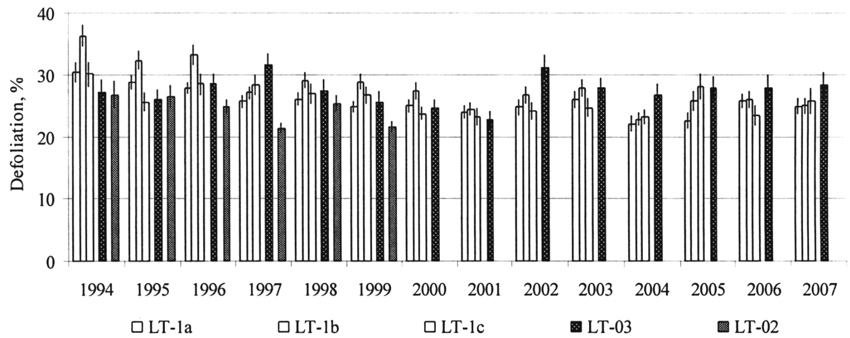

Figure 7. Mean defoliation and standard error of estimation of the considered stands (LT-01 - Aukštaitija IMS; LT-02 - Dzūkija IMS; LT-03 - Žemaitija IMS)

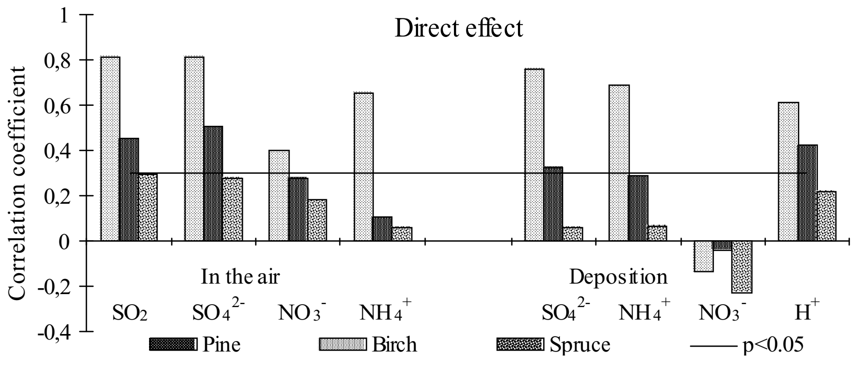

Figure 8. Direct effect of concentrations of considered contaminators in the air, and their deposition on tree defoliation

This fully agrees with ICP Forest Monitoring data (Lorenz and Mues 2007, De Vries et al. 2000a, 2003a, UN-ECE 2005). Changes in crown defoliation of Birch were less related to changes in acidifying compounds, whereas changes in defoliation of Norway spruce were least related, mainly due to damage caused by forest pests, primarily *Ips typographus* L.

An analysis of possible indirect effects of the considered contaminators revealed that pine crown defoliation demonstrated the highest correlation with NH_4^+ concentrations in the soil and ground water, following close to the pH of soil water. The effect of these contaminators on birch defoliation was lower. Only nitrate concentrations both in ground and soil water seemed to have a positive effect on crown condition of these particular tree species. Changes in spruce crown defoliation were least related to the indirect effect of acidifying compounds (Fig. 9).

nificant (r^2=0.355) than the direct effect through the air (r^2=0.258) (Table 2).

The indirect effect of sulphur compounds was not significant. However, its concentrations in surface water and runoff, which reflect chemical processes carried out in the ecosystem, quite well reflects changes in tree crown defoliation.

The detected very week relationships between spruce defoliation and considered pollutants demonstrated no regular pattern. Ips typograpphus had significant impact on vitality of spruce trees over the considered period. Due to their activity about 2 % of the monitored spruce trees died annually. Maximal value reached in 2002 – 4.5%.

Discussion

Nitrogen (N) deposition, meaning inorganic ammonium (NH_4^+) and nitrate (NO_3^-) disrupting the ratio

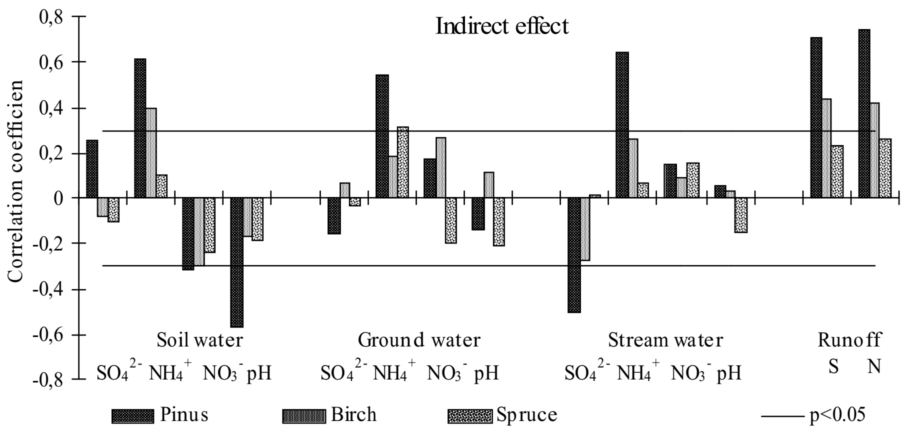

Figure 9. Indirect effect of concentrations of considered contaminators in ground, soil and runoff water on tree defoliation

Contributions of effect character to their integrated impact on tree defoliation

Findings have revealed that the changes in pine defoliation were most significantly related to changes in SO_4^{2-}, NH_4^+ and NO_3^- air concentrations. Deposition of these compounds as well as ozone concentration, did not significantly increase the explanation of pine defoliation variability. Changes in concentrations of the considered compounds in soil and ground water affected pine crown condition remarkably less (Table 2). However, this indirect effect increased the explanation of pine defoliation variability significantly - by 15%, up to 89%.

Controversial results were obtained in explaining changes in the defoliation of birch trees. The highest correlation was detected between birch defoliation and N concentration in ground water, followed closely by NO_3^- concentration in the air and NH_4^+ deposition. The results revealed that the indirect effect of N compounds through soil and ground water was more sig-

between dissolved organic and inorganic N available to plants, together with surface ozone and sulphur air concentrations and their deposition, are among key factors resulting in spatial and temporal changes in the condition of forest ecosystems. While the anthropogenic emissions of SO_2 have declined in Europe by 67 % since the early 1980s due to enactment of strict pollution control strategies, the NO_x and NH_3 emissions have only been reduced by approximately 20% (EMEP 2004). Therefore, even after complete implementation of the Gothenburg Protocol and other current legislation, the effect of N deposition with commensurate adverse biological effects still remains the most relevant problem to confront in Europe as well as in the USA and Canada (Wright et al. 2005).

Ambient ozone (O_3) is considered to be one of the most important and pervasive phytotoxic agents, the effects of which are likely to increase in the future (Krupa and Manning 1988, Hutunnen et al. 2002, Percy et al. 2003, Vingarzan 2004), not only in the South-

Table 2. Contribution of different pollutant compounds in an integrated impact on pine, birch and spruce crown defoliation

Variables		Models, F(a.b)																				
		Pinus							Spruce							Birch						
		3.43	2.44	2.44	3.43	2.44	5.41	9.37	1.51	2.50	2.50	3.49	2.50	4.48	5.47	1.54	2.53	2.53	3.52	3.52	4.51	8.47
In the air:	O_3																					
	SO_4	+						+								+						+
	SO_2	+							+						+							
	NH_4	–						–														–
	NO_3																					
Deposition:	SO_4		+					+		+							+					
	NH_4																					
	NO_3		–					–		–							–					–
In soil water:	pH			–			–							-	–							
	SO_4																					
	NH_4			+			+	-			+							+			+	+
	NO_3										–							–			–	–
In ground water:	deep				–		+	+				–		–	–				+		+	+
	pH				–		-	–											–		–	–
	SO_4																					
	NH_4				+		+	+				+		+					+			
	NO_3											–		–	–							
In stream water:	pH																					
	SO_4												+									
	NH_4																			+		
	NO_3					–														–		–
Runoff:	S																					
	N					+		+					+		+					+		
r^2 ,		0.747	0.662	0.560	0.496	0.560	0.687	0.897	0.094	0.101	0.060	0.296	0.104	0.473	0.502	0.258	0.150	0.206	0.355	0.234	0.387	0.690
p <		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.026	0.071	0.177	0.001	0.063	0.000	0.012	0.000	0.013	0.002	0.000	0.001	0.000	0.000

Note: F(a.b) - models identified by F - test symbol with numbers of degrees of freedom: a - of the predictor variables; b - of the observations. Effect on the defoliation: + - relevance; – inverse.

ern parts of Europe (Bussotti et al. 2005, Ferrety et al. 2003, Paoletti 2005, 2006), but also in the Centre and eventually in the Northern parts of Europe (Augustaitis and Bytnerowicz 2008, Bytnerowicz et al. 2004, 2007, Karlsson et al. 2002, 2006, Utrainen and Holopainen 2000). Its combined effects with acid deposition differ significantly from the sum of individual effects due to their complex synergistic or antagonistic interactions (Bytnerowicz et al. 2004). Our earlier data indicated that only the effect of the peak ambient ozone concentrations on Scots pine defoliation in Lithuania is statistically significant (Augustaitis et al. 2007b, 2007c, 2007d), and its effect on pine defoliation is significant but lower than the integrated effect of acidifying compounds in the air and their deposition. Therefore, the primary attention of investigating the direct and indirect effects of pollutants on tree defoliation was focused on the direct and indirect effects of acidifying compounds and nitrogen compounds.

An indirect effect of the considered pollutants was detected through analysis of the relationships between their concentrations in soil, ground and surface water and tree defoliation. Our data revealed that changes in the concentration of NH_4^+ in the soil and ground water could be explained by variation of concentrations in the air, precipitation and deposition (Augustaitis et al. 2008). These concentrations increased with an increase in NH_4^+ deposition level and the concentrations in the air and precipitation. NO_3^- concentrations in soil and ground water were less related to the concentrations in the air, precipitation or deposition. However, these changes often were related to the NH_4^+ concentration in the air, precipitation and deposition, indicating the occurrence of nitrification in the soil. Other authors investigating key factors of changes in soil water obtained similar results (De Vries et al. 2003b).

Despite these earlier derived, very complicated relationships, what is not entirely understood is which of these effects, direct or indirect, is more significant to the forest ecosystem. In Lithuania, in the vicinity of the nitrogen fertilizer plant *"Achema"* over a period of 25 years, the distance over which chemical changes in forest soils were observed became 3-8 fold less than the distance over which emission compounds damaged pine stands (Armolaitis 1998, Armolaitis and Stakenas 2001). These results revealed a more significant effect of N compounds through the air and deposition on the condition of pine stands, than through the soil. The findings of this study revealed the same regularities. Defoliation of pine trees was more related to changes in N concentrations in the air and deposition than in the soil and ground water.

Controversial results were obtained in explaining changes in the defoliation of birch trees. The indirect effect of N compounds through the soil and ground water was more significant than the direct effect through the air. The main reason is that, when air concentration of N compounds and their deposition reach their highest values, birch trees are leafless and direct effects do not occur. It is therefore most likely that their indirect effect through changes in the soil, ground and surface water is more significant than their direct effect.

Only detected key factors resulting in spruce crown defoliation (Table 2) could be attributed to the uncertainties of this study due to very intensive damage of spruce by forest pests, especially *Ips typographus* L.

Generalizing these results, we could state that in most cases, nitrates in the soil and ground water have a positive effect on crown condition as do a higher value of pH of these waters, while ammonia has a negative effect. Notwithstanding this, the direct effect of air concentration of acidifying compounds, reinforced by the effect of surface ozone, was more significant for pine crown defoliation than their indirect effect through the soil and ground water. Needles, which are present on trees all year round, seem to be more efficient aerosol collectors than leaves. This was confirmed by the results of other authors (Blood et al. 1989, Rothe et al. 2002). To test this hypothesis, detailed analysis of the seasonal effect of considered pollutants on pine defoliation is needed.

Conclusions

Changes in the mean defoliation of pine trees were mainly related to changes in air concentration of aerosolic NO_3^-, NH_4^+ and SO_4^{2-}. Deposition of these compounds as well as ozone concentration did not significantly increase the explanation of pine defoliation variability. The indirect effect of acidifying compounds in the soil and ground water affected pine defoliation remarkably less; however, this indirect effect increased the explanation of pine defoliation variability significantly by 14%, from 75% up to 89%.

Contrary to pine crown defoliation, the indirect effect of N compounds through the soil and ground water on birch defoliation was more significant (r^2=0.355) than the direct effect through the air on leaves (r^2=0.258). Therefore, we could state that needles, which are present on trees all year round, seem to be more efficient aerosol collectors than leaves. The death of spruce trees due to *Ips typographus* L., prevented completion of this task.

In most cases, the considered contaminators had a negative effect on crown condition, while nitrate - its deposition and concentration in the soil and groundwater - had a positive effect.

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НЕПОСРЕДСТВЕННОЕ И КОСВЕННОЕ ВЛИЯНИЕ РЕГИОНАЛЬНОГО ЗАГРЯЗНЕНИЯ НА ДЕФОЛИАЦИЮ КРОН ДЕРЕВЬЕВ

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Резюме

Изменения состояния крон деревьев чаще всего зависят от комплексного влияния натуральных и антропогенных факторов природной среды, где S и N осадки через комбинацию непосредственного влияния атмосферного SO_2, NO_x и NH_4^+ на листву и косвенного влияния на корневую систему, играют предрасположительную, сопроводительную или иногда даже основную роль. Поэтому, в представленной работе мы попытались проверить, которое из влияний является более значительным для состояния крон деревьев. Результаты исследования показали, что непосредственное влияние атмосферного загрязнения является более значительным для состояния крон сосны обыкновенной, чем косвенное влияние через изменения состава почвенной, грунтовой и поверхностной воды. Хвоя, продолжительность которой составляет несколько лет, по-видимому, является более хорошим коллектором аэрозольных соединений, чем листья. Противоположно этому, непосредственное влияние исследуемых загрязнителей, особенно азотных соединений на состояние крон березы, является менее значительным чем их косвенное влияние. В большинстве случаев, кислотные соединения негативно влияют на состояние крон деревьев за исключением только нитратов, влияние которых на состояние крон деревьев положительное. Влияние короедов *Ips typographus* L., впоследствии которых погибло значительное число исследуемых деревьев ели, не позволило подтвердить представленное.

Ключевые слова: кислотные осадки, почвенная, грунтовая и поверхностная вода. дефолиация кроны