

Impact of Meteorological Parameters on Responses of Pine Crown Condition to Acid Deposition at Aukštaitija National Park

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Abstract

Climate changes may affect sustainable forest development, its adaptation to unfavourable environmental conditions and first of all to air concentration of acidifying species. However, it is still unclear whether local meteorological conditions reduce or enhance air pollution effects on forest health. To reduce these uncertainties data set of crown defoliation of 20 Scot pine stands located in north-eastern part of Lithuania and obtained since 1994 was used. The findings revealed that meteorological parameters being less significant in relationships with pine crown condition than acidifying species resulted in their deposition variation. Increase in precipitation amount over the dormant period and decrease over vegetation enhance negative effect of the acidifying species on pine crown condition. However, if climate changes follow the long term tendencies, reduction of the negative effect of air concentrations of acidifying species and their deposition can be expected, despite the rise in negative effect of new threat for forest ecosystem – the increase in deposition of the ammonia compounds.

Key words: climate change, acidifying species, interaction, pine defoliation

Introduction

The variation of forest growth and vitality conditions in relation to natural and anthropogenic factors is of main concern for researchers who are studying the impact of air pollution on forest ecosystems (Paoletti et al. 2003). Long range transboundary pollution and ensuing acidification of the environment were implicated as major causes of forest decline on a regional scale (Knabe 1981, Bauer 1982, Mehne-Jakobs 1990). The findings of our earlier study allowed us to make an assumption, that temporal and spatial changes in pine defoliation are first of all related to air concentrations of acidifying compounds, their deposition and meteorology, while ozone concentrations only reinforced the integrated impact of these factors (Augustaitis et al. 2005, 2007, Augustaitis and Bytnerowicz 2008).

Climate is among key factors comprising the global change threat to forest condition (Percy and Ferretti 2004, Langner et al. 2005, Zlatev and Moseholm 2008, Jacob and Winner 2009). Its effect on the distribution and deposition of air pollutants can alter air quality (Meleux et al. 2007), because chemical reactions

and biogenic emissions are dependent on temperature (Zlatev and Moseholm 2008). Vice versa, pollutants can modify responses of ecosystems to specific climatic change impacts (Bytnerowicz et al. 2007).

Therefore, in the present study, as a working hypothesis we wanted to see if the detected changes in mean temperature and amount of precipitation of different periods through the interaction with air concentrations of acidifying species and their deposition could have had some additional effect on crown defoliation of the prevailing tree species in Lithuania – Scots pine (*Pinus sylvestris* L.) especially under climate change condition.

In order to meet the objectives the following research tasks were carried out:

- an analysis of temporal variation in data on crown defoliation of pine stands;
- an analysis of seasonal variation in data on air pollutant concentrations, their deposition, and meteorological parameters, and their interdependence;
- a correlative and multiple regression analyses of data on pine defoliation versus seasonal changes in meteorology, air concentration of acidifying compounds, and their deposition;

• an estimation of the effects of local meteorological conditions on key air pollutant variations in order to predict pine stand condition in the eastern part of Lithuania.

The Integrated Monitoring Programme, which has been annually performed since 1994 in Lithuania, provides all the necessary data to meet the objectives of the study.

Material and methods

Study sites and their location

The study was based on monitoring data on crown defoliation of more than 3000 Scots pine (*Pinus sylvestris* L.) trees from 20 permanent observation stands (POS) annually obtained between 1994 and 2009 in the Aukštaitija national park (ANP), located in the eastern part of Lithuania (Fig. 1). Over 16yr period mean values of crown defoliation of every sample pine stand were computed annually. Therefore the total number of treatments made N=320. These stands were classified according to stand maturity: 4 sapling stands (45 to 50 years – Table 1, section “Stand maturity”, group 1), 5 middle aged stands (61 – 80 years – group 2), 4 premature stands (81-100 years – group 3), 3 mature stands (101-120 years – group 4) and 4 over mature stands (> 121 years – group 5). Main characteristics of the considered stands are compiled in table 1. All stands represent the prevailing *Pinetum vaccinomyrtilosum* forest type in Lithuania.

Monthly data on air concentrations of sulphur dioxide (SO₂), sulphate (SO₄²⁻), the sum of nitrate species

Table 1. Characteristics of the permanent observation stands (POS) at the beginning of observation (1994)

POS index	Stand and site parameters					
	Stand maturity group	Mean diameter cm	Mean height m	Sum of basal area m ² ·ha ⁻¹	Volume m ³ ·ha ⁻¹	Tree density unit·ha ⁻¹
ANP-01	5	40.4	32.6	39	577	260
ANP-02	4	38.4	29.1	36	479	245
ANP-03	2	18.5	19.6	14	141	422
ANP-04	5	41.4	29.5	31	417	221
ANP-05	2	18.5	19.7	17	173	600
ANP-06	5	42.4	31.0	32	456	220
ANP-07	2	17.8	19.4	17	173	611
ANP-08	5	41.7	29.9	28	380	193
ANP-09	2	21.2	20.9	27	281	521
ANP-10	1	16.8	17.2	20	181	772
ANP-11	3	33.0	27.8	24	310	263
ANP-12	3	32.8	27.0	29	359	297
ANP-13	3	32.9	25.5	25	292	272
ANP-14	1	17.5	14.6	20	148	717
ANP-15	1	18.8	16.7	20	168	656
ANP-16	4	32.4	27.5	22	279	225
ANP-17	2	31.4	27.1	24	308	275
ANP-18	3	27.8	24.5	22	254	336
ANP-19	1	22.1	21.5	30	315	733
ANP-20	4	34.5	28.1	22	285	220

Note: stand maturity group: 1 – sapling stands, 2 – middle aged stands, 3 – premature stands, 4 – mature stands, 5 – over mature stands.

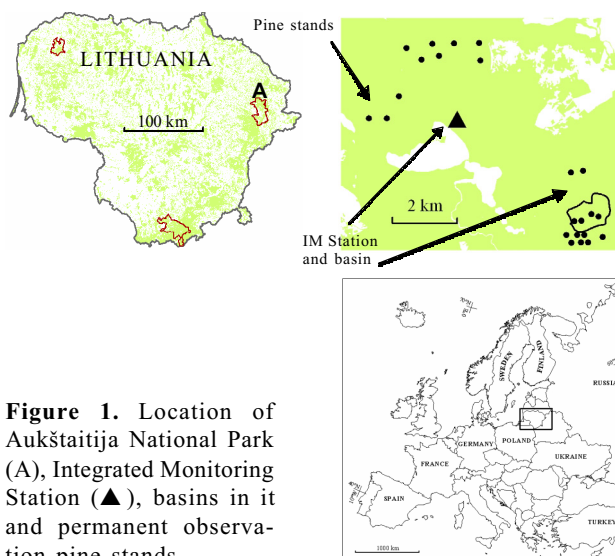


Figure 1. Location of Aukštaitija National Park (A), Integrated Monitoring Station (▲), basins in it and permanent observation pine stands

and ammonium species ($\Sigma\text{NH}_4^+ = \text{NH}_4^+ + \text{NH}_3$) and wet deposition of SO₄²⁻, NO₃⁻, NH₄⁺ as well as general meteorological data were obtained from the Integrated Monitoring Station (IMS) established in this national park (Sopauskiene et al. 2001) and presented in detail in our earlier publications (Augustaitis et al. 2005, 2007, 2010).

Main predictor variables and methods of their estimation

Air pollution and deposition

Air sampling was carried out at weekly intervals over the entire year. The sampling equipment for SO₂ and particulate sulphate consisted of a two-stage filter pack sampler with a cellulose filter (Whatman 40). SO₂ was collected by retention of particles using 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.

Weekly precipitation samples were collected in a polyethylene bulk-collector from December to March (snow collector) 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. The NH₄⁺ concentration in precipitation as well as in the solutions extracted from Whatman 40 filters impregnated with oxalic acid was analysed spectrophotometrically, using the indophenol blue method. Precipitation pH and electrical conductivity were determined with a pH glass electrode and an electric conductance meter, respectively (Sopauskiene and Jasineviciene 2006).

The measurements 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).

Meteorological parameters

The effect of meteorological conditions on pine defoliation was analysed by beginning in autumn of the previous year (in September) and ending in summer of the current year (in August). The quality of the data was assured according to the requirements of the World Meteorological Organization (WMO 1983) and ICP IM methodology (UN-ECE 1993).

Dendrometric parameters

Data on tree and stand parameters (mean diameter and height of trees, tree density, basal area and tree volume per hectare) were available from the permanent observation stands established according to the methodology developed by Lithuanian scientists (Juknys et al. 2003). A three-stage sampling pattern was used for the collection of the field material: (1) sampling of the research stands; (2) sampling of the circular plots within each research stand; (3) sampling of trees for more detailed measurements of the tree crown parameters and tree ring analysis (data are not included). Each pine stand included twelve circular sample plots with an average of 15-20 trees on each plot. Sample plots were distributed systematically in a grid system. Stem diameter and crown defoliation for all sample trees were measured and assessed annually in August, due to which the considered response variables (meteorology and pollutants) were analysed over the period from September of the previous year to August of the current year. Mean annual values of crown defoliation as well as stem diameter were computed for every POS using the data of its sample trees from 12 circular plots. European forest monitoring methodology was employed to assess tree defoliation (UN-ECE 1994).

Statistical methods

The Fisher test was used to test for significant differences in mean defoliation of Scots pine stands between (spatial changes) and within (temporal changes) the stands. The integrated impact of the air pollutants and their deposition on mean defoliation of pine trees was analyzed by a multiple stress approach, using the linear multiple regression technique of Statistica 6.0 software. In order to detect the effect of climate changes on air quality, the relationships between air pollutant concentrations, aqueous pollutant concentrations in atmospheric wet deposition and response meteorological parameters were derived. To detect seasonal effect of meteorological parameters and pollutants on pine condition, and their interdependence, the values of these parameters were computed over different periods:

- i. – over dormant period: a) September-December; b) September-February; c) January-April; d) March-April; e) total September-April;
- ii. – over vegetation – May-August.

The quality of the created models was assessed by determining the coefficient of determination (R^2) and the level of statistical significance (p). Stress factors were excluded from the regression model by a stepwise procedure based on the level of significance of each stress factor. Finally, variables with a high level of significance compiled the models.

Results

Temporal changes in mean defoliation of Scots pine trees

F-test statistics revealed statistically significant differences in spatial and temporal variance of the pine stand mean defoliation within each POS as well as among them (Table 2).

Table 2. Fisher test data on spatial and temporal changes in mean defoliation of Scots pine trees (ANOVA 2 factorial analysis)

Source of variation	Statistical parameters of F - test					
	SS	df	MS	F	F crit	p
Among years	1681,4	15	112,1	40,34	1,702	0.000
Among stands	771,6	19	40,61	14,61	1,623	0.000

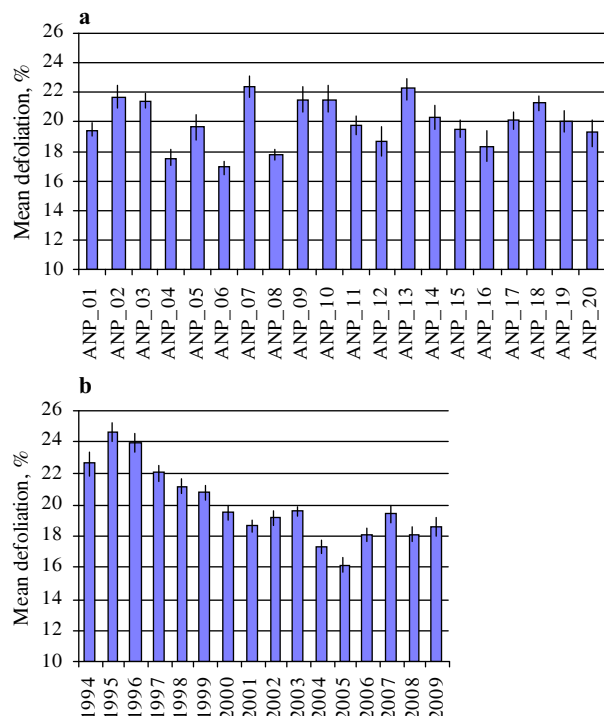


Figure 2. Spatial changes in mean defoliation of each sample stand over the considered period (a, n=20), and temporal changes in annual mean defoliation of all POS (b, n=16) over the entire considered period (Total N=320)

The highest level of mean defoliation of pine trees in the Aukštaitija NP was observed in 1995 (Fig. 2). Thereafter, the crown conditions were gradually improving until 2005, approximately by an average of 1% per year. This trend in pine crown defoliation was statistically significant ($p < 0.05$). Since 2005 crown defoliation increase has been observed approximately on average 0.5% per year ($p > 0.05$).

Seasonal variation of air temperature and precipitation, and their effect on pine defoliation

Mean annual precipitation in the Aukštaitija NP over 16yr period was 705 mm and did not demonstrate significant trend (decrease by 2 mm per year, $p = 0.74$). However, seasonal changes in the precipitation amount were a little more significant. Over September and December it decreased by -3.2 mm per year ($p = 0.21$), over January and April by -4.5 mm per year ($p = 0.15$), and over the vegetation increased by +5.9 mm per year ($p = 0.28$). Despite these changes, between 2004 and 2009 when pine crown defoliation demonstrated a tendency towards increasing, precipitation amount over the vegetation period started decreasing by almost 30 mm per year.

Mean annual temperature for the long term period made +6.7°C and tended to increase between 1994 and 2009 by 0.038°C per year. This increase was in full agreement with the data presented by the SRES A1 B Project (3.5°C per 100 years) (IPCC 2007), and was most pronounced in the first half of the dormant period (September–December) when it made 0.16°C per year ($p = 0.034$). From January to April mean monthly temperature showed a tendency towards decreasing by -0.05 per year ($p = 0.68$) while over the vegetation period (May–August) it was stable ($p = 0.995$). It is highly probable that only changes in temperature over the first half of the dormant period due to their high variation had a considerable effect on pine defoliation changes. Changes in mean air temperature from 2004 to 2009 did not differ considerably from those of the entire considered period.

The direct correlation derived between pine defoliation and precipitation amount in the first half of the dormant period was found to be the strongest, followed by a weaker direct correlation with precipitation in the second half of the dormant period, i.e. from January to April (Table 3). These relationships revealed the improvement of pine stands condition (lower defoliation level) when precipitation amount decreased. Negative correlation was established between defoliation and precipitation during the vegetation period, i.e., from May to August, when more abundant precipitation resulted in a better tree crown condition.

Table 3. Correlative analysis of Scots pine stands defoliation vs. meteorological parameters: precipitation (Pr, mm) and temperature (Tm, °C)

Parameters	Periods						
	IX-XII	I-IV	II-IV	IX-II	III-IV	IX-IV	V-VIII
Precipitation, (mm)							
r	0.334	0.224	0.263	0.272	0.230	0.349	-0.375
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
Temperature (°C)							
r	-0.323	0.056	0.083	-0.199	0.066	-0.138	0.043
p	p=0.000	p=0.314	p=0.137	p=0.000	p=0.239	p=0.013	p=0.446

Note: Effect of pollutants: (+) – direct, (-) – adverse; significant correlation in bold.

Forward stepwise procedure was chosen to derive relationships between Scots pine stands defoliation and meteorology. Created multidimensional model (F1) indicated that precipitation amounts should have explained about 21% variability of the pine stands mean defoliation (F1):

$$F1 = 19.1 - 0.011 \times Pr_{(V-VIII)} + 0.019 \times Pr_{(IX-XII)}; \\ R^2 = 0.207; \quad p < 0.000, \tag{1}$$

where: $Pr_{(V-VIII)}$ – precipitation amount of May–August, mm; $Pr_{(IX-XII)}$ – precipitation amount of September–December, mm.

Changes in temperature over the first half of dormant period, as it was expected, significantly and adversely affected pine defoliation variation indicating that warmer dormant period should have resulted in lower defoliation level (Table 3). Effect of temperature over the rest of periods (second half of the dormant and vegetation periods) was not as significant as those mentioned above. Created multidimensional model (F2) indicated that mean temperature of the different periods explained up to 12 % of mean defoliation variability of the pine stands:

$$F2 = 23.3 - 0.783 \times Tm_{(IX-XII)} + 0.185 \times Tm_{(I-IV)}; \\ R^2 = 0.116; \quad p < 0.000, \tag{2}$$

where: $Tm_{(IX-XII)}$ – mean temperature of September–December, °C; $Tm_{(I-IV)}$ – mean temperature of January–April, °C.

Obtained relationships revealed that the effect of both meteorological parameters (precipitation and temperature) of the dormant period and precipitation amount over vegetation increased the explanation rate of mean defoliation variability of the pine stands up to 27 % (F3 model):

$$F3 = 20.55 + 0.0204 \times Pr_{(IX-XII)} - 0.5780 \times Tm_{(IX-XII)} - 0.0082 \times Pr_{(V-VIII)}; \\ R^2 = 0.269; \quad p < 0.000, \tag{3}$$

where: $Pr_{(V-VIII)}$ – precipitation amount of May-August, mm; $Pr_{(IX-XII)}$ – precipitation amount of September-December, mm; $Tm_{(IX-XII)}$ – mean temperature of September-December, °C.

Decrease in precipitation amount over the dormant period following the increase in mean monthly air temperature from September to December and the increase in precipitation amount over the vegetation period had to ensure a better pine crown condition in future. However, changes in these parameters since 2004 have not confirmed long-term changes. Precipitation over the dormant period had a tendency to increase, meanwhile over the vegetation to decrease. The continuation of these changes will most probably result in decline of pine tree condition.

Seasonal variability of air pollution and acid deposition, and their possible effect on pine defoliation

IMS data showed a significant decrease in main acidifying species until the year 2000. Over the dormant period air concentrations of both sulphur compounds (SO_2 and SO_4^{2-}) decreased by 80% (from 3.7 to 0.7 $\mu gS/m^3$), and their (SO_2 and SO_4^{2-}) wet deposition by 68% (from 500 to 160 mgS/m^2) (Fig. 3). Over the vegetation period air concentration of sulphur species was remarkably lower than that over the dormant period and their decrease was as follows: SO_4^{2-} from 2.6 up to 0.6 $\mu gS/m^3$ and SO_2 from 1.5 up to 0.2

$\mu gS/m^3$. This significant decrease ($p>0.05$) in annual sulphur compounds was most likely the result of a reduction in SO_2 emission in Europe including Lithuania (CLRTAP 2005).

The decrease in ΣNH_4^+ air concentration over this period made up 60% (from 3 to 1.2 $\mu gN/m^3$) and 65% (from 350 to 120 mgN/m^2) in deposition. Changes in NO_3^- deposition were rather stable over the entire considered period and fluctuated around 220 mgN/m^2 . (Sopauskiene and Jasineviciene 2006; Augustaitis et al. 2010).

Between 2000 and 2009 a further decrease in SO_2 air concentrations up to 0.08 $\mu gS/m^3$ over the dormant period and up to 0.05 $\mu gS/m^3$ over the vegetation as well as in aerosol SO_4^{2-} air concentration up to 0.49 $\mu gS/m^3$ was observed. Only aerosol SO_4^{2-} air concentration over the dormant period increased again by approximately 0.02 $\mu gS/m^3$ per year and reached 0.92 $\mu gS/m^3$ level in 2009 (Fig. 3). Wet deposition of sulphur and its concentration in precipitation over the dormant period of these years demonstrated tendencies towards increasing by 13 mg/m^2 and 0.02 mg/l per year respectively, meanwhile over the vegetation period wet deposition of sulphur further decreased by 5-8 mg/m^2 and its concentration in precipitation was almost stable, fluctuated around 0.32 mg/l . These changes in air concentrations and deposition of sulphur species were not significant ($p>0.05$).

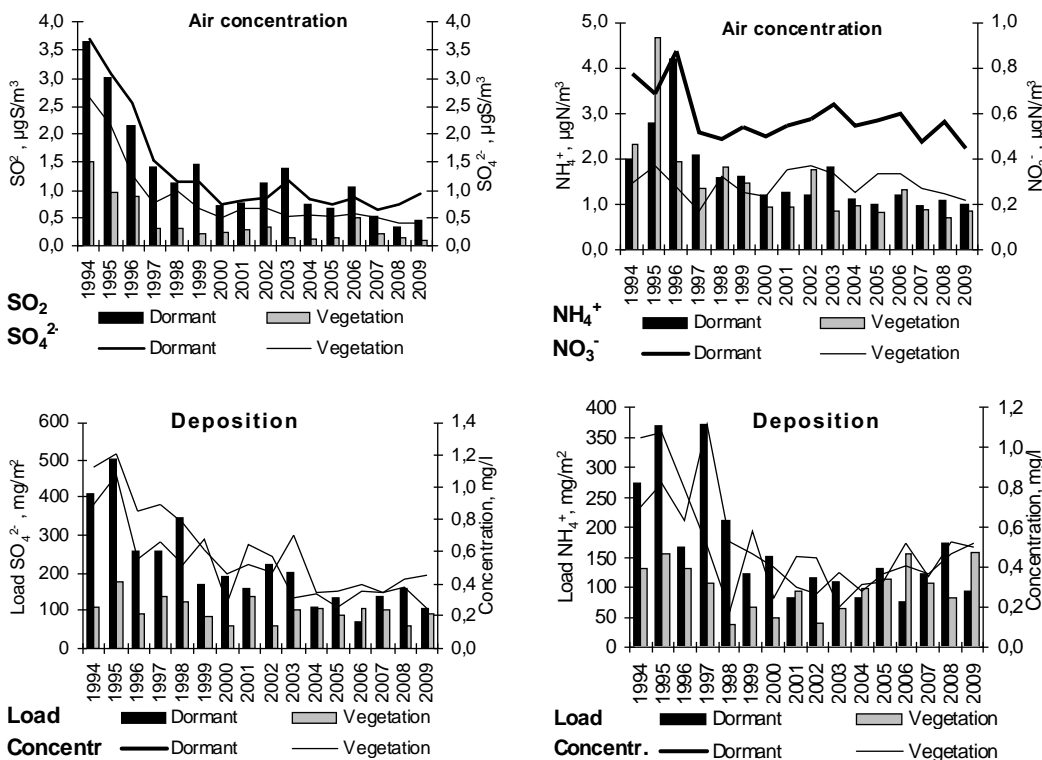


Figure 3. Changes in air concentration and deposition load of the main acidifying species over the considered period at Aukštaitija IMS

Over this period NH₄⁺ deposition and its concentration in precipitation, contrary to its air concentration, started increasing (Fig. 3). From 2001 to 2009 it made 3.9 mg/m² per year (p>0.05) over the dormant period and from 2000 to 2009 it made 9.3 mg/m² per year (p<0.05) over the vegetation period. Increase in NH₄⁺ concentration in precipitation over the dormant period was most pronounced from 2002 to 2009 and made 0.03 mgN/l per year meanwhile from 2003 to 2009 by 0.05 mgN/l per year (p<0.05) over the vegetation.

Relationships between pine defoliation and sulphur and ammonia air concentrations and their concentrations in precipitation were almost of the similar significance over all considered periods (Table 4). Correlation coefficient fluctuated around 0.55-0.60. Significance of the effect of changes in air concentration of NO₃⁻ and its concentration in precipitation explaining pine defoliation variation was found to be the lowest. In most cases these relationships were direct, i.e. higher pollution level lead to higher defoliation rate. The exception was the nitrate deposition over the vegetation period, when this relationship was indirect, i.e. higher pollution level lead to lower defoliation rate.

Integrated effect of the considered pollutants on pine defoliation

Integrated effect of ΣNH₄⁺ air concentration, their deposition (DNH₄⁺) and aqueous concentration in pre-

Table 4. Correlative analysis of Scots pine stands defoliation vs. air pollution and acid deposition

Parameters	Periods						
	IX-XII	I-IV	II-IV	IX-II	III-IV	IX-IV	V-VIII
Air concentration, (µg·m ⁻³)							
SO ₂							
r	0.560	0.541	0.479	0.591	0.488	0.576	0.516
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
SO ₄ ²⁻							
r	0.630	0.518	0.512	0.592	0.558	0.592	0.538
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
ΣNH ₄ ⁺							
r	0.518	0.643	0.624	0.563	0.624	0.598	0.558
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
NO ₃ ⁻							
r	0.334	0.348	0.222	0.475	0.039	0.411	-0.008
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.488	p=0.000	P0.885
Deposition, (mg·m ⁻²)							
SO ₄ ²⁻							
r	0.602	0.501	0.479	0.602	0.374	0.600	0.351
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
NH ₄ ⁺							
r	0.519	0.502	0.483	0.519	0.335	0.532	0.124
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.026
NO ₃ ⁻							
r	0.188	0.332	0.194	0.255	0.317	0.147	-0.367
p	p=0.001	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
Concentration in precipitation, (mg·l ⁻¹)							
SO ₄ ²⁻							
r	0.642	0.596	0.512	0.656	0.419	0.652	0.561
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
NH ₄ ⁺							
r	0.516	0.479	0.444	0.527	0.198	0.518	0.493
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000	p=0.000
NO ₃ ⁻							
r	0.200	0.360	0.279	0.317	0.144	0.342	0.182
p	p=0.000	p=0.000	p=0.000	p=0.000	p=0.010	p=0.000	p=0.001

Note: Effect of pollutants: (+) – direct, (-) – adverse. Significant relationships in bold.

cipitation (CNH₄⁺) on pine defoliation was described by the following equations:

$$F=15.41+1.162\times\Sigma NH_4^+_{(V-VIII)}+0.801\times\Sigma NH_4^+_{(IX-XII)}+3.403\times CNH_4^+_{(IX-XII)}; \\ R^2 = 0.452; p<0.000 \quad (4)$$

$$F = 14.83+0.459\times\Sigma NH_4^+_{(IX-XII)}+2.05\times\Sigma NH_4^+_{(I-IV)}+0.014\times DNH_4^+_{(I-IV)}; \\ R^2 = 0.451; p<0.000 \quad (5)$$

Integrated effect of SO₂ air concentration, S deposition (DSO₄²⁻) and its aqueous concentration in precipitation (CSO₄²⁻) on pine defoliation was described by the following equations:

$$F = 15.81+1.669\times SO_{2(V-VII)} + 5.73\times CSO_4^{2-}_{(IX-XII)}; \\ R^2 = 0.436; p<0.000 \quad (6)$$

$$F = 16.48+1.914\times SO_{2(V-VII)} + 0.63\times SO_{2(IX-XII)} + 0.0196\times DSO_4^{2-}_{(IX-XII)}; \\ R^2 = 0.413; p<0.000 \quad (7)$$

Integrated effect of aerosol SO₄²⁻ air concentration, its deposition (DSO₄²⁻) and aqueous concentration in precipitation (CSO₄²⁻) on pine defoliation was described by the following equations:

$$F = 15.98+1.142\times SO_4^{2-}_{(IX-XII)} + 4.20\times CSO_4^{2-}_{(IX-XII)}; \\ R^2 = 0.446; p<0.000 \quad (8)$$

$$F = 16.42+1.636\times SO_4^{2-}_{(IX-XII)} + 0.014\times DSO_4^{2-}_{(IX-XII)}; \\ R^2 = 0.437; p<0.000 \quad (9)$$

Effect of air concentration of ΣNO₃⁻, their deposition (DNO₃⁻) and aqueous concentration in precipitation (CNO₃⁻) demonstrated the lowest explanation rate. Effect of these species of pollutants explained up to 30% variability in pine defoliation:

$$F = 14.07+9.1\times\Sigma NO_3^-_{(IX-XII)}-0.043\times DNO_3^-_{(V-VIII)}+0.053\times CNO_3^-_{(IX-XII)}; \\ R^2 = 0.290; p<0.000 \quad (10)$$

$$F = 7.32+8.79\times\Sigma NO_3^-_{(IX-XII)} + 8.08\times CNO_3^-_{(IX-XII)}+9.77\times CNO_3^-_{(I-VI)}; \\ R^2 = 0.273; p<0.000 \quad (11)$$

Air concentration of ΣNH₄⁺, its deposition and concentration in precipitation most likely have had the most significant effect on Scots pine condition. Their integrated effect explained more than 45% variability

in pine defoliation. For the first time over the entire considered period the effect of air concentrations of SO₂ and aerosol SO₄²⁻ and their deposition and concentrations in precipitation was a little lower. Explanation rate ranged from 41.3 to 44.6% pine defoliation variability. Gradual increase in NH₄⁺ deposition and its concentration in precipitation since 2002, which well coincided with the increase in pine crown defoliation, could have been attributed to the deciding factors affecting this phenomenon.

Generalizing the obtained results multiple regression model of Scots pine defoliation in the eastern part of Lithuania vs. changes in acidifying species and ozone was elaborated:

$$F = 13.05 + 0.0621 \times O_3(VI-VIII) + 1.146 \times SO_{2(IX-XII)} + 0.673 \times NH_4^+(IX-XII) + 0.014 \times DSO_4^{2-}(IX-XII) - 0.0083 \times DNO_3^-(V-VIII);$$

$$R^2 = 0.478; p < 0.000 \quad (12)$$

According to it air concentration of ΣNH₄⁺, SO₂ and wet deposition of sulphur over the first half of dormant period (September-December) as well as wet deposition of NO₃⁻ over the vegetation explained up to 48% of temporal variability in pine defoliation under regional pollutant load in the eastern part of Lithuania.

Interaction of meteorological parameters and key parameters of pine defoliation

Meteorological parameters, the direct effect of which explained up to 30% variation in pine defoliation, in integrated effect with acidifying species was not significant. Consequently, we attempted to evaluate the integrated effect of meteorological parameters on air concentrations of the considered pollutants and especially on the amount of acid deposition. The obtained data revealed that precipitation in conjunction with temperature had direct impact on SO₂ air concentration and SO₄²⁻ deposition, indicating that increase in temperature and precipitation over the dormant period might have resulted in increase in SO₂ air concentration and SO₄²⁻ deposition:

$$SO_{2(IX-XII)} = -0.2075 + 0.0064 \times Pr(IX) + 0.0147 \times \frac{Pr(IX-XII)}{Tm(IX-XII)};$$

$$R^2 = 0.342; p < 0.042 \quad (13)$$

$$DSO_4^{2-}(IX-XII) = -71.27 + 0.568 \times Pr(IX-XII) + 12.01 \times \frac{Pr(IX)}{Tm(IX)};$$

$$R^2 = 0.693; p < 0.000 \quad (14)$$

The developed regression model revealed that precipitation amount in conjunction with air temperature over the dormant period is a key factor reflecting the changes in NH₄⁺ air concentration:

$$NH_4^+(IX-XII) = -0.281 + 0.0218 \times Pr(IX) + 0.0146 \times \frac{Pr(IX-XII)}{Tm(IX-XII)};$$

$$R^2 = 0.564; p < 0.004 \quad (15)$$

Nitrate wet deposition over the vegetation period which was the only of the key parameters with increase of which pine crown condition improved, demonstrated the direct relationship with precipitation amount and indirect – with air temperature. More abundant precipitation and lower air temperature over the vegetation should have improved pine crown condition.

$$DNO_3^-(V-VIII) = 650.4 + 2.77 \times Pr(V-VIII) - 41.16 \times Tm(V-VIII) - 40.58 \times \frac{Pr(V-VIII)}{Tm(V-VIII)};$$

$$R^2 = 0.503; p < 0.033 \quad (16)$$

The obtained results revealed that such indirect effect of meteorological parameters on pine crown condition, through close interdependencies with selected pollutant species (Langner et al. 2005, Sanderson et al. 2006, Bytnerowicz et al. 2007, Zlatev and Moseholm 2008, Jacob and Winner 2009) is the object of a very deep discussion when investigating forest responses to global environmental changes in future.

Discussion

Air pollution and climate change are two key factors comprising the global change threat to forest health and sustainability (Percy and Ferretti 2004). Their impacts on forest ecosystems have been traditionally treated separately. However, their integrated effects on forest ecosystem may significantly differ from a sum of separate effects due to an array of various synergistic or antagonistic interactions (Langner et al. 2005, Bytnerowicz et al. 2007). To avoid overlaps between two lines of traditional research, what is suggested by most of the authors (Swart et al. 2004, Bytnerowicz et al. 2007); we expressed pine defoliation changes through the changes in main environmental contaminants whereas meteorological data were chosen only as predisposing factors of those changes. Due to increasing interest in the potential impact of climate change on the distribution and reduction of air pollutants (Langner et al. 2005) we attempted to evaluate the integrated effect of meteorological parameters on air concentrations of the considered pollutants and especially on the amount of acid deposition.

It is well established that the increase in temperature in conjunction with changes in intensity of precipitation determine the atmospheric concentration and deposition of acidifying compounds (Mayerhofer et al. 2002, Buda and Walle 2002, Langner et al. 2005, Sanderson et al. 2006, Bytnerowicz et al. 2007, Jacob and

Winner 2009). The data obtained on relationships between considered meteorological parameters and SO_2 air concentration and SO_4^{2-} deposition well agreed with the state of knowledge in this field.

The statement that climate change increases NH_4^+ air concentration and deposition mainly during the dormant season (Buda and DeWalle 2002) agreed well with the obtained results too. The developed regression model revealed that precipitation amount over this period is the key parameter reflecting the changes in NH_4^+ air concentration best.

Climate change increases the amount of produced nitric acid deposited down to soils, which in turn may result in further acidification of soils what will worsen the problem of acidification in future (Sanderson et al. 2006). Therefore, precipitation due to below-cloud scavenging of HNO_3 was the key factor affecting changes in NO_3^- wet deposition (Buda and DeWalle 2002). More abundant precipitation resulted in increase in NO_3^- deposition. However, in forest ecosystem where N is often a limiting nutrient, NO_3^- deposition may increase wood production and accumulation of soil organic matter, thus demonstrating fertilization effect and increasing foliage biomass (Juknys et al. 2003) especially over the vegetation period. Therefore, negative effect of NO_3^- deposition could be expected only through the effect on soil mediated parameters and soil biota, mainly enforcing acidification processes in the soil.

Studies on the integrated effect of considered pollutant species allowed us to select possibly the most dangerous pollutants and periods when they had the greatest negative effect on Scots pine in Lithuania. Our earlier studies revealed that values of acidifying species obtained over the period from September to December explained, in most cases, more than 90% of variability in their annual values and over this period their integrated effect on changes in pine stands condition was, in most cases, more significant than over the rest periods (Augustaitis et al. 2010). This could be explained firstly by a few times higher concentrations; secondly by the combination of direct, above-ground, impacts of acidifying species 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 or on the other hand, fertilisation effect on growth on nutritionally poor sites (Augustaitis et al. 2010, Juknys et al. 2003). These findings were in full agreement with the earlier findings not only in Lithuania but also in the other parts of Europe (Lorenz and Mues 2007). Positive effect of the considered pollutant species on pine crown condition was mainly related to more abundant precipita-

tion over the vegetation which being a key factor of tree growth could result according to the created model in additionally higher nitrate deposition and lower surface ozone concentration.

Taking into account last tendencies in variation of precipitation amount detected between 2000 and 2009 – the increase over the dormant period, followed in turn by gradual increase in sulphur and ammonia deposition should result in decline of the pine crown condition. Pine vulnerability to the considered pollutants should be enhanced by the direct lack of humidity over the vegetation period as well, if the precipitation amount over this period further decreased. These pessimistic prognoses could not come true only in case if further changes in meteorological parameters and air pollutants would occur on long term basis.

Conclusion

Acidifying species and meteorological parameters are key factors that contribute to Scots pine stands condition and sustainability in Lithuania. Complete implementation of the Gothenburg Protocol helped to significantly reduce concentrations of the main acidifying species in air and their deposition, what resulted in significant pine stand condition improvement. Despite these international efforts air concentrations of N species and especially ammonium remain at the same level or even start demonstrating upwards trend, what should result in further acidification of soils and act to worsen the problem of forest ecosystem acidification in future. These changes in the considered pollutant species and their effects do not enable to predict further reliable pine condition changes without comprehensive studies on the direct effect of meteorological parameters on air pollutants and acid deposition. The Integrated Monitoring Programme, which has been annually performed since 1994 in Lithuania, provides all the necessary data to join scientists' community for solving one of the most actual problems, i.e. to evaluate the effect of climate changes on forest response to changing pollution load.

Generalizing the obtained results, we can conclude that meteorological parameters which are the result of climate change could become key factor contributing to Scots pine responses to acid deposition. Being less significant in relationships with pine crown condition than acidifying species, meteorological parameters resulted in variation of acid deposition. In case if precipitation amount increased over the dormant period and decreased over vegetation the negative effect of acidifying species could be enhanced. If changes occur according to long term tendencies reduction of the negative effect of air pollutants of acidifying species

and their deposition can be expected, despite the rise in negative effect of new threat for forest ecosystem – the increase in deposition of the ammonia compounds.

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ВЛИЯНИЕ МЕТЕОРОЛОГИЧЕСКИХ ПАРАМЕТРОВ НА ИЗМЕНЕНИЯ СОСТОЯНИЯ СОСНЯКОВ В ОТВЕТ НА ВОЗДЕЙСТВИЕ КИСЛОТНЫХ ОСАДКОВ В НАЦИОНАЛЬНОМ ПАРКЕ АУКШТАЙТИЯ**А. Аугустайтис***Резюме*

Потепление климата становится основным фактором, влияющим на развитие лесов, их адаптации к неблагоприятным условиям окружающей среды и, в первую очередь, к увеличению кислотных осадков. Несмотря на это, до сих пор не выяснено усиливает или уменьшает потепления климата влияние загрязнителей на лесные экосистемы. Поэтому, представленная работа посвящена установить потенциальный эффект изменения метеорологических параметров на состояние сосняков в ответ на воздействие кислотных осадков. Установлено, что метеорологические параметры влияют более значимо на изменения количества кислотных осадков чем на дефолиацию крон сосны. При по настоящему сценарию происходящие изменения в количестве осадков и температуры воздуха должны уменьшить повреждения сосняков кислотными осадками и повысить их жизнеспособность к неблагоприятным условиям окружающей среды. Только постепенное увеличение азотных соединений может усилить кислотные процессы в лесных экосистемах и повысить дефолиацию крон сосняков.

Ключевые слова: осадки, температура воздуха, кислотные осадки, взаимодействие, дефолиация кроны сосны.