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Air Pollutants, Their Integrated Impact on Forest Condition under Changing Climate in Lithuania

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1. Introduction

Changes in forest ecosystems, in most cases, are 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 & Freer-Smith, 1995, Cronan & Grigal, 1995, Manion & 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 effects 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 above mentioned indirect, soil-mediated effects of air pollutants on the vitality of forests are frequently more relevant than the direct above ground effects especially in local polluted areas (Roberts et al., 1989, De Vries et al., 2000b). However, on a regional scale long range transboundary air pollution and ensuing acidification of the environment could only be detected when data of the repeated surveys for a relatively long period become available (De Vries et al., 2003a).
Ambient O_3 is also among the key factors resulting in spatial and temporal changes of tree crown defoliation (Reich, 1987; Guderian, 1985; Takemoto et al., 2001; Sandermann, 1996). Integrated effects of ozone with acidifying species differ significantly from the sum of their individual effects due to the complex synergistic or antagonistic interactions (Bytnerowicz et al., 2007). Therefore, the process based on plant damages due to O_3 exposure is still not fully clarified (Zierl, 2002; Matyssek et al., 2005), and the relationship between forest tree crown condition and O_3 concentrations as well as acid deposition is still not well established. O_3 is one of the most important and pervasive phytotoxic agents whose effects are likely to increase in future (Krupa & Manning, 1988; Hutunnen et al., 2002; Percy et al., 2003; Vingarzan, 2004). In contrast to SO_2, the continuing rise in the emissions of precursor substances (NO, NO_2) (Fowler, et al., 1998; Ryerson, et al., 2001) resulted in a rise in ozone concentrations (Matyssek & Innes, 1999; Coyle, et al., 2003; Fuhrer, 2000). If since the middle of the last century ozone (O_3) air pollution has been recognized as a major phytotoxic agent in North America and South Europe (Smith, 1981; Schmieden & Wild, 1995; Hill, et al., 1970; Alonso, et al., 2002), then recently - in Central and Northern Europe (Utrainen & Holopainen, 2000; Muzika, et al., 2004; Karlsson, et al., 2002).

Therefore, due to the expected increase in O_3 concentrations (Fowler et al., 1999; Percy et al., 2003) it is necessary to determine if exposure to O_3 levels actually affects tree growth and crown condition in natural forests (Manning, 2005) under regional pollution load. 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 the acidifying compounds and their deposition meanwhile meteorological parameters only reinforce or mitigate the integrated impact of these factors (Augustaitis et al. 2003, 2005, 2007d). In this study we attempted to investigate the possible effect of natural and anthropogenic environmental factors on pine defoliation and stem growth, and quantify of O_3 contributions to the integrated impact of these factors.

However, in estimating the effect of air pollutants and their deposition on forest conditions, only the mean annual value of air pollutants and sum deposition over the year, or sum concentrations of ozone from April to September is usually used (Klap et al., 1997, 2000; De Vries et al., 2000a). Seasonal effects of the considered pollutants are attributed to the knowledge gaps in this area. Our earlier study showed that pine needles, which are present on trees all year round, seem to be more efficient aerosol collectors than leaves (Augustaitis et al., 2008a). Following this assumption, the negative effect of the considered contaminants, with the exception of O_3, should occur during the dormant periods as well. Therefore, in the present study, we set out to see whether it is possible to detect seasonal effects of air pollutants and acid deposition on the mean defoliation of pine stands employing correlative or multiple regression analysis, and evaluate their significance in explaining the variance in mean defoliation of the pine stands.

The Integrated Monitoring Programme, which has been performed for more than 15 years in Lithuania, provides all the necessary data to identify the key environmental factors, effect character and periods when the effect of the considered contaminants is most pronounced. To meet the objectives of the study the following studies were performed:

- an analysis of temporal and spatial variation of data on defoliation, meteorological parameters, air pollutant concentrations, including surface ozone and acid deposition;
- an analysis of variation of data on quality of soil, ground and stream waters in relation to air pollutants, their deposition and meteorological parameters;
- an estimate of the contributions of effect character to the integrated impact of air and soil mediated pollutants on tree defoliation
- an evaluation of the significance of the seasonal effects of the considered contaminants, explaining variance in mean defoliation of the pine crowns.
- an evaluation of the significance of the surface ozone concentration in the integrated effect with acidifying species, their deposition and meteorological parameters on Scots pine defoliation and stem increment reduction

The findings should increase our knowledge in the field of diagnostic and mechanistic understanding of processes occurring in ecosystem. It may help: (i) to estimate the integrated effect of environmental factors on forest ecosystem, (ii) detect peculiarities of the seasonal effects of acidifying compounds, surface ozone on pine defoliation, (iii) specify the periods when the effect of the considered contaminants is most pronounced, and (iv) predict state of forest ecosystem under the pressure of global changes.

2. Objects, materials and methods

In order to meet the emerged tasks the different data sets obtained in three national parks (NP), representing different Lithuanian landscape types (Aukstaitija NP – eastern part, Dzukija NP – southern part and Zemaitija NP –western part) (Fig. 1) were used. There integrated monitoring stations have been under operation since 1994.

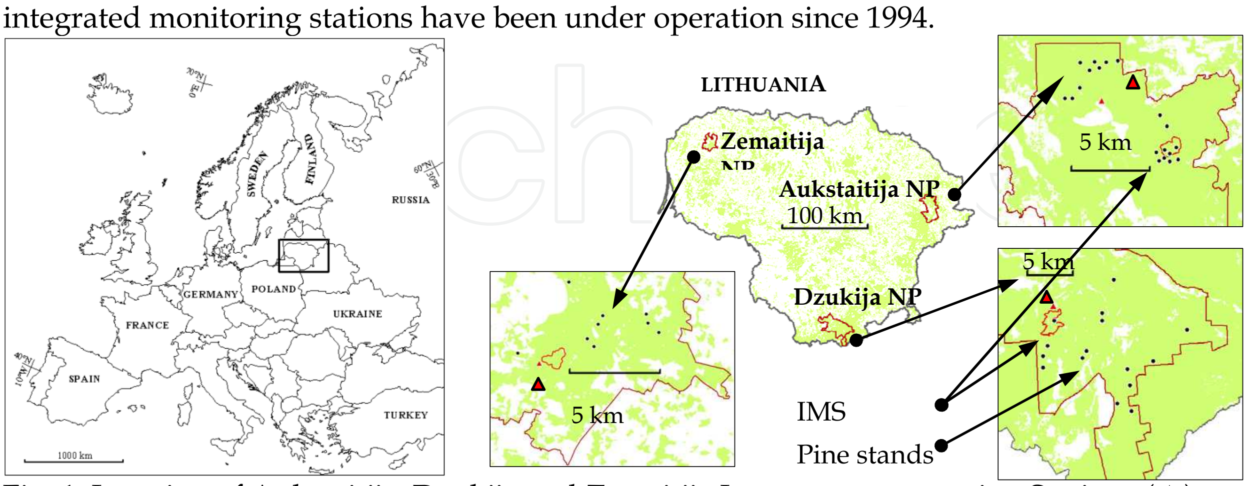

Fig. 1. Location of Aukstaitija, Dzukija and Zemaitija Integrated Monitoring Stations (▲), rivulet basins and POS in Lithuanian National Parks.

2.1 Objects for estimation of the direct effect of acidifying species and surface ozone

The study of direct effects of acidifying species, their deposition, surface ozone and meteorological parameters on forest health was based on annual observations of crown defoliation of more than 8,000 Scots pine (*Pinus sylvestris* L.) trees from 44 permanent observation stands (POS) between 1994 and 2007. Aukstaitija NP had 20 POS, Dzukija NP 16 POS and Zemaitija - NP 9 POS. These stands were located in the surroundings of Integrated monitoring stations (IMS) established there, and were selected according to stand maturity: 8 sapling stands (45–50 years), 10 middle aged stands (61–80 years), 10 premature stands (81–100 years), 10 mature stands (101– 120 years) and 7 over mature stands >121 years).

2.2 Objects for estimation of the indirect effect of acidifying species

Investigations were performed on 5 vegetation plots established for intensive monitoring at IMS catchments where dendrometric parameters and crown condition of the three tree species – Scots pine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* Karst.) and birch (*Betula pendula* 'Crispa' and *B. pubescens* Ehrh.) were assessed. Over the period 1994-2007 about 600 trees (pine trees– 30%, spruce – 50% and birch – 20%) in Aukstaitija NP (LT-01) on 3 intensive plots (IPs): A, B and C; in Dzukija NP (LT-02 A) about 100 trees on 1 IP (pine trees – 90%, spruce and birch – 10%), and in Zemaitija NP (LT-03 A) about 100 trees on 1 IP (pine trees – 20%, spruce – 70% and birch – 10%) were monitored annually.

2.3 Objects for estimation of the effect of surface ozone on pine increment

To get closer insight into the relationships between different O_3 indices and pine increment we chose 12 out of 44 pine permanent observation stands (POS). More than 200 trees, for which defoliation was assessed from 1994 to 2007, were chosen for the increment boring.

2.4 Predictor variables and methods of their estimation

The considered predictor variables were classified into 3 groups. The first group included 3 site-specific variables (forest type, soil typological groups with respect to fertility, and moisture conditions) and 5 tree and stand variables: mean tree age (A), height (H), diameter (D), basal area (BA), and stand volume (M).

The second group consisted of 10 meteorological variables (air temperature and the amount of precipitation for 8 seasons of 2 years and 2 year-long periods from September to August).

The third group of 13 variables included data on: air concentrations of sulfur dioxide (SO_2), sulfate (SO_4^{2-}), the sum of nitrates in aerosols and gaseous nitric acid ($\Sigma NO_3^- = NO_3^- + HNO_3$), the sum of gaseous ammonia and ammonium in aerosols ($\Sigma NH_4^+ = NH_4^+ + NH_3$) and surface ozone ($O_3$); concentration of SO_4^{2-}, NO_3^-, $NH_4^+$and H^+ in precipitation as well as their wet deposition (Sopauskiene & Jasineviciene, 2006; Girgzdiene et al., 2007).

2.5 Methods

Dendrometric and health parameters of trees and stands. Data on tree and stand parameters as well as crown defoliation were available from the permanent observation stands. A three-stage sampling pattern was used for the collection of the field material:

(i) sampling of the research stands;

(ii) sampling of the circular plots within each research stand;

(iii) sampling of the trees for more detailed measurements of tree crown parameters and tree ring analysis. Each pine stand included 12 circular sample plots distributed systematically in a grid system with on average 15-20 trees on each plot, i.e., about 150-250 sample trees per stand. There, tree stem diameter was permanently measured and crown defoliation assessed for all sample trees annually. In addition three closest to the centre of sampling plot trees were sampled for measurement of crown parameters (height, diameter and length) and analysis of annual radial increment. Radial growth was assessed by measuring the width of annual rings in stem cores. Based on the ring widths the stem basal area increment (BAI) was computed.

Crown defoliation was assessed at the end of August. Forest monitoring methodology was employed to assess tree defoliation: healthy trees (defoliation up to 10%); slightly damaged trees (defoliation ranging from 11 to 25%); moderately damaged trees (defoliation 26–60%); severely damaged trees (defoliation 61–99%); dead trees (defoliation 100%) (UN-ECE, 1994).

Soil-mediated parameters. Glacioaquatic accumulation forms with sand, gravel and stones are typical of the Aukstaitija IMS catchment. In this area, LT-01A and LT-01B, bores No 1 and 2 for ground water (GW) studies and lysimeters for soil water (SW) collection are arranged. Soil water samples were collected at 20 cm, 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 Dzukija IMS catchment is formed by more intensive glacioaquatic and eolian processes, than in LT-01. All 3 bores for GW collection are arranged in fine-grained sand dominated 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 of Zemaitija NP is different from that of the other stations. 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 LT-03A, bore No 1 and lysimeters for soil water collection are arranged, dominates in the Zemaitija IMS (Augustaitis et al., 2008a).

Air pollution and deposition. Data on regional air concentrations of acidifying chemical species, the concentrations and fluxes of these chemical species in wet deposition and general meteorological data were obtained from the IMS established in the national parks and used for all stands located in the park. Due to a lack of funding Dzukija IMS was closed in the year 2000. Therefore, data from this region represented the period 1994–1999.

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 (Sopauskiene & Jasineviciene, 2006).

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.

Ozone. Ozone concentrations were measured continuously using commercial UV-absorption monitors O_3 41M (Environment S.A., France) and ML9811 (Monitor Labs) with an air inlet at the height of 2.5 m above ground. The instruments were calibrated periodically every year. Hourly data on peak O_3 value, their annual average, and average from April through August were used in the analysis. AOT40 values, which define the potential risk of O_3 for vegetation (Fuhrer et al., 1997), were calculated according to the requirements of the 2002/3/EC directive. For crops, the critical level is set to 3.0 ppmh (AOT40-1); for forest trees, 10 ppmh (AOT40-2) (NABEL, 1999). Exceedance of the AOT40-2 threshold would indicate a risk of tree biomass loss of more than 10% (LRTAB, 2004).

Meteorological parameters. The effect of meteorological conditions on pine defoliation was analyzed for two yearlong periods from September to 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). Meteorological data were collected at IM stations according to the requirements of the WMO Guidelines (1989) to ensure their comparability with the data from official weather stations and other monitoring sites.

Statistics. The Fisher test was employed for estimating the significance of spatial and temporal differences in changes in pollution level and tree defoliation. The integrated impact of natural and anthropogenic factors on mean defoliation of pine trees was analyzed by a multiple stress approach, using the linear multiple regression technique of "Statistica 6.0" software. 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.

The impact of ambient O_3 on pine crown defoliation and BAI was examined in a 2-step multi-regression procedure (Neirynck & Roskams, 1999). The annual mean defoliation was regressed on site and stand predictor variables using a stepwise regression with a forward selection procedure. Then, the residual defoliation was regressed on air concentrations of acidifying compounds, their concentration in precipitation and deposition, and meteorology using the same stepwise regression procedure. The annual BAI was regressed on stand predictor variables and crown defoliation using a stepwise regression with a forward selection procedure. Then, the BAI residuals were regressed on air concentrations of acidifying compounds, their concentration in precipitation and deposition, and meteorology using the same stepwise regression procedure. Finally, the predictor variable ''peak ozone concentration'' was included in the created models and its contribution to the integrated impact of different combinations of natural and anthropogenic factors on pine crown defoliation and BAI was quantified.

3. Spatial and temporal variation of predict and response variables

3.1 Air pollution and acid deposition

IMS data showed a significant decrease in pollutants until the year 2000 (Sopauskiene *et al*. 2001, Augustaitis *et al*. 2005, Sopauskiene & Jasineviciene, 2006, Augustaitis *et al*. 2007b). The air concentration of SO_2 at Aukstaitija IMS decreased by 82% (from 2.73 to 0.49 μgS/m^3), at Zemaitija IMS by 79% (from 2.22 to 0.47 μgS/m^3), and at Dzukija IMS by 57% (from 3.0 to 1.3 μgS/m^3) (Fig. 2). Thereafter, the concentration was stable at the level of 0.5 – 1.0 μgS/m^3. Air concentration of aerosolic SO_4^{2-} changed in a similar to SO_2 air concentration pattern.

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). Since 2001 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.

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 Aukstaitija NP decreased by 58% (from 600 to 250 mgS/m^2), at Zemaitija by 60% (from 750 to 300 mgS/m^2), and at Dzukija by 52% (from 660 to 320 mgS/m^2). Since 2001 at LT-01, sulphur deposition further decreased to 200 mgS/m^2, while at LT-03, it drastically increased again in 2002-2003 and 2007, reaching around 600 mgS/m^2 (Sopauskiene & Jasineviciene, 2006).

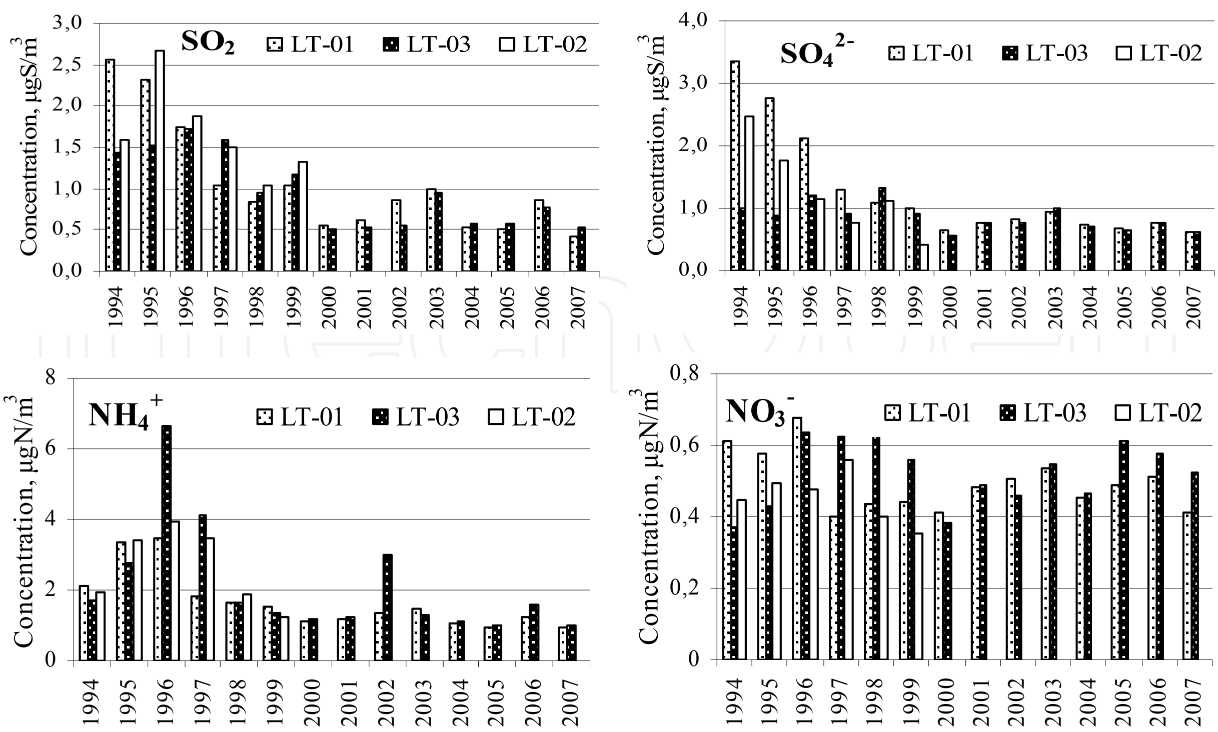

Fig. 2. Changes in the mean air concentration of the considered contaminators.

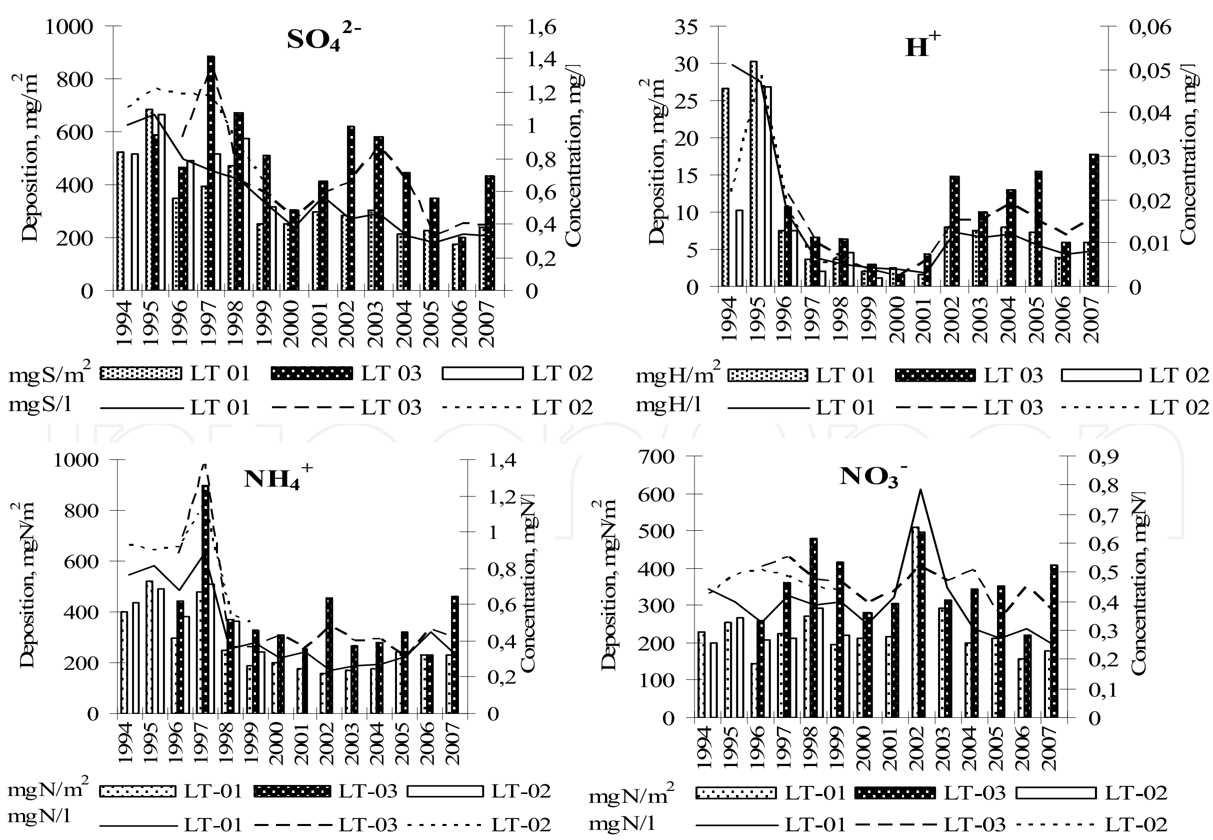

Fig. 3. Changes in acid deposition and their concentrations in precipitation (LT-01 – Aukstaitija IMS; LT-02 – Dzukija IMS; LT-03 – Zemaitija IMS)

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 occurred until 2001. Afterwards, a gradual increase in deposition of the contaminant was observed at all stations. Contrary to this, 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. Despite this, the total N deposition since 2001 started to increase again mainly due to the increase in ammonium deposition. As a result of these changes in acidifying compounds until 2001 a more than tenfold decrease in H^+ concentration in precipitation and its deposition was observed at the stations, however, since afterwards an increase especially at LT-03 was recorded. Acidity of precipitation started to increase again mainly due to repeated increase in NH_4^+ deposition especially in western part of Lithuania.

Therefore, even after a 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).

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

3.2 Surface ozone

Ozone concentration data at IMS showed no clear trend in temporal changes in the annual mean, and mean values from April through August as well as in the AOT40 (Girgzdiene et al., 2007). However, decline in the peak concentrations from 215 to 125 μg/m^3 was observed until 2001 (Fig. 4). After 2001 gradual increase in both means (annual and April-August period), and peak concentrations was observed. The peak hourly O_3 concentrations ranged from 125 μg/m^3 to 165 μg/m^3 during the summer period and were typical of other parts of Central Europe (Pell et al., 1999; Solberg et al., 2005; Bytnerowicz et al., 2004). In Lithuania high (more than 120 μg/m^3) ozone concentrations were mostly observed when air masses were transported from Western Europe. Instead of rather similar ozone concentrations at all stations (Fig. 4), AOT40 values varried more significantly due to different meteorological conditions. Ozone concentrations higher than 80 μg/m^3 were mostly observed on sunny days the number of which differed significantly in the western (LT-03) and eastern parts (LT-01) of Lithuania. The computed AOT40 values for the protection of forest at LT-01 and LT-02 ranged from 8000 to 21000 μg/m^3 h while at LT-03 only from 5000 to 12000 μg/m^3 h.

The concentration of O_3 among the concentrations of the other monitored air pollutants (SO_2, ΣNO_3^- and ΣNH_4^+) reached the closest to critical phytotoxic level. The AOT40 value for the protection of vegetation (6000 μg/m^3 per h) was exceeded at all stations for almost all considered years (Girgzdiene et al., 2007). The critical level 20000 μg/m^3 per h for the protection of forest was observed only at LT-01 in the year 1999. The highest peak ozone value 213 μg/m^3 was observed only in the year 1995, while higher than 160 μg/m^3 value – at the beginning of observation and recently, in the years 2002 and 2005. Peak ozone concentrations most often were observed in spring, i.e., in April and May.

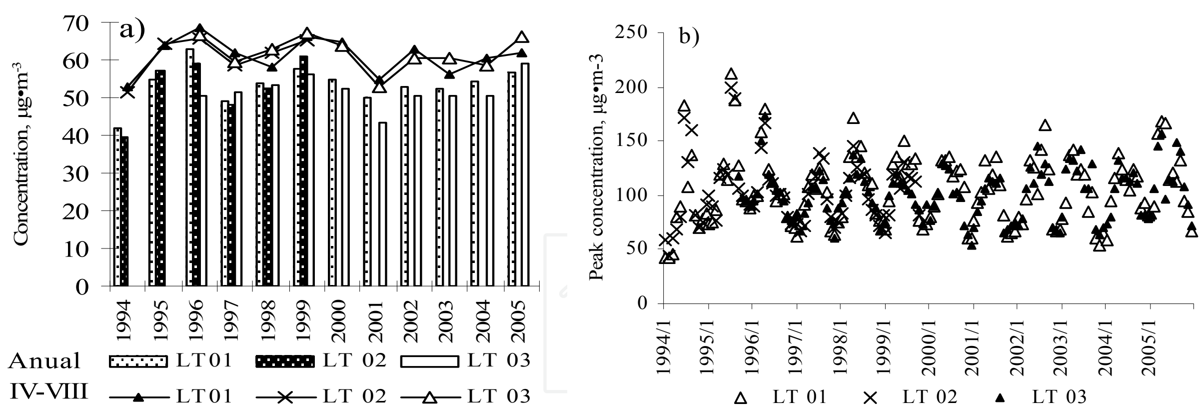

Fig. 4. Variation in ozone annual and April-August period means (a) and peak concentrations (b)

3.3 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. 5): 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 it increased by 0.021 mgN/l and 0.075 mgN/ l per year, respectively. The detected changes in NO_3^- 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, 2007a; 2008a). Consistently, the acidity of soil water at LT-01 decreased while at LT-03 it increased.

3.4 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 shallow bores showed a decreasing trend, and in the water of the deeper bores an increasing trend. NH_4^+ concentration showed a trend towards decreasing in all bores of all IM stations.

SO_4^{2-} concentration changes had no regular patterns at 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.

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 in 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.

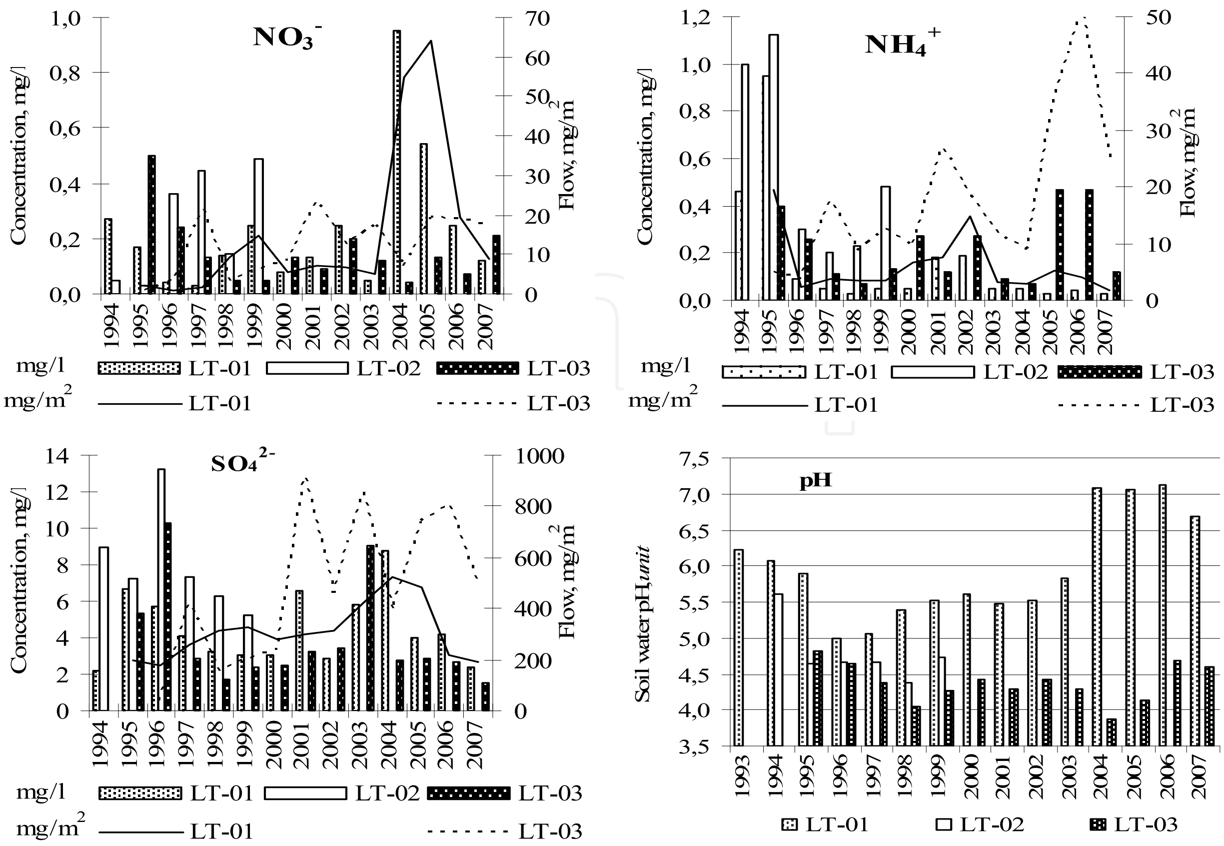

Fig. 5. Concentrations of the acidifying species in soil water and their flows at 20 cm depth. (LT-01 – Aukstaitija IMS; LT-02 – Dzukija IMS; LT-03 – Zemaitija IMS) were also observed at LT03, mainly due to higher air concentrations of the considered pollutants and their deposition (Augustaitis et al., 2005, 2008a).

3.5 Runoff water quality

Stream water quality and runoff of the main chemical compounds from ecosystems reliably reflect a common tendency of the chemical processes occurring in forest ecosystems. Therefore, this parameter was used in the analysis to detect the indirect effect of deposition on tree conditions. Concentrations of NO_3^- and SO_4^{2-} in the surface water had no statistically significant trends over the considered period (Fig. 6), however, since 1998 at LT-01 a significant decrease in SO_4^{2-} concentration was observed. Concentration of NH_4^+ had a tendency to decrease in all stations over the entire observation period. The year 2007 was an exception, when concentration of this contaminator at LT-03 increased drastically. These detected changes resulted in a gradual decrease of surface water acidity at all sites.
Despite rather similar character of the changes in NO_3^- and NH_4^+ concentrations in runoff water, there was an 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 a tendency to increase (Fig. 7). Similarly, the output of sulphur at LT-03 increased significantly while at LT-01 and LT-02, it decreased only until 2000, and afterwards some increase was observed.

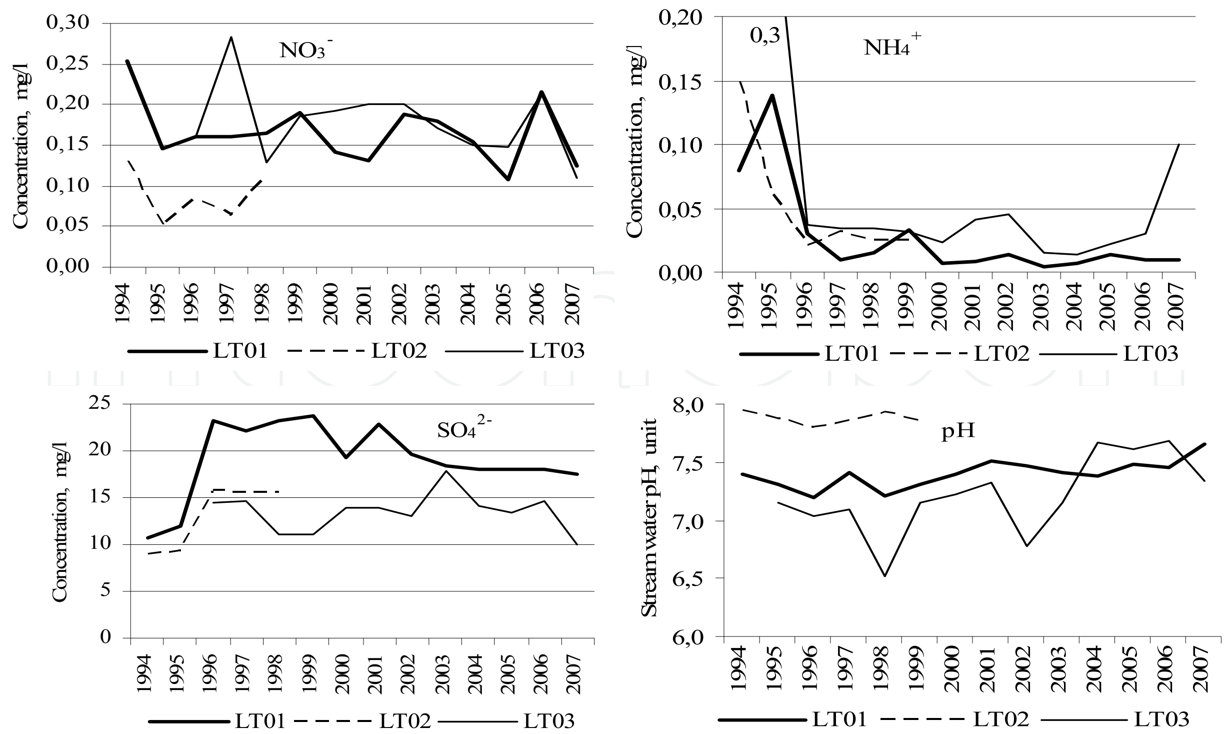

Fig. 6. Concentrations of the acidifying compounds in surface water of IM sites

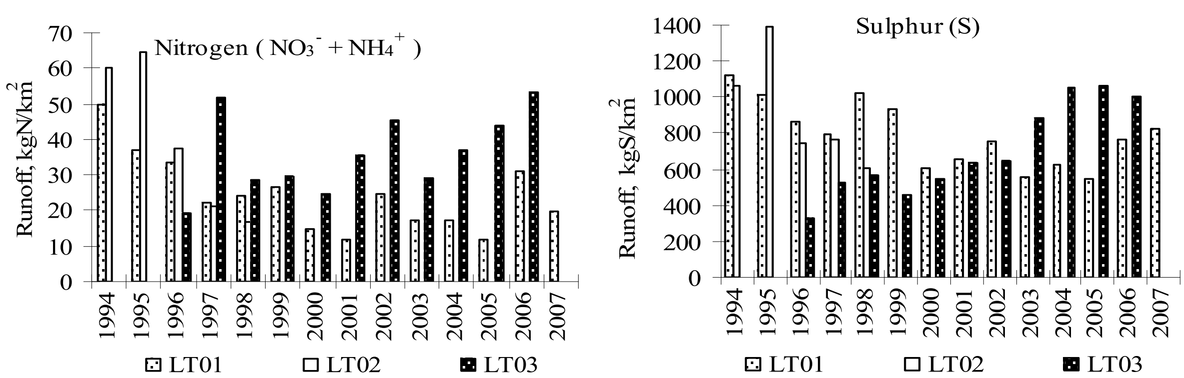

Fig. 7. Runoff of sulphur and nitrogen from the considered forest ecosystems

3.6 Temporal and spatial changes in meteorology

Annual amounts of precipitation did not demonstrate significant temporal tendency in NP during the 1994-2007 period meanwhile difference among them was significant ($p<0.05$). Mean annual precipitation was 930 mm at Zemaitija NP, 710 mm at Aukstaitija NP, and 650 mm at Dzukija NP (Fig. 8). The precipitation amount from October through February accounted for these differences (Augustaitis et al., 2005; 2007b; 2007d).

Mean annual temperature tended to increase between 1994 and 2007 ($p > 0.05$). At Aukstaitija there was an average increase of 0.013ºC per year, at Dzukija 0.061ºC, and at Zemaitija 0.069ºC per year. The increase was most pronounced in autumn (September-November). Mean annual temperature in Zemaitija (+6.9ºC) was significantly higher than in Aukstaitija (+6.4ºC), and Dzukija (+6.5ºC) ($p<0.05$).

3.7 Tree crown defoliation

Scots pine trees in Aukstaitija NP showed the best condition as illustrated in Fig. 8. On the poorest sites (*Pinetum cladoniosum* forest type) in Dzukija NP, outbreaks of the forest pests (*Diprion pini* L. and *Ocneria monacha* L.) started in 1992 after a hot and dry vegetation period in 1991 and caused very serious crown damage. In 1996 biological insecticide Foray-48B was applied to suppress the outbreak, and recovery of the damaged Scots pine trees started. The highest level of mean defoliation of pine trees in Aukstaitija and Dzukija NPs was observed in 1995, whereas in Zemaitija NP it was observed in 1997. Afterwards, crown condition showed obvious improvement that lasted until 2005. Since this year pine defoliation started to gradually increase again. The detected temporal changes in mean defoliation of pine trees were quite common throughout most of Europe (UN-ECE, 2005; Lorenz & Mues, 2007).

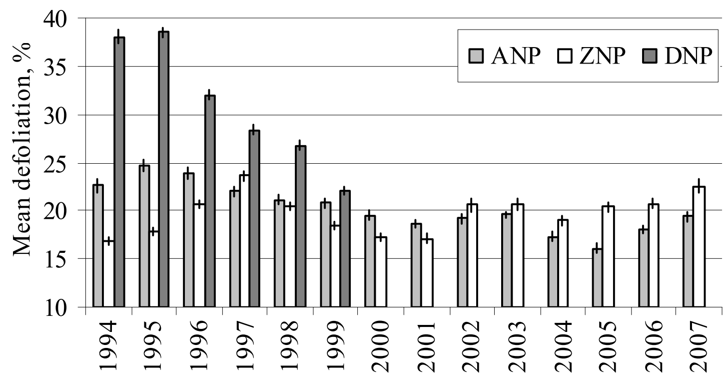

Fig. 8. Spatial and temporal changes in pine crown defoliation

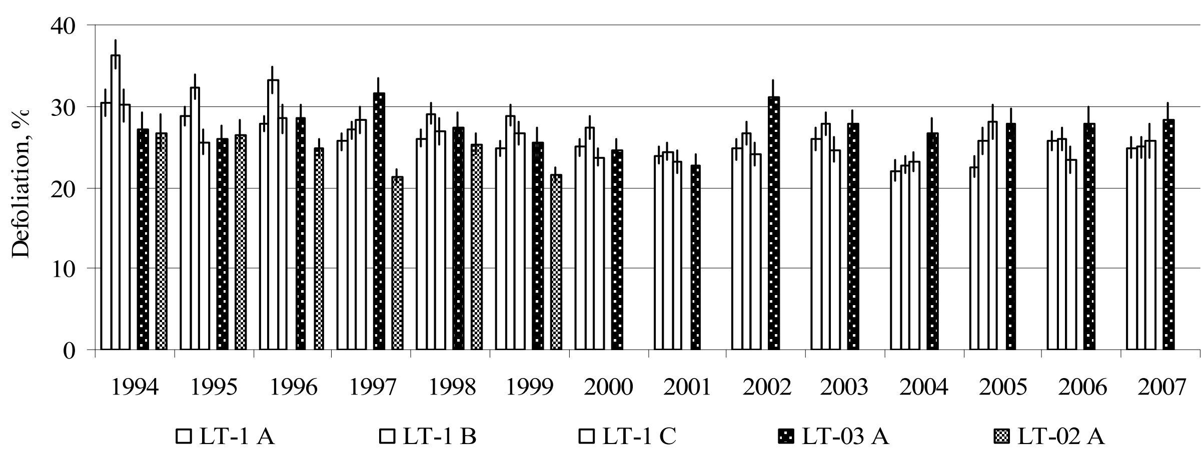

Fig. 9. Mean defoliation and standard error of estimation of the considered stands (LT-01 – Aukstaitija IMS; LT-02 – Dzukija IMS; LT-03 – Zemaitija IMS)

F test statistics indicated significant differences in mean defoliation of the prevailing tree species ($p<0.05$) among and within the intensive plots at Aukstaitija IMS (LT-01). It follows from figure 9 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 at an average by 0.61% per year. At LT-01B over the same period, the decrease made 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-03A had no regular pattern. Peaks in defoliation were observed in 1997 and 2002, when they exceeded 30% (Fig. 9). During the rest of the period, defoliation fluctuated between 22% and 27%. The changes in mean defoliation at LT-02A demonstrated a significant trend towards decreasing ($p<0.05$). Between 1994 and 2001 it decreased from 26.7% to 21.6% (Augustaitis et al., 2005, 2007a; 2007b).

3.8 Stem radial increment of Scots pine trees

Significant temporal changes in annual stem increment were established at all sites ($p < 0.05$) (Fig. 10) and reflected changes in crown defoliation (Fig. 8). Correlation coefficients between crown defoliation and stem increment ranged between r=-0.25 in Zemaitija NP and r = -0.91 in Dukija NP. Over the considered period increment of the pine stems in Aukstaitija NP increased by an average of 0.033 mm per year in middle aged and by 0.018 mm per year in matured stands. In contrast, in Zemaitija NP radial increments of the pine stems decreased by an average of 0.024 mm per year. Over the period from 1994 to 1999 pine stem increment increase in Dzukija NP was the most significant, by 0.178 mm per year (Fig. 10).

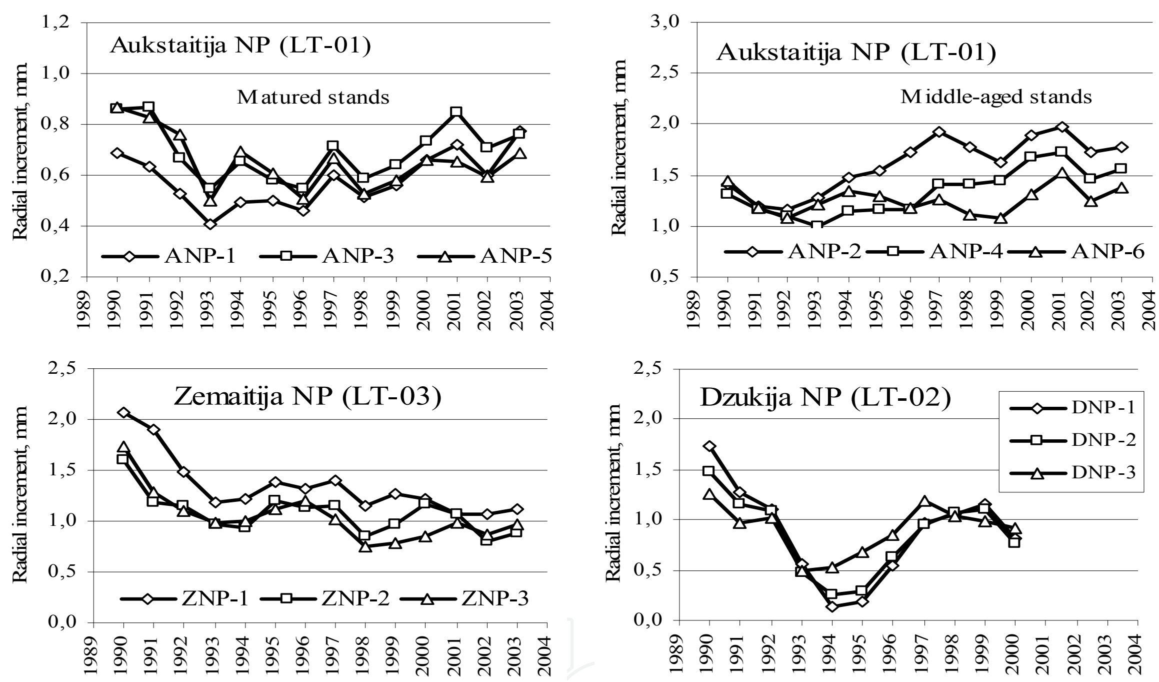

Fig. 10. Radial increment of pine trees at different sites.

4. Effect of predict variables on tree health and increment

4.1 Effect of meteorological parameters on Scots pine defoliation

Derived relationships between meteorological variables and tree defoliation showed that the correlation between pine defoliation and the amount of precipitation was generally the strongest one, followed by the correlation with air temperature. The direct correlation between pine defoliation and precipitation in September was found to be the strongest, followed by a weaker direct correlation with precipitation in the second half of the dormant period (February-April). Temporal changes in precipitation during these months (precipitation decrease) indicated improvement of pine stand conditions (lower defoliation

level). A negative correlation was established between defoliation and precipitation during the vegetation period (May-August), when the increase in precipitation should have resulted in better tree crown conditions as well (Augustaitis et al., 2010a).
The seasonal effect of monthly air temperature on pine defoliation was less significant than that of precipitation. In most cases, the effect of temperature on pine crown defoliation was negative and only higher temperatures in November-December should have resulted in better pine crown condition. The established significance of these relationships was in full agreement with the findings of the ICP Forest monitoring programme (De Vries et al., 2000a). An exceptional case, however, was the effect of the precipitation during early spring (March and May), a higher amount of which resulted in an increase in defoliation. Therefore, the hydrothermal index had no significant impact on tree crown defoliation.
An increase in both precipitation during the vegetation period and mean monthly temperature from September to December, as well as a decrease in temperature and precipitation over the rest of the dormant period, represented the conditions of climate change during the 14-year period in Lithuania, where the monitored pine stands were located. The positive effect of warmer and dryer dormant periods on tree condition is well known (Kozlowski et al., 1991), and our results confirmed that. An increase in temperature over the dormant period, particularly for the 2-month period of November and December, should have improved the condition of pine stands in Lithuania. The positive effect of more abundant precipitation over the vegetation period is well known (Makinen et al., 2003; Kahle and Spiecker, 1996). Gradual increase in temperature over vegetation, what could result in increase in surface ozone concentration should result in tree condition deterioration. Despite this, positive effects of changing climate conditions could be expected. By contrast, this process is opposite to the official scenario of climate change presented by the SRES A1 B Project (IPCC 2007), in which an increase in drought effect during the vegetation period is expected. Further investigation should allow us to check our assumptions.

4.2 Effect of air pollutants and acid deposition on tree crown condition

The air concentrations of SO_2 and NH_4^+ as well as their wet deposition showed the strongest statistically significant relationships with temporal and spatial changes in mean defoliation of Scots pines ($p<0.05$) (Fig. 11). There was the weakest relationship between defoliation and the air concentration of ΣNO_3^- ($r=0.23$). The obtained results are in agreement with the ICP Forests Monitoring data (De Vries et al., 2000a, 2003; Klap et al., 2000).

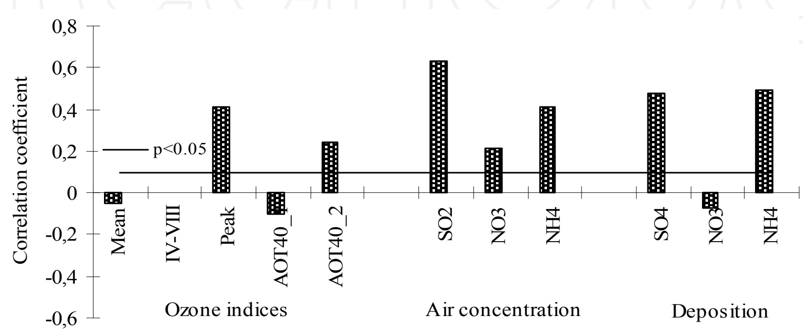

Fig. 11. Correlation coefficients of mean defoliation of pine stands and considered pollution variables in Lithuanian NPs (AOT40-1 for vegetation; AOT40-2 for forest)

Correlation coefficient between peak ozone concentration and temporal and spatial changes in mean pine defoliation was weaker. In Aukstaitija NP correlation coefficient between mean stand-wise defoliation and peak O_3 concentration in 13 of 22 stands was statistically significant ($p<0.05$, $r>0.56$), in Dzukija NP in 5 of 16 stands ($r>0.80$), and in Zemaitija NP – in 3 of 9 stands (Table 3). Data revealed that the changes in mean stand-wise defoliation of pines were most likely related to changes in acidifying compounds and peak concentration of ambient ozone. Significance of AOT40-2 for forest was twofold lower, meanwhile AOT40-1 for vegetation as well as other considered ozone indices were not significant ($p>0.05$).

4.3 Integrated effect of the considered variables on tree crown condition

A multiple regression analysis showed that 79% of the spatial and temporal variance in defoliation of the pine trees was explained by the variation in regional air pollution and meteorological, site and stand variables (Table 1). These findings suggest that the worst conditions of Scots pines which were found at Dzukija NP were not due to forest pests, alone. The poor site conditions, the higher level of pollutants, and the low precipitation amount explained together 75% of the defoliation variance of the pine trees in this park. Outbreaks of forest pests increased crown defoliation only by 7–19%, which accounted for 25% of the variance (Augustaitis et al., 2007d).

Model	Predictor variables (parameters) of:							Regression summary of model			
	Stand-site		Meteorology		Pollution						
	Stand	Site	Precipitation	Temperature	In air	In precipitation	Load	F	R^2	p	Std. err.
			mm	°C	µg/m³	mg/l	mg/m²				
F (4.400)	+	+						129.4	0.564	0.00	5.2
F (4.400)	+							61.1	0.337	0.00	6.2
F (1.403)		+						174.3	0.301	0.00	6.5
F (6.398)			+	+				43.0	0.393	0.00	6.2
F (5.399)			+					48.0	0.376	0.00	6.2
F (3.399)				+				42.2	0.178	0.00	7.2
F (6.398)					+	+	+	89.8	0.575	0.00	5.2
F (2.402)					+			146.9	0.422	0.00	6.0
F (2.402)						+		154.4	0.434	0.00	5.9
F (2.402)							+	115.0	0.364	0.00	6.3
F (5.399)					+	+		99.8	0.555	0.00	5.3
F (5.399)						+	+	84.3	0.514	0.00	5.5
F (5.399)					+		+	85.1	0.516	0.00	5.5
F (14.390)			+	+	+	+	+	50.8	0.646	0.00	4.8
F (10.394)		+	+	+				98.9	0.715	0.00	4.2
F (8.396)		+			+	+	+	158.5	0.762	0.00	3.8
F (16.388)		+	+	+	+	+	+	89.8	0.787	0.00	3.7

Table 1. Multiple regression analysis of the impact significance of the group of predictor variables on changes in crown defoliation of Scots pine trees

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.

The group of pollution variables (air concentrations of the acidifying species, and their wet deposition) was regarded as the most relevant one. The integrated impact of these variables accounted for 58% of the variance in pine tree defoliation. A separate regression with air temperature and amount of precipitation (meteorological factors) accounted for 39%.
The integrated impact of the variables from both of these groups accounted for 65% of the variance, and inclusion of stand and site parameters increased the degree of explanation by another 14% (totally 79%). Based on the obtained results we could state, that regional air pollution and acid deposition had the most significant effect on temporal and spatial changes in Scots pine crown defoliation in Lithuania.

4.4 Significance of the effect character on tree defoliation

Changes detected in mean defoliation of the prevailing tree species on vegetation plots of IMS 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 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. 12). This fully agrees with our data obtained in National parks and 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. (Augustaitis et al., 2010b).
An analysis of possible indirect effects of the considered contaminators revealed that pine crown defoliation demonstrated the highest correlation with NH_4^+ concentrations in soil and ground water, following similar with 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.
The 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. The effect of changes in concentrations of the considered compounds in soil and ground water on pine crown condition was remarkably lower (Table 2). However, this indirect effect increased the explanation of pine defoliation variability significantly - by 15%, up to 89%.

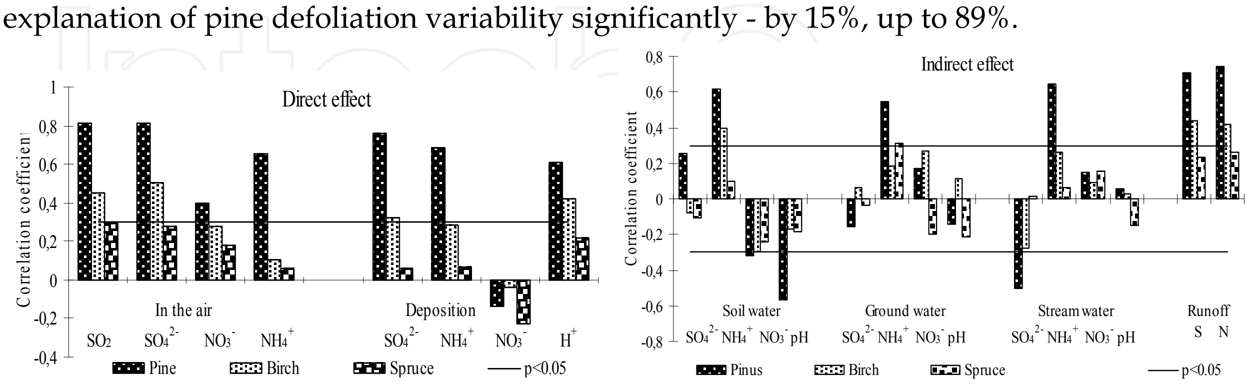

Fig. 12. Direct and indirect effect of concentrations of considered contaminators in the air, and their deposition on tree defoliation.

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																					
$\sum NH_4$	+							+						+							
$\sum NO_3$	–						–														–
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

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

Note:. F(a.b) - models identified by F - test symbol. Effect on the defoliation: + - relevance; – inverse when $p<0.05$.

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

The detected very weak 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 was reached in 2002 – 4.5%.

Generalizing these results, we could state that in most cases, nitrates in soil and ground water have a positive effect on crown condition as does a higher value of pH of these waters, while ammonia has a negative effect. The indirect effect of sulphur compounds was less significant. However, its concentrations in surface water and runoff, which reflect chemical processes occurring in the ecosystem, reflected changes in tree crown defoliation rather well. Nonetheless, the direct effect of air concentration was more significant for pine crown defoliation than their indirect soil-mediated effect. Needles, which are present on trees all year round, seem to be more efficient aerosol collectors than leaves what was confirmed by other authors (Blood *et al*. 1989, Rothe *et al.* 2002). To test this hypothesis, analysis of the seasonal effect of considered pollutants on pine defoliation was performed.

4.5 Seasonal variation of the effect of air pollution and acid deposition on pine defoliation

The seasonal variation in air pollutant concentrations and acid deposition resulted in different significances of the relationships of the considered pollutants and pine stand defoliation. Despite a very high seasonal variation in monthly air concentrations of SO_2 ($p<0.05$) and SO_4^{2-}($p<0.05$), the relationships between the considered contaminants and defoliation of pine stands did not differ significantly. Over a two year period, correlation coefficients fluctuated between 0.5 and 0.7. The findings revealed that the integrated effect of monthly values of air concentrations of sulphur compounds did not explain pine defoliation variability more significantly than their annual values. Although all possible monthly effects of this air pollutant on pine defoliation over the two year period were accounted for, the explained portion of variation in pine stand mean defoliation increased by about only 10% ($p>0.05$) (Augustaitis et al., 2010a).

The most stable monthly air concentrations of ΣNH_4^+ demonstrated rather stable effects on pine crown conditions during the entire year. In contrast, insignificant seasonal variation in NH_4^+ deposition demonstrated the pronounced seasonal character of NH_4^+ deposition effects on pine defoliation. Correlation coefficients fluctuated between 0.1 and 0.4 during vegetation and between 0.5 and 0.8 during dormant periods.

Analysis of the relationships derived from monthly values of NO_3^- (air concentrations or deposition) and pine defoliation revealed controversial results. High variations in monthly values resulted in the highest variation of the relationships. Correlation coefficients ranged from +0.4 indicating a negative effect during the dormant period to -0.2 indicating a positive effect during the vegetation. The integrated effects of the monthly values of ΣNO_3^- air concentrations in comparison with its mean annual values on pine stand mean defoliation increased the explained portion of pine defoliation variance by more than two times.

The variation in seasonal O_3 concentrations and its effect on pine defoliation demonstrated a very similar pattern, from positive in the January to May period, to negative in the June to November period; however, this effect was not significant in most cases ($p>0.05$).
Obtained results confirmed our assumption that the negative effects of acidifying compounds could occur not only during vegetation but also during the dormant period, while the negative O_3 effect – only in summer and autumn. This is confirmed by the current state of knowledge in this field. Finally, we could conclude that the accounting for integrated effects of the considered pollutants was generally more significant than the effect of their annual value including ozone; this should be taken into account when investigating key factors resulting in changes in tree crown defoliation (Augustaitis et al., 2010a).

5. Effect of surface ozone on pine tree condition

Ambient ozone (O_3) is considered to be one of the most important and pervasive phytotoxic agents whose effects are likely to increase in future (Krupa and Manning, 1988; Hutunnen et al., 2002; Percy et al., 2003; Vingarzan, 2004). However, too little is known about the effect of O_3 on tree growth on a regional scale, where its effects may be subtle and difficult to detect (Paoletti, 2006; Percy and Ferretti, 2004) and studies often fail despite using sophisticated statistically-based approaches (Muzika et al., 2004). Therefore, in this study we attempted to quantify O_3 contributions to the integrated impact of environmental factors on pine defoliation and stem growth by means of multiple regression analysis.

5.1 Contribution of the surface ozone effect on pine crown condition increment

Correlation between pine defoliation and stand density was found to be strongest ($r = 0.38$), followed by a weaker negative correlation with stand volume ($r = -0.23$) and age ($r = -0.19$, $p<0.05$). Significant differences in defoliation were established among stands from different sites. The highest mean defoliation (36.4%± 1.0) was recorded on *Pinetum cladoniosum* forest type, the lowest on *Pinetum vacciniosum* FT (19.2%± 0.2) and *Pinetum oxalidosum* FT (19.6%± 1.4). These data revealed that stand and site parameters had significant effect on spatial distribution of pine defoliation (Augustaitis et al., 2007b). Integrated impact of these parameters accounted for more than 60% variation in defoliation:

$$F = 11.61+1.378\times A-0.169\times\sum G+0.012\times N+1.884\times FType;\quad R^2= 0.610,\ p<0.05 \tag{1}$$

F – crown defoliation, %; A – stand age, years; $\sum G$ – sum of tree basal area, m^2/ha; N – tree number, units/ha; FType – forest type (categorical value).

In order to meet the objectives of the present study, relationships between pine defoliation and pollution and meteorology were examined after the influences of site and stand parameters had been accounted. This procedure had not considerable effect on the level of significance of the relationships between defoliation residuals and meteorology, meanwhile resulted in a decrease in significance of the relationship between defoliation residuals and acidifying compounds from 1.5 times (air concentration) to 2 times (deposition), and increase in the significance of the relationship between defoliation residuals and peak O_3 concentrations from 0.315 to 0.439 (Fig. 13).

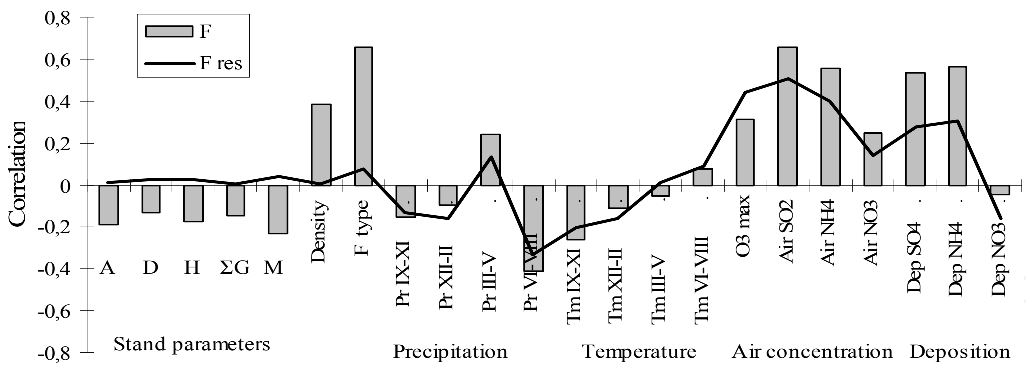

Fig. 13. Relationships between crown defoliation (F), its residuals (F_res) and considered parameters of stand, pollution and meteorology

Models, F(a.b)	1	2	3	4	5	6	7	8	9
Variables	(1.419)	(2.418)	(2.418)	(3.417)	(4.416)	(2.418)	(2.418)	(5.415)	(6.414)
In the air: SO_2	+								+
NH_4^+							+		
NO_3^-						+			
In precipitation: SO_4^{2-}		+							+
NH_4^+		+							
NO_3^-									
Deposition: SO_4^{2-}			+						
NH_4^+							+		
NO_3^-			+			+			
Precipitation:									
last season: IX-XI									
XII-II								+	+
III-V									
VI-VIII				+				+	+
current season: IX-XI				+					
XII-II									
III-V									
VI-VIII				+				+	
Temperature:									
last season: IX-XI					+				
XII-II					+			+	+
III-V									
VI-VIII									
current season: IX-XI					+			+	
XII-II					+				
III-V									
VI-VIII									
r^2, %	25.4	18.1	17.3	20.0	11.5	7.1	18.0	24.0	29.0
r^2 with O_3 effect, %	27.7	26.1	23.2	25.7	22.9	19.5	25.8	26.7	29.9
O_3 effect $(r^{2*} - r^2)$, %	2.4	8.0	5.9	5.7	11.4	12.4	7.8	2.7	1.0
O_3 significance: p<	0.00	0.000	0.000	0.000	0.000	0.010	0.010	0.000	0.034

Table 3. Contribution of the ozone to integrated impact of different environmental factors on pine defoliation. Note: individual impact of O_3 on defoliation residual: r^2=19.3% and p<0.0001.

Despite the statements that below the phytotoxic level no direct threat to vegetation from SO_2 (Bytnerowicz et al., 1998; Hjellbrekke, 1999) or synergetic interaction between SO_2 and O_3 could be expected (Krupa and Arndt, 1990), the presented data confirm our earlier findings that acidifying air compounds and their deposition are the key factors resulting in pine defoliation changes (Guderian, 1985; Takemoto et al., 2001). They could explain from 23–28% variance of residual defoliation of pine trees (Table 3). The effect of peak O_3 concentrations was less significant (19.3%), however, the presented data verified the statement that O_3 could reinforce their effects (Guderian, 1985; Takemoto et al., 2001). Ozone increased the explanation rate of defoliation residual variability by air concentration of acidifying species and their wet deposition by almost 3–8% (Augustaitis et al., 2007d).
Drought, especially during the vegetation period, is often mentioned as one of the key factors resulting in defoliation changes. However, there are contrary statements indicating that the effect of O_3 and drought might counterbalance each other (Zierl, 2002). Closed stomata protect foliage from the highest concentrations of O_3. This contrary interaction could be explained by the fact that despite increase in O_3 concentrations from north towards south (Matyssek and Innes, 1999; Karlsson, 2002), O_3 exposure in northern latitudes often leads to plants becoming more susceptible to injury than in southern areas (Matyssek and Innes, 1999). Long, bright days and high humidity in air and soil are typical for the situation in large parts of the Nordic countries (Karlsson, 2002). The peak concentrations are typical in spring (Utrainen and Holopainen, 2000). Not very high air temperature, low vapor pressure deficits, and sufficient soil water supply is characteristic for this period. Therefore, in these areas, O_3 flux inside the leaves could be higher if compared with southern areas.

5.2 Contribution of the surface ozone effect on pine stem increment

Contribution of the ozone effect to the changes in residual increment was quantified after the influences of tree dendrometric parameters (age, diameter) and crown defoliation had been accounted for. Correlation between pine stem basal area increment (BAI) and crown defoliation was strongest (r=-0.512), followed closely by positive correlation with tree diameter (r=0.382), and a weaker negative correlation with stand age (r=-0.081) (Fig. 14). Integrated impact of these parameters was analyzed by the means of multiregression model:

$$Zq = 7.330-0.189\times F-0.088\times A+0.50\times D;\ R^2= 0.795,\ p<0.05 \quad (2)$$

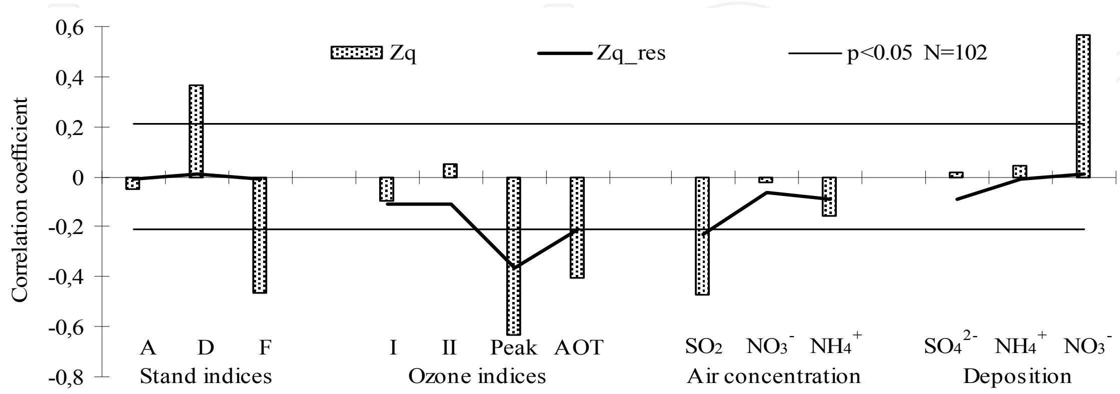

Fig. 14. Relationships between basal area increment (Zq), its residuals (Zq_res) and considered parameters of stand and pollution.
(I – mean value of ozone for April-August; II – annual mean value of ozone)

	Models, F(a.b)	1	2	3	4	5	6	7	8	9
Variables		(3.98)	(3.98)	(3.98)	(4.97)	(4.97)	(2.99)	(4.97)	(4.97)	(8.93)
In the air:	SO_2	+			+					–
	NH_4	–				–				
	NO_3	–			+	–				+
In precipitation:	SO_4		–							
	NH_4		–			–				
	NO_3		–			–				
Deposition:	SO_4			–						+
	NH_4			–	+					
	NO_3			–	+					+
Precipitation:										
last year:	IX-XI									
	XII-II									
current year:	III-V						–		–	
	VI-VIII						–		–	–
Temperature:										
last year:	IX-XI							+	+	+
	XII-II							–		–
current year:	III-V							+	+	
	VI-VIII							–		–
r^2, %		6.3	0.4	2.5	18.5	0.7	0.2	7.7	10.2	21.7
r^{2*} with O_3 effect,%		15.3	16.4	19.4	27.2	23.9	14.4	17.2	17.2	31.6
O_3 effect (r^{2*} - r^2) ,%		9.0	16.0	16.9	8.7	23.2	14.2	9.5	7.0	9.9
O_3 significance: p<		0.002	0.000	0.000	0.001	0.000	0.000	0.001	0.005	0.000

Table 4. Contribution of ambient ozone to the integrated impact of different environmental factors on residual of pine stems basal area increment. Note: individual impact of O_3 on stem basal area increment residual: r^2=13.3% and p<0.0002. Variables (+) - p<0.05 and (–) – p>0.05 in models.

Crown defoliation (*F, %*), tree age (*A, years*) and diameter (*D, cm*) accounted for 80% of spatial and temporal variability in pine stem BAI (*Zq, cm*²). Data on correlation analysis revealed the highest significance of the effect of peak ozone concentrations on BAI residuals (p < 0.001) and the least - on acid deposition (Fig. 15). This procedure allowed elimination of the impact of the air concentration of the acidifying compounds and their deposition on BAI through the decrease in foliage thus verifying strong interaction of acid compounds with crown defoliation. These findings indicate a possible direct effect of ambient O_3 on changes in pine BAI, probably due to disturbances in CO_2 assimilation and carbohydrate movement within the trees. The elimination of the defoliation impact on tree increment by regression methods was a good example of an attempt to separate the effects of different pollutants, i.e., acidifying compounds and ambient O_3 (Table 4) (Augustaitis & Bytnerowicz, 2007d).
Integrated impact of air acidifying compounds and their deposition accounted for 18.5% of variability in BAI residual. O_3 increased the degree of the explanation by 8.7% up to 27.2%. Integrated impact of meteorological parameters accounted for 10.2% of residuals variability, and O_3 increased this rate up to 17.2%. Integrated impact of air acidifying compounds, acid deposition and meteorological parameters accounted for up to 21% of variability in BAI residual. Ozone increased this rate of explanation by approximately 10% up to 31.6%.

Climate warming might have had an additional effect enhancing the phytotoxic O_3 effect on forest. Synergistic O_3 effects with high temperature and moisture stress are well known (McLoughlin & Downing, 1996). However, the statement that the effect of O_3 and drought might counterbalance each other (Zierl, 2002) is more significant when investigating phytotoxic O_3 effect on trees. Closed stomata protect foliage from the uptake of high O_3 concentrations into the leaves, which is typical of periods characterized by high temperature and moisture stress. Most likely therefore, the O_3 effect in northern latitudes (where moisture stress is less frequent) often leads to plants becoming more susceptible to injury than in southern areas, despite the increase in O_3 concentrations from North to South (Matyssek & Innes, 1999; Karlsson et al., 2002; Paoletti, 2006).

Data from the ICP IMS, where air pollutants have been continuously monitored offered a possibility to get closer insight into O_3 effect on tree growth. Peak O_3 concentration has more significant effect on tree defoliation and increment than other indices, such as AOT40, for vegetation and forest as well as mean O_3 concentrations for a vegetation period. Therefore, more thorough studies with continuous active monitors are needed, especially in the northern countries where O_3 concentrations seldom reach the level of toxicity. However, impact of ambient O_3 on native forest ecosystems could be higher than that in the southern countries where O_3 concentrations often exceed the phytotoxic level of the AOT index, but significant relations with tree damages fail (Paoletti, E. 2006).

Recent findings clearly show that O_3 exposure does not adequately characterize the potential for plant injury, because plant response is more closely related to the amount of O_3 absorbed into leaf tissue and modified by detoxification processes (effective flux) (Matyssek et al., 2007). Newly developed concepts based on O_3 flux into leaves require profound knowledge of physiological processes, e.g. of both stomatal functioning, which determines stress avoidance through the degree of opening, and of stress tolerance, which is determined by structural and physiological leaf differentiation and related capacities in primary and secondary metabolism (Paoletti et al., 2007). Therefore, a well-coordinated and enhanced international cooperation in various disciplines such as atmospheric chemistry, forestry, botany, entomology, soil science, and dendrochronology in various regions of Europe is recommended. Since climatic changes in the Baltic region manifest themselves by the earlier (up to 15 days) beginning of the growing season, when the levels of O_3 are high and plants have high stomatal conductance, a potential for phytotoxicity is much higher than in other parts of Europe where levels of ambient O_3 are higher, but frequently occurring droughts may prevent plants from taking up high levels of O_3, thus reducing the risk of severe phytotoxic effects (Ferretti et al., 2007; Paoletti, 2006; Paoletti et al., 2007). In this context, the Baltic region seems to be a new and relevant European region for future studies on potential O_3 phytotoxicity and the evaluation of risk to temperate forest ecosystems.

Despite this, a new threat for forest ecosystem in Lithuania, changing climate, which occurs through the increase in precipitation amount during the vegetation period and mean monthly temperature from September to December, as well as a decrease in amount of precipitation during the dormant period, should mitigate the negative effect of acidifying compounds and enhance forest sustainability to unfavorable environmental factors, first of all expected increase in surface ozone concentration. Only in cases of extreme conditions such as heat and drought during the vegetation or hard frost in winter, the frequencies of which are too difficult to forecast, would not confirm our assumption.

6. Conclusions

The same detected character of changes in meteorology, surface ozone, acidifying species and pine defoliation, which from 1994 to 2001 changed towards decreasing of air pollution and improving of forest health, since 2001 adversely, indicated possible causative relationships among them. Air concentrations of SO_2, and SO_4^{2-} and NH_4^+ deposition, as well as dormant period and vegetation precipitation and mean winter temperature were shown to be the key factors most significantly affecting changes in tree crown defoliation in Lithuania.

The acidifying compounds accounted for nearly 58% of the variance in pine defoliation. Meteorological factors increased the degree of explanation to 65%, and stand and site variables to 79%. Indirect effect of acid deposition and meteorological parameters was less pronounced, however they significantly increased explanation rate of pine crown defoliation up to 89%. Indirect effect of acidifying species on birch defoliation was more significant than the direct effect through the air on leaves what allows to state that needles, which are present on trees all year round, are more efficient aerosol collectors than leaves. The death of spruce trees due to *Ips typographus* L., prevented completion of this task.

Data revealed that O_3 were among key pollutants that significantly affected tree condition in Lithuania. Correlation coefficient between temporal and spatial changes in the peak O_3 concentrations and changes in mean defoliation of Scots pine trees where the AOT40 values are commonly below their phytotoxic levels was statistically significant. However, the significance was lower than it was between defoliation and the SO_2 air concentration, approximately the same as between defoliation and the acidifying compounds in precipitation, acid deposition, and amount of precipitation, but considerably higher than between defoliation and mean air temperature. Contribution of peak O_3 concentrations to the integrated impact of acidifying compounds and meteorological parameters on pine stem growth was found to be more significant than its contribution to the integrated impact of acidifying compounds and meteorological parameters on pine defoliation

NH_4^+ air concentrations and its deposition, which show a the tendency to increase due to enhanced acidification processes in soil, with surface ozone could be the key threats to forest ecosystem in future. However, recent stable or downward tendencies in annual SO_2 air concentrations, SO_4^{2-} and NO_3^- wet deposition, as well as an increase in precipitation amount over the vegetation following the increase in mean monthly temperature and decrease in precipitation from September to December, which represented the climate change condition, should mitigate negative effect of acidifying species and enhance resiliency to phytotoxic effect of surface ozone, ensuring sustainable development of Lithuanian forest under global environmental pressures.

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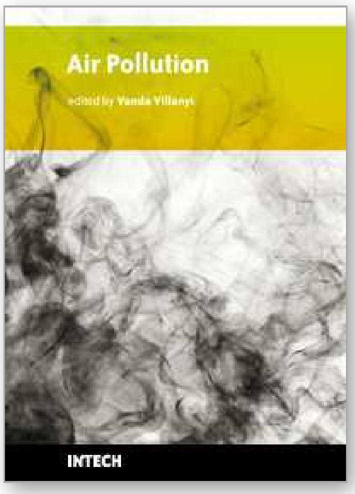

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Although the climate of the Earth is continually changing from the very beginning, anthropogenic effects, the pollution of the air by combustion and industrial activities make it change so quickly that the adaptation is very difficult for all living organisms. Researcher's role is to make this adaptation easier, to prepare humankind to the new circumstances and challenges, to trace and predict the effects and, if possible, even decrease the harmfulness of these changes. In this book we provide an interdisciplinary collection of new studies and findings on the score of air pollution.

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