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ECOLOGY

Integrated impact of sulphur and nitrogen deposition and ozone on forest ecosystems in Lithuania, 1995–2015

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Abstract. The present study considers temporal variability in concentrations of the throughfall fluxes and bulk fluxes of sulphate, nitrate, and ammonium in precipitation in Lithuania evaluated employing a non-parametric Mann-Kendall test along with Sen's slope estimator during the period 1995–2015. Decreased air concentrations of sulphur species and ammonium as well as their deposition were the main drivers of the reduction of mean defoliation of Scots pine trees in Lithuania. Their effect on Norway spruce crown defoliation was less expressed, but no effect of acidifying species and surface ozone was detected on birch tree crown defoliation. The established relationships revealed that a deterioration of the health of coniferous tree species, first of all Scots pine, could be expected only in forest where the deposition of the acidifying species exceeded the critical loads.

Key words: sulphur deposition, nitrogen deposition, ozone, crown defoliation, critical load.

INTRODUCTION

Scientific studies have associated exposure to ambient pollutants with a variety of problems (Kumar et al., 2014). In most cases effects of environmental factors on the tree growth and crown condition in natural forests are observed through direct impacts of sulphur dioxide (SO_2), nitrogen oxides (NO_x), and ammonia (NH_3) on foliage and soil-mediated impacts on roots (Dornelas et al., 2014; Loehle et al., 2016). It is well recognized that forest ecosystems are influenced by atmospheric deposition of sulphate (SO_4^{2-}) and nitrogen (N) resulting from acid precursors (SO_2, NO_x, and NH_3) emitted by natural and man-made sources since the 1950s. Forest monitoring networks in Europe initiated in 1986 and 1994 after a 'Pan-European Programme for Intensive and Continuous Monitoring of Forest Ecosystems' started contributing to a better understanding of causal relationships between the condition of forest ecosystems and abiotic stress factors (Reis et al., 2012). Tree defoliation data provided by European forest ecosystem monitoring networks have led to the publication of numerous studies conducted in various countries such as Finland (Nevalainen et al., 2010), France (Ferretti et al.,

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2014), Spain (De la Cruz et al., 2014), Italy (Pollastrini et al., 2016), and Lithuania (Augustaitis et al., 2018).

In North European forested regions, trees have suffered an irreversible damage due to air pollution (Juknys et al., 2014). Therefore, temporal trends in acidic deposition of both S and N are of extensive attention as such pollution causes various changes in forest ecosystems (Rizzetto et al., 2016). A similar but opposite change of surface ozone (O_3) concentrations demonstrated a stable trend towards increasing in many countries of Central and Southern Europe (Paoletti et al., 2014). In Lithuania, the O_3 level is strongly affected by local fresh emissions and long-range transport of O_3 and its precursors from the sources in south-western Europe (Girgždienė et al., 2009). Recent studies have indicated that despite the fact that high-level O_3 episodes (>150 μg m^{-3}) are seldom observed, elevated O_3 concentrations remain potentially harmful for forest vegetation in Lithuania (Augustaitis et al., 2007).

Despite the newest approach to the O_3 effect on forest through its flux evaluation (Sicard et al., 2016), a critical loads approach to other air contaminants has been widely used in Europe. The latter is a practical tool that allows researchers to provide evidence of the effects of air pollution exposure to shape a new strategy towards improving the air quality in the European Union.

Climate change and new threats can significantly reduce the adaptive capacity of prevailing in Lithuania tree species to mitigate the main threats of global changes. Therefore, new forest treatments are needed at forest sites where S and N depositions can exceed their critical loads. The new national research programme initiated by the Lithuanian Council of Research is mainly addressed to enhance the sustainability of the ecosystem in Lithuania, including the forest ecosystem. New possibilities allow researchers to gain a better insight into the problem of forest response to changing environmental conditions with a special view on the ecophysiological reaction of trees to unfavourable environmental conditions including meteorology, acidifying compounds, and surface ozone. It will become possible to fulfil the main tasks of the programme and to present new forest management approaches to enhance the sustainability of the forest ecosystem under the pressures of global changes. It should be noted that elevated aerosol concentrations can influence leaf temperatures and transpiration rates (Baldocchi et al., 2002).

The objective of this study was assessment of long-term trends in bulk precipitation and throughfall and of bulk concentrations of SO_4^{2-}, NO_3^-, NH_4^+, and O_3 at International Cooperative Programme (ICP) Integrated Monitoring Stations (IMSs) in Lithuania during the period 1995–2015. The aim was to establish trends by applying the Mann–Kendall and Theil–Sen approaches (see Sicard et al., 2016), their significance, and effects on mean defoliation of the prevailing in Lithuania tree species: Scots pine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* Karst.), and birch (*Betula pendula* 'Crispa' and *B. pubescens* Ehrh.). On the basis of the obtained results in deposition changes and by applying the Simple Mass Balance model, spatial estimates of the critical loads for sulphur and nitrogen deposition in Lithuania were performed to prepare new strategies to increase forest sustainability under pressures of global changes.

METHODS

Sampling site, sampling, and analyses

The study was carried out at the Aukštaitija (LT01) and Žemaitija (LT03) IMSs (Fig. 1). Continuous sampling of the bulk precipitation and throughfall has been performed at these IMSs since 1994. At the Aukštaitija IMS forests cover 70% of the site area where Scots pine stands prevail, while at the Žemaitija IMS, Norway spruce dominates. Two factors, thoroughfall flux and bulk deposition flux, both widely used to study vegetation conditions in ecosystems, were applied for describing and classifying the studied forest ecosystems.

At each site five throughfall collectors (on monthly basis) were located randomly within the forest stand at 10 m intervals with one precipitation sampler at a nearby unforested area. Precipitation was sampled with a continuously opened collector. It consisted of a polyethylene funnel with a collecting area of 314 cm^2 connected to a 5 L polyethylene vessel. Larger collectors with a collecting area of 405 cm^2 were used during winter. The same samplers were used for throughfall. At each site, amounts of precipitation collected by individual samplers were measured individually, and then were combined into one integrated sample. Water samples were kept at 4 °C until laboratory analysis. Later, after filtering through a 0.45-μm membrane filter, nitrate (NO_3^-), sulphate (SO_4^{2-}), and chloride (Cl^-) concentrations were determined with ion chromatography (Dionex). Ammonium (NH_4^+) was analysed spectrophotometrically, and calcium (Ca^{2+}), magnesium (Mg^{2+}), potassium (K^+), and sodium (Na^+) were determined by flame atomic absorption spectrophotometry (FAAS). To determine the pH and electrical conductivity of precipitation samples a pH meter including a pH glass combination electrode and a conductivity meter were used.

The concentration of ozone was measured continuously by an O341M ozone analyser based on the principle of ultraviolet absorption.

Fig. 1. Location of the Aukštaitija (LT01) and Žemaitija (LT03) stations.

MAKESENS

A non-parametric MAKESENS (Mann–Kendall test and Sen's slope estimate method) was applied to investigate annual mean concentrations and throughfall fluxes of SO_4-S, NO_3-N, and NH_4 and to determine the monotonic trend over 1995–2015 at different statistical significance levels ($\alpha = 0.001$, 0.01, 0.05, and 0.1) (Bari et al., 2016). The Theil–Sen approach was applied to estimate the magnitude of the rate of change per year (Sen et al., 1968).

Crown defoliation

Crown defoliation of the three tree species prevalent in IMSs – Scots pine, Norway spruce, and birch – were assessed on vegetation plots for the 1995–2015 period. The tree crown condition of about 600 trees (spruce 50%, pine 30%, and birch 20%) was assessed annually, employing the methodology of the ICP Forest Monitoring Programme over the period 1994–2015 (UNECE ICP FORESTS, 2016).

Critical loads

Critical loads of total nitrogen were calculated by the Simple Mass Balance (SMB) model where the following formula is used for the mass balance of total N:

$$N_{dep}+N_{fix}=N_{AD}+N_i+N_u+N_{de}+N_{eros}+N_{fire}+N_{vol}+N_{le}, \quad (1)$$

where N_{dep} is the total N deposition, N_{fix} is the N 'input' by biological fixation, N_{ad} is the N adsorption, N_i is the long-term net immobilization of N in soil organic matter, N_u is the net removal of N harvested by vegetation and animals, N_{de} is the flux of N to the atmosphere due to denitrification, N_{eros} is the N losses through erosion, N_{fire} is the N losses in smoke due to (wild or controlled) fires into the atmosphere, N_{vol} is the N losses to the atmosphere via NH_3 volatilization, and N_{le} is leaching of N below the root zone.

Annual critical loads and total deposition (dry and wet) values of oxidized S and N and nutrient N were mapped on 50×50 km^2 EMEP grid taking into account the annual mean temperature, annual precipitation, and soil map.

RESULTS AND DISCUSSION

Changes in acid deposition

Long-term trends in annual emissions, atmospheric deposition fluxes, and deposition throughfall and bulk concentration of nitrogen, ammonia, and sulphur emissions from all sources in Lithuania totalled 45, 18, and 29 Gg yr^{-1}, based on the 2018 inventory under CLRTAP.

Total NO_x emissions decreased 62% from 120 Gg in 1990 to 45 Gg in 2015. The road transport sector is the major source of NO_x emissions, contributing ~33% (26 Gg) of the national total amount in 2015. The contribution of the electricity and heat production in 1990 to the national total NO_x emissions decreased by 9.7% to 4.2 Gg in 2015 as a result of the decreases in fuel consumption due to the economic crisis. Total sulphur dioxide emissions decreased by 89.5%, from 173 Gg in 1990 to 18 Gg in 2015. The public electricity and heat production sectors remained the principal (58%) sources of SO_2 emissions in 2015. The large reduction of sulphur emissions occurred due to desulphurization processes and use of fuels with lower content of sulphur such as natural gas. Almost all emissions of NH_3 result from agricultural activities (94%) such as manure application for soil fertilization (28%). Various chemical and physical processes affect how much of these emissions eventually get deposited to surface ecosystems through either wet or dry deposition.

Over 1995–2015, throughfall and bulk deposition fluxes decreased by approximately 70–77% (from 813 to 147 mgS m^{-2} yr^{-1} in throughfall fluxes and from 652 to 149 mgS m^{-2} yr^{-1} in bulk deposition fluxes) at the Aukštaitija IMS, while the annual values of throughfall fluxes for SO_4^{2-} decreased from 1663 to 429 mgS m^{-2} yr^{-1} and bulk deposition fluxes from 786 to 237 mgS m^{-2} yr^{-1} at the Žemaitija IMS (Table 1). This significant decrease ($p < 0.001$ level of significance) in annual SO_4^{2-} (by 27.73 mgS m^{-2} yr^{-1} for throughfall and by 18.01 µgS m^{-2} yr^{-1} for bulk deposition) was most likely the result of a reduction in SO_2 emissions in Europe, including Lithuania. Starting in the late 1980s, SO_4 concentrations in bulk precipitation decreased in most catchments in Lithuania until the year 2000 (Juknys et al., 2014). Changes in annual throughfall and bulk deposition fluxes had a very similar pattern to that of the volume weighted concentration of SO_4 in throughfall and bulk deposition in both sites. The SO_4^{2-} concentration in throughfall for the same period decreased by 80% (from 1.64 to 0.32 mgS L^{-1}) at the Aukštaitija IMS and by 81% (from 3.29 to 0.61 mgS L^{-1}) at the Žemaitija IMS in line with both emission reductions (LRTAP Convention, 2018) over the last 20 years.

A decrease in the annual deposition of NH_4^+ from 389 to 57 mgN m^{-2} yr^{-1} (85%) at the Aukštaitija IMS and from 397 to 88 mgN m^{-2} yr^{-1} (78%) at the Žemaitija IMS was observed during 1995–2015. The most significant decrease in the ammonium concentration in throughfall lasted until 2001, bringing about an 86% reduction in this concentration at the Žemaitija IMS (0.83 to 0.29 mgN L^{-1}) and a 77% reduction at the Aukštaitija IMS (from 0.68 to 0.40 mgN L^{-1}). During the 2001–2005 period a stabilization of the NH_4^+ concentration in throughfall and bulk deposition at 1.1–1.3 mgN L^{-1} at both sites was observed. The annual means of the NO_3^- concentration in throughfall and bulk deposition were stable at 0.44–0.47 µgN m^{-3} at all the stations considered.

The difference between the critical loads for acidity and the total depositions of SO_4-S, NO_3-N, and NH_4-N was calculated, whose negative values represent exceedances of the critical load (Fig. 2). The calculated differences of critical loads and deposition of oxidized sulphur (–398 to 8500 eq·$ha^{-1}yr^{-1}$) are shown in Fig. 2. Critical loads of acidity for terrestrial ecosystems are defined in units of 'equivalents' (ionic charge × moles).

As can be seen, critical loads of oxidized sulphur were mostly exceeded in the southern, southwestern, and small northern parts of Lithuania. The obtained data revealed that the critical load of NO_3-N was not exceeded over the whole territory of Lithuania. The highest exceedances of critical loads of NH_4-N were calculated for the southern part of Lithuania and the lowest for the northern parts of Lithuania. Exceedance of the critical loads of sulphur and ammonium deposition pose the main risk for the forest ecosystem in Lithuania.

Relationships between acid deposition and crown defoliation

Data collected since 1995 on the mean defoliation of the prevailing in Lithuania tree species revealed differences in their statistical significance on temporal and spatial scale ($p < 0.05$). The worst condition of coniferous tree species was recorded at the beginning of the investigation in 1996–1997; afterwards the mean defoliation value fell, and within an equally short period their increase was observed. No reliable trend in the crown defoliation of birch trees was detected.

From 1995 to 2015, the mean defoliation of pine trees decreased on average by 0.24% per year (Fig. 3). Over the same period, the decrease at the Žemaitija IMS was 0.22% per year. The detected temporal changes in the mean defoliation of pine trees were quite common throughout most of Europe (Lorenz and Mues, 2007).

Table 1. Throughfall and bulk deposition fluxes and throughfall and bulk deposition concentrations of SO_4, NO_3, and NH_4, O_3 concentration, throughfall, and precipitation (bulk) in the European Monitoring and Evaluation Programme stations LT01 and LT03, 1995–2015

	n	Test Z	*p*	Q	Q min99	Q max99	Q min95	Q max95	B	B min99	B max99	B min95	B max95
SO_4 fluxes, mgS m^{-2} yr^{-1}													
LT01 Throughfall	21	-5.10	***	-27.73	-40.32	-16.72	-36.82	-19.05	898.05	1142.88	651.27	1100.82	690.80
Bulk deposition	21	-4.80	***	-18.01	-25.30	-12.13	-23.99	-13.12	636.18	781.18	518.15	753.74	533.94
LT03 Throughfall	21	-4.44	***	-50.52	-78.02	-31.60	-70.72	-39.46	1940.2	2500.34	1602.74	2383.45	1721.90
Bulk deposition	21	-4.44	***	-26.18	-35.52	-14.54	-31.46	-18.52	938.60	1088.71	685.11	1037.66	769.98
NO_3 fluxes, mgN $m^{-2}yr^{-1}$													
LT01 Throughfall	21	-4.50	***	-9.04	-13.41	-5.85	-12.06	-6.92	340.07	416.03	273.62	392.01	299.43
Bulk deposition	21	-3.38	***	-6.06	-10.26	-1.71	-8.93	-3.21	346.68	404.91	255.21	390.27	287.04
LT03 Throughfall	21	-2.02	*	-9.39	-21.42	2.37	-17.60	-0.15	520.67	724.85	298.48	659.01	346.40
Bulk deposition	21	-1.84	+	-4.97	-11.95	3.17	-9.78	0.60	456.24	585.20	304.75	550.40	338.17
NH_4 fluxes, mgN $m^{-2}yr^{-1}$													
LT01 Throughfall	21	-4.14	***	-9.84	-20.97	-4.67	-19.00	-5.90	312.65	544.28	198.39	506.93	222.30
Bulk deposition	21	-2.54	*	-9.41	-18.37	0.44	-15.64	-2.09	455.96	617.87	257.66	570.96	289.03
LT03 Throughfall	21	-2.20	*	-10.33	-30.37	1.57	-22.58	-2.28	415.26	793.82	212.19	674.41	286.46
Bulk deposition	21	-0.66		-2.79	-22.22	7.09	-16.77	4.33	402.54	819.16	212.49	710.15	272.04
SO_4 concentration, mgS L^{-1}													
LT01 Throughfall	21	-5.01	***	-0.05	-0.08	-0.03	-0.07	-0.04	1.73	2.22	1.24	2.02	1.38
Bulk deposition	21	-5.04	***	-0.03	-0.04	-0.02	-0.04	-0.02	0.94	1.21	0.77	1.14	0.81
LT3 Throughfall	21	-4.08	***	-0.11	-0.17	-0.06	-0.15	-0.06	3.60	4.85	2.50	4.37	2.69
Bulk deposition	21	-4.92	***	-0.03	-0.05	-0.02	-0.04	-0.02	1.05	1.38	0.82	1.29	0.91
NO_3 concentration, mgN L^{-1}													
LT01 Throughfall	21	-4.02	***	-0.02	-0.03	-0.01	-0.02	-0.01	0.64	0.80	0.51	0.74	0.54
Bulk deposition	21	-3.17	**	-0.01	-0.02	0.00	-0.02	0.00	0.53	0.68	0.39	0.65	0.42
LT03 Throughfall	21	-2.81	**	-0.02	-0.05	0.00	-0.04	0.00	0.91	1.31	0.53	1.22	0.58
Bulk deposition	21	-2.72	**	-0.01	-0.02	0.00	-0.02	0.00	0.61	0.80	0.41	0.77	0.45
NH_4 concentration, mgN L^{-1}													
LT01 Throughfall	21	-4.32	***	-0.02	-0.04	-0.01	-0.03	-0.01	0.56	1.11	0.36	0.87	0.40
Bulk deposition	21	-1.96	*	-0.01	-0.03	0.00	-0.02	0.00	0.65	0.96	0.32	0.87	0.41
LT03 Throughfall	21	-2.33	*	-0.01	-0.06	0.00	-0.05	0.00	0.65	1.50	0.29	1.30	0.42
Bulk deposition	21	-1.84	+	-0.01	-0.03	0.00	-0.03	0.00	0.57	1.03	0.30	0.95	0.39
O_3 concentration, µg m^{-3}													
LT01	21	-0.33		-0.10	-0.55	0.47	-0.41	0.36	55.50	66.34	41.72	62.77	44.71
LT03	19	-1.51		-0.23	-0.67	0.11	-0.51	0.06	53.83	61.80	48.61	59.07	43.25
Throughfall, mm													
LT01	21	0.21		2.53	-9.72	14.72	-6.92	11.33	483.1	725.60	329.46	674.99	373.06
LT03	21	1.12		5.86	-12.50	24.52	-7.91	18.73	563.16	931.00	213.20	842.53	329.74
Precipitation (bulk), mm													
LT01	21	-0.03		-0.19	-10.58	14.40	-8.77	11.34	638.3	824.47	451.54	798.82	493.63
LT03	21	1.90	+	10.18	-4.40	25.60	-0.18	21.80	718.36	1031.84	410.87	953.21	497.71

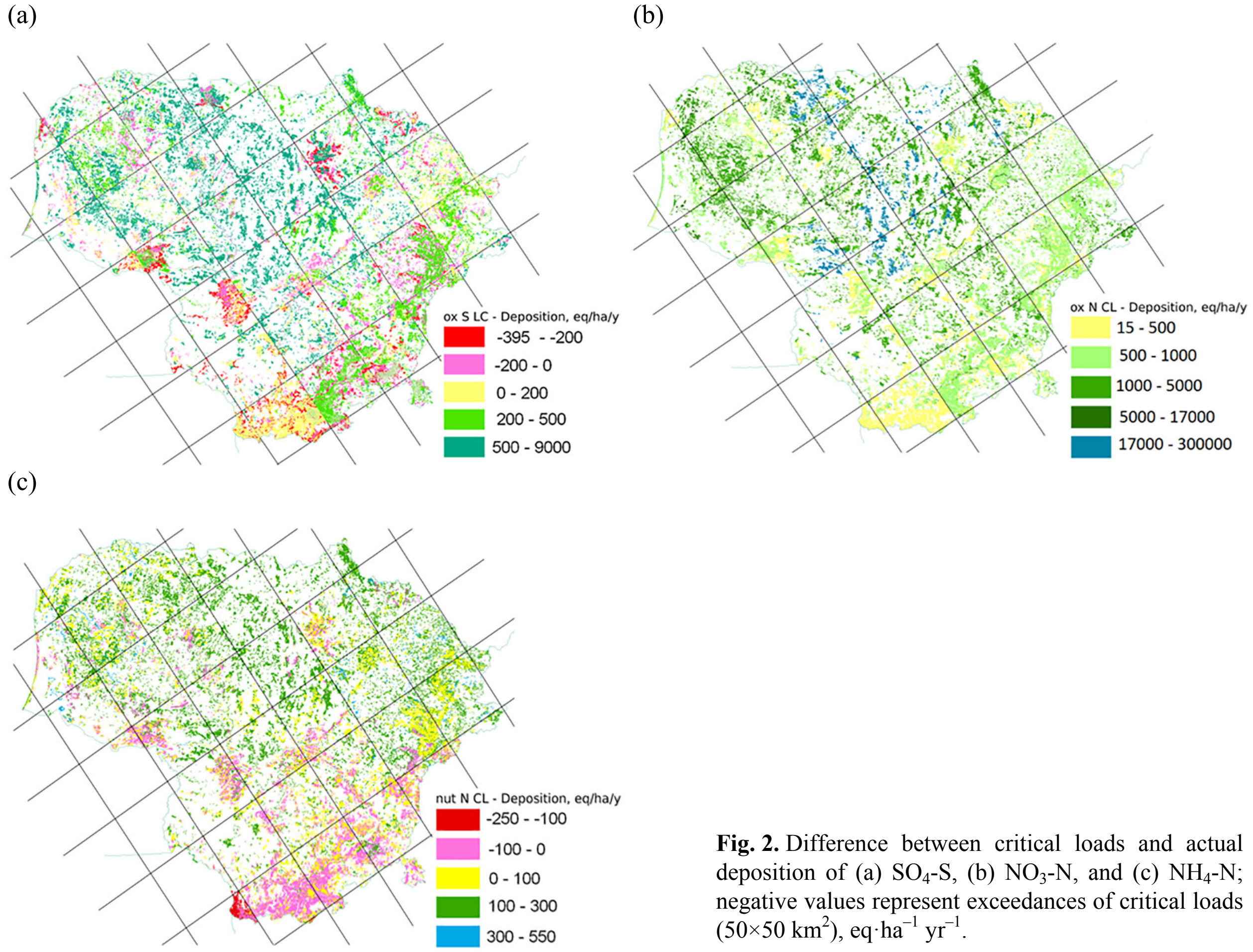

Fig. 2. Difference between critical loads and actual deposition of (a) SO_4-S, (b) NO_3-N, and (c) NH_4-N; negative values represent exceedances of critical loads (50×50 km^2), eq·ha^{-1} yr^{-1}.

Through the 1995–2008 period the mean crown defoliation of spruce trees varied almost equally at both investigation sites (Fig. 3) with the highest level of the mean defoliation of spruce trees observed in 1996 (31.98% at the Aukštaitija IMS and 31.42% at the Žemaitija IMS). The highest differences in the variation of the mean crown defoliation of spruce trees between the two sites started in 2009 (Fig. 3). During the 2009–2015 period the mean defoliation levels of spruce crowns were higher at the Aukštaitija IMS with a reduction from 29.67% in 2011 to 22.08% in 2015 (Fig. 3). At the Žemaitija IMS the mean crown defoliation levels of spruce trees were lower but more stable. There the level was lower not only during the 2009–2015 period, but also during the entire investigation period, varying from 19.19% in 2015 to 23.28% in 2011. However, the mean crown defoliation levels were the highest for Norway spruce trees compared to the other investigated species during the entire 1995–2015 period (Fig. 3), except for the mean crown defoliation of birch trees at the Žemaitija IMS in 2007, when the highest defoliation level of 35.64% was recorded (Fig. 3).

The highest level of mean crown defoliation of birch trees at the Aukštaitija IMS was observed in 2010 (25.00%). Birch trees at the Žemaitija IMS showed a better condition compared to the Aukštaitija IMS during the most of the investigation period as illustrated in Fig. 3, except the extreme worsening of the conditions at the Žemaitija IMS in 2007 and in 1998, 2000, 2005–2006, and 2009 when the conditions were slightly better at the Aukštaitija IMS. During the 2010–2014 period the condition of birch crowns demonstrated stability, and only in 2015 a notable improvement at both investigation sites was observed, with the overall lowest mean defoliation levels of 13.63% at the Aukštaitija IMS and 11.80% at the Žemaitija IMS (Fig. 3).

The largest variability of mean crown defoliation levels of birch trees between the two investigation sites during the same periods indicates that deciduous trees could be more sensitive to the changes of the local environmental and meteorological conditions than coniferous species. We quantified the contribution of the predictor variable, ‘peak ozone concentration’, to the integrated impact of different combinations of natural and

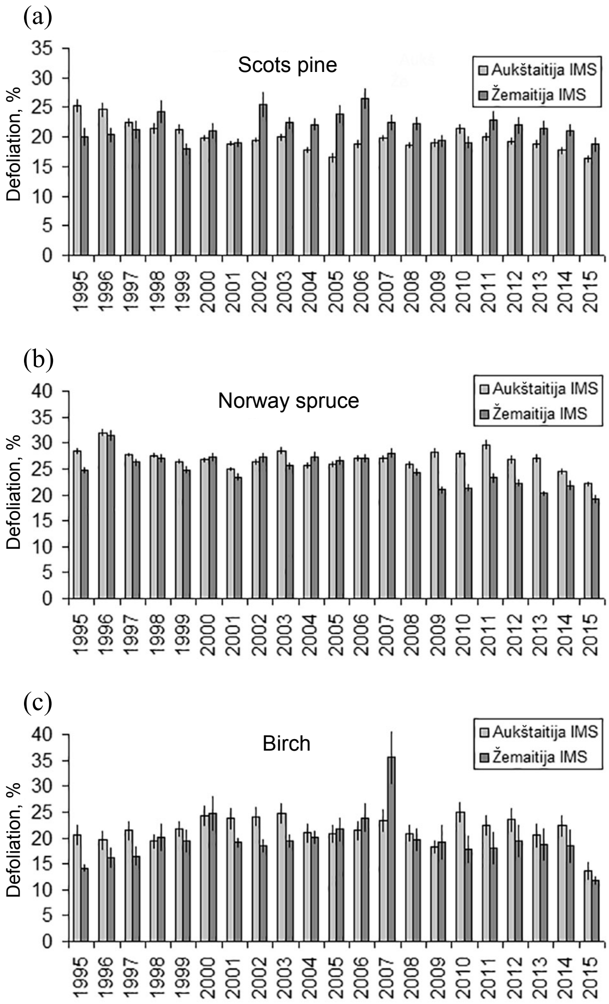

Fig. 3. Crown defoliation of Scots pine, Norway spruce, and birch trees during the 1995–2015 period, %.

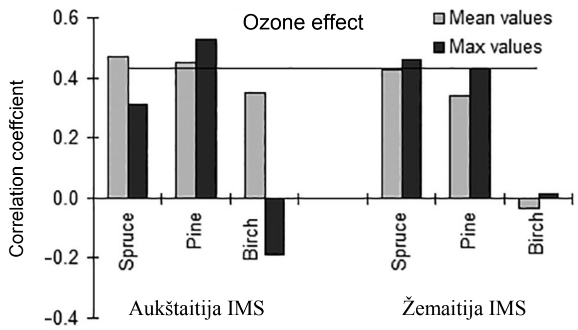

Fig. 4. Relationship between the mean and maximal values of surface ozone and crown defoliation of the prevailing in Lithuania tree species.

anthropogenic factors on Scots pine, Norway spruce, and birch species crown defoliation (Fig. 4).

This index was shown to contribute most significantly to the changes in different components of forest biota. The most significant direct effect of maximal values of ozone was detected on pine defoliation at the Aukštaitija IMS and on spruce defoliation at the Žemaitija IMS. Norway spruce is the tree species that generally prevails at the considered IMSs. A significant effect of these ozone concentrations was detected also on pine crown defoliation at the Žemaitija IMS. This coincides with the results obtained by Augustaitis et al. (2007) for the 1994–2005 period. No significant effect of maximal values of surface ozone on birch crown defoliation was observed at either IMS.

Mean values of surface ozone of similar level had a direct effect on crown defoliation of both considered coniferous tree species at the Aukštaitija IMS, whereas at the Žemaitija IMS only spruce trees were affected. This finding corroborates the state of knowledge in the field that *Picea abies* is one of the most sensitive tree species to the effect of surface ozone. The effect of mean values of ozone on crown defoliation of birch trees was close to the level of significance at only the Aukštaitija IM station.

In contrast, there was no statistical evidence that surface ozone could have had a significant effect on birch crown defoliation. It is known that ozone can cause a wide range of symptoms: increased respiration, mineral nutrient deficiencies, a decrease in the foliar chlorophyll content, and development of necrotic spots causing a reduction in photosynthesis (Utrainen and Holopainen, 2000) and growth and productivity (Matyssek and Innes, 1999). However, the intensity of such damage primarily depends on the concentration level, time of exposure, and uptake through the open stomata (Matyssek et al., 2015).

The ability of birch species to maintain a high midday leaf water potential by reducing their crown-level stomatal conductance with the decrease in soil water availability could be evaluated as the highest adaptive capacity to reduce the uptake of ozone at its highest mid-day concentrations through the stomata. Gradual reduction in leaf stomata density on a single birch tree as result of acclimation to extreme changes in environmental conditions also strengthened this preventative capacity against damage by high levels of ozone. The negative effects of ozone mostly affect the relative growth rate of birch trees, especially young ones. Most probably it is why no regular pattern was observed when investigating the effect of ozone on crown defoliation of birch trees. Analysis of the relationships between environmental contaminants and crown defoliation of Scots pine, Norway spruce, and birch trees revealed the highest susceptibility to the impact of air pollution by sulphur and nitrogen compounds (Table 2).

Table 2. Matrix in Pearson correlation coefficients between the mean defoliation of the prevailing in Lithuania tree species and the considered air pollutants, 1994–2015

		LT01	LT03	LT01	LT03	LT01	LT03
Scots pine (*Pinus sylvestris* L.) crown defoliation, %							
		SO_4, mgS m^{-2} yr^{-1}		NO_3, mgN m^{-2} yr^{-1}		NH_4, mgN m^{-2} yr^{-1}	
Throughfall fluxes		0.70	0.79	0.72	0.48	0.79	0.69
Bulk deposition fluxes		0.73	0.67	0.40	0.02	0.69	0.55
		SO_4, mgS L^{-1}		NO_3, mgN L^{-1}		NH_4, mgN L^{-1}	
Concentration	Throughfall	0.73	0.66	0.65	0.57	0.76	0.55
	Bulk	0.75	0.63	0.37	0.15	0.69	0.50
Spruce (*Picea abies* L.) crown defoliation, %							
		SO_4, mgS m^{-2} yr^{-1}		NO_3, mgN m^{-2} yr^{-1}		NH_4, mgN m^{-2} yr^{-1}	
Throughfall fluxes		0.34	0.56	0.21	0.13	0.65	0.47
Bulk deposition fluxes		0.37	0.53	0.03	0.05	0.63	0.35
		SO_4, mgS L^{-1}		NO_3, mgN L^{-1}		NH_4, mgN L^{-1}	
Concentration	Throughfall	0.37	0.67	0.12	0.49	0.66	0.59
	Bulk	0.33	0.71	–0.12	0.31	0.67	0.57
Birch (*Betula* L.) crown defoliation, %							
		SO_4, mgS m^{-2} yr^{-1}		NO_3, mgN m^{-2} yr^{-1}		NH_4, mgN m^{-2} yr^{-1}	
Throughfall fluxes		0.05	–0.12	–0.05	–0.27	–0.36	–0.32
Bulk deposition fluxes		–0.11	–0.28	0.15	–0.11	–0.26	–0.32
		SO_4, mgS L^{-1}		NO_3, mgN L^{-1}		NH_4, mgN L^{-1}	
Concentration	Throughfall	0.04	–0.16	–0.09	–0.25	–0.36	–0.28
	Bulk	–0.11	–0.27	–0.08	–0.13	–0.38	–0.27

The highest statistically significant correlation of 0.75 was observed between Scots pine defoliation and SO_4^{-2} concentration at the Aukštaitija IMS. At the Žemaitija IMS, SO_4^{-2} fluxes and concentrations were among the key contaminants negatively affecting the pine crown condition. The effect of NH_4^+ concentrations and their fluxes on the condition of pine trees was quite similar to that detected for SO_4^{-2} values. Only higher concentrations of NO_3^- in throughfall and their fluxes stimulated crown defoliation significantly. Bulk deposition of these contaminants had no significant effect on changes in crown defoliation. All defoliation levels increased directly with increasing concentrations and fluxes in throughfall and bulk deposition of the considered acidifying compounds. Detected relationships allow us to state that Scots pine is one of the most sensitive tree species to the effects of acidifying compounds.

The highest statistically significant relationship was observed between Norway spruce crown defoliation and SO_4^{-2} concentrations both in throughfall (0.67) and bulk (0.71) as well as between spruce defoliation and NH_4^+ concentrations (from 0.63 to 0.67) at the Žemaitija IMS. All other correlations between spruce defoliation and SO_4^{-2}, NO_3^-, and NH_4^+ fluxes and concentrations were low or weak for both investigation sites, while NO_3^- bulk concentration demonstrated even a negative correlation (–0.12). A weak and non-significant relationship was evaluated between birch defoliation with all air pollutants at both IMSs. These data could be presented as a sign of a high tolerance capacity of birch trees to the effect of elevated concentrations and fluxes in throughfall and bulk deposition of the considered acidifying compounds.

CONCLUSIONS

Long-term trends in throughfall and bulk deposition fluxes and concentrations of NO_3-N, NH_4-N, and SO_4-S were analysed using long-term monitoring data from two integrated monitoring sites over a 20-year period (1995–2015). Trends were derived by the Mann–Kendall test. Changes detected in the mean defoliation of Scots pine were directly related to changes in air pollutant concentrations and deposition of the acidifying compounds. Changes in annual NO_3-N, NH_4-N, and SO_4-S fluxes in throughfall and bulk deposition had a very similar pattern to that of the concentration at both sites. The SO_4 concentration in throughfall for the same period at the Aukštaitija IMS decreased by 80% (from 1.64 to 0.32 mgS L^{-1}) and by 81% (from 3.29 to 0.61 mgS L^{-1}) at the Žemaitija IMS in line with both emission reductions over the last 20 years. The detected temporal changes in the mean defoliation of pine trees were rather common across Europe. A significant direct effect of maximal values of ozone was detected on pine defoliation at the Aukštaitija IMS and on spruce defoliation at the Žemaitija IMS where these tree species prevail. The mean values of surface ozone also significantly affected changes in the crown defoliation of pine and spruce trees at the Aukštaitija IMS, whereas at the Žemaitija IMS surface ozone had a significant effect only on the defoliation of spruce trees. The concentration of neither acidifying species nor surface ozone had a significant effect on birch trees crown defoliation. This indicates a high tolerance of this tree species for the negative effect of these contaminants on crown status. The highest critical load values (762 eq·ha^{-1} yr^{-1}) of SO_4-S were calculated for the northern and central parts of Lithuania, while the lowest (16 eq·ha^{-1} yr^{-1}) were for southern parts. Also for NO_3-N the lowest critical load values (from 269 to 1896 eq·ha^{-1} yr^{-1}) were evaluated in the southern part of Lithuania, and the highest for the northern and western parts of Lithuania. The detected relationships revealed that the condition of coniferous tree species should have a similar pattern in spatial changeability as the critical loads of the considered contaminants.

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Väävli ja lämmastiku depositsiooni ning osooni koosmõju metsaökosüsteemidele Leedus, 1995–2015

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On uuritud sulfaatide, nitraatide ja ammooniumi kontsentratsioonide ajalist varieeruvust sademetes ning selle mõju hariliku kuuse, hariliku männi ja kase defoliatsioonile Leedus ajavahemikul 1995–2015. Leiti, et kõige suuremat mõju avaldas väävli ja lämmastiku kontsentratsiooni vähenemine õhus ning samuti nende depositsiooni vähenemine hariliku männi keskmise defoliatsiooni langusele. Mõju hariliku kuuse ladva defoliatsioonile oli väiksem. Samas ei mõjutanud happelisust suurendavad ühendid ega osoon kase defoliatsiooni. Leiti, et okaspuude, eriti hariliku männi tervise halvenemist võib oodata vaid juhul, kui happelisust suurendavate ühendite depositsioon ületab kriitilise koormuse.