

Trends of Scots pine forest health and element flow changes in the ICP Forests monitoring sites in Latvia

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Abstract

Scots pine (*Pinus sylvestris* L.) is one of the most widespread and economically most important tree species in Latvia. Tree health and element flow changes in Scots pine forests have been monitored within the International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) with assessment of crown condition and damaging agents at 70 Level I monitoring sites (mostly oligotrophic and mesotrophic *Vacciniosa*, *Myrtillosa* and *Hylocomiosa* forests) and with sampling and analyses of environmental samples at three Level II monitoring sites (*Myrtillosa* site type). All sites represent typical Scots pine forests under hemiboreal conditions in Latvia. This study presents the trends of forest health, carbon turnover and environmental condition in Scots pine forests during a 10-year period from 2009 to 2019. In general, defoliation rate in Scots pine stands remained stable during the studied period, with some yearly fluctuations, possibly related to regional insect outbreaks, especially well demonstrated in two Level II plots. The share of damaged trees varied by year from 12.8% to 19% of the total number; the main cause of damage was direct human influence. Flows of chemical elements in Scots pine forests in Level II monitoring plots were relatively stable as well, except the decreasing trend in total N concentration in deposition and SO₄-S concentration in soil solution and increasing trends in DOC concentration in soil solution that is in line with common trends in Europe. Carbon input with above-ground litter was relatively stable during the whole period; however, inter-annual variations were rather wide.

Keywords: forests on dry mineral soils, Scots pine, ICP Forests, monitoring, environmental condition, forest health, litter

Introduction

Forest is not only a characteristic element of the landscape with an important stabilizing role in the balanced development of the environment, but also a key natural resource (Baumanis et al. 2014, Nikodemus et al. 2018). The forest area and standing volume in Latvia display an increasing trend. The data of National Forest Inventory (NFI) show that as of 1 January 2019 forest area in Latvia was 3,285 thousand ha or 51% of total country area, of which 1,519 thousand ha were owned by the state (46.2% of the total forest area) and the rest of the area – 1,766 thousand ha – by other owners (53.8% of the total forest area, CSP 2019). In 2020, Latvia had the fifth highest forest cover among all EU countries, surpassed only by Finland (66%), Sweden (63%), Slovenia (61%) and Estonia (54%) (Eurostat 2021).

Scots pine (*Pinus sylvestris* L.) is one of the most common native conifer species in the Nordic and Baltic countries (Houston Durrant et al. 2016, Rytter et al. 2016, Mason and Alia 2000). In Latvia, the area of Scots pine forests now exceeds 950 thousand ha, comprising over 30% of the productive forest area (VMD 2019). Scots pine forests cover the whole Latvia territory (Laiviņš et al. 2009), although their distribution is not uniform. Scots pine is more common in the coastal areas on nutrient-poor sandy soils, but in fertile sites it is often outcompeted by other species, usually spruce or broadleaved trees (Baumanis et al. 2014, Houston Durrant et al. 2016). The Baltic region provides optimal growing conditions for Scots pine, and the trees are characterized by especially straight stems, rather narrow crowns, thin branches and good quality timber

(Baumanis et al. 2014). Thus, it is of considerable importance as a timber producing species (Rytter et al. 2016, Mason and Alia 2000).

The major stressors affecting health and vitality of European forests to be considered in their sustainable management are climate change and air pollution (Requardt et al. 2007, De Marco et al. 2014). Scots pine is tolerant towards varying environmental conditions, including drought and frost; therefore, it is suitable for a wide range of site types (Houston Durrant et al. 2016). However, its adaptability to changes in climate or environmental conditions differs among sites depending on geographical location – northern or southern parts of the distribution range, site fertility, stand origin and mixture (Houston Durrant et al. 2016, Samec et al. 2020). Over the last decade, several studies have highlighted that climate change and especially drought tend to be more important causes of Scots pine stand decline and mortality than other environmental factors (e.g. Zheng et al. 2012, Wong and Daniels 2017, Buras et al. 2018, Buras and Menzel 2019). At the same time, Scots pine has low tolerance towards atmospheric pollution (Campbell 2011), acidification exceeding critical loads and nitrogen (N) surpluses from acid deposition (Bernal et al. 2012, Samec et al. 2020) or salty sea winds (Houston Durrant et al. 2016). Scots pine meets many of the requirements of a good bioindicator plant (Yilmaz and Zengin 2004, Likus-Ciešlik et al. 2020) and is a very good archive of changes in local conditions and in ecosystems in general (Elferts 2007, Sensuła et al. 2015).

Long-term monitoring provides a valuable tool to identify status and to document changes in forest health (Paoletti et al. 2007, Lorenz et al. 2008), including the slow biogeochemical processes induced by natural and anthropogenic factors (Prietz et al. 2020). It can be used for development and evaluation of abatement strategies and for tracking possible chemical recovery from long-term inputs (Verstraeten et al. 2012). One of the most comprehensive international initiatives for this purpose is The International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests), a transnational forest monitoring and research network, launched in 1985 under the Convention on Long-range Transboundary Air Pollution of the United Nations Economic Commission for Europe (IPC Forests 2020). ICP Forests monitors forest condition in Europe at two monitoring intensity levels. Level I gathers information on forest health by using crown condition as a main tool on a systematic transnational grid of observation plots throughout Europe and beyond. Level I plots are typically integrated in the NFI system. The Level II intensive monitoring is performed on highly instrumented plots in selected forest ecosystems with the aim to better understand the processes and causes affecting the condition of forest ecosystems, as well as the effects of different anthropogenic and natural stressors (IPC Forests 2020). In Latvia, monitoring at Level I was started in 1990 (it was integrated in

the NFI system in 2009), but monitoring at Level II plots began in 2004.

The aim of the present study was to evaluate trends of forest health, carbon turnover and environmental condition in Scots pine forests in Latvia in 2009–2019, by assessing changes in air quality, crown defoliation, tree damage, tree growth and chemical element concentrations in water, litter and tree needles in ICP Forests monitoring sites to provide background information and reference for further studies.

Materials and methods

Study sites and observations

We used data on observations conducted at 70 Level I and three Level II monitoring plots in Scots pine forests in Latvia during time period 2009–2019. The Level I plots are evenly distributed across the country. The Level II plots are in south-western, central and eastern part of the country, representing the same site type but different landscape ecological regions. Locations of Scots pine forests in the Level I and Level II monitoring plots in Latvia are shown in Figure 1, characteristics of the Level I and Level II monitoring plots are summarized in Table S1 and Table S2 of supplemental material (see Appendix). All Level II plots are surrounded by a 10-m wide buffer zone. Meteorological conditions in 2009–2019 are summarized in Table S3 of supplemental material, but frequency of assessment, measurement and sampling is summarized in Table S4 of supplemental material.



Figure 1. Location of Scots pine forests at the Level I and Level II monitoring plots in Latvia

Visual assessment of crown condition and damaging agents in the Level I and Level II plots

Defoliation and damages of 15 dominant and co-dominant trees were assessed at each Level I plot each year within the period between July 1 and August 31. Defoliation, defined as needle loss in the assessable crown as compared to a reference tree, was assessed with the score of 5%

steps in the classes of 0 (no defoliation), 5 (0–5%), 10 (5–10%) and so forth. Assessable crown is determined as the part of the crown unaffected by the competition of nearby trees, excluding individual branches below the crown and branches dried off a long time ago. The widest span of the crown is the lower limit for the assessment. Tree damage symptoms, damage type, extent and causal agents responsible for the damage were determined according to Eichhorn et al. (2016). The main causal agent groups are game and grazing, insects, fungi, abiotic agents, direct human influence, fire, atmospheric pollutants and other.

Tree growth in the Level II plots

Fifteen trees of the main tree species corresponding to the 1st, 2nd and 3rd Kraft classes were selected randomly at each Level II plot for measurements. Permanent manually readable tree girth bands (UMS, accuracy 1/10 of mm) were installed slightly above breast height. The diameters of the trees through the circumference were read monthly (Dobbertin and Neumann 2016).

Monitoring of air quality in Level II plots

Air quality monitoring (Schaub et al. 2016) was performed only at one Level II plot (in Valgunde). Following variables were measured using diffusive sampling with IVL passive samplers (Swedish Environmental Research Institute, Stockholm, Sweden): nitrogen dioxide (NO₂), sulphur dioxide (SO₂), ozone (O₃), and ammonia (NH₃). Passive samplers were installed at 2 m above soil level in the open-field plot (at 200 m distance from stand). The sampling was carried out on a 2-week basis and covered the period between May 1 and October 30. Samplers were analysed at the Swedish Environmental Research Institute according to the SS-EN ISO 13395:1997/ISO 15923-1:2013, ISO 10304-1:2007, and modified ISO 11732:2005 standards. Limits of determination (LODs) for NO₂, SO₂, O₃, and NH₃ were 0.1 µg NO₂ m⁻³, 0.1 µg SO₂ m⁻³, 1 µg O₃ m⁻³, and 0.2 µg NH₃ m⁻³, respectively.

Sampling and analysis of deposition in the Level II plots

Three different types of deposition were collected (Clarke et al. 2016): bulk deposition (BOF), throughfall

deposition (THF), and stemflow deposition (STF). Bulk deposition was sampled continuously (2 replicates per plot, installed at 1.5 m height above soil level, collecting area was 0.020–0.059 m²) in the open field close to the forest stand with a continuously open plastic funnel connected to a sample bottle. The funnel also collects parts of particulate and gaseous deposition during dry periods; contributions from occult deposition are also included. Throughfall (sampled during vegetation season, 10 replicates per plot, collecting area was 0.020–0.059 m²) is the deposition sampled beneath the forest canopy and containing bulk + leached + dry deposition – adsorbed ions and contributions from occult deposition are included. Stemflow (sampled during vegetation season, 10 replicates per plot, installed at 1.3 m height, collecting area was 0.050–0.064 m²) is the deposition sampled on stems and it contains precipitation, occult deposition and leachate from the bark and leaves. Sampling was carried out at a time interval of two weeks. Analysed chemical parameters and used methods are summarized in Table 1.

Sampling and analysis of soil solution in the Level II plots

Zero-tension plate lysimeters (7 replicates per plot) were installed immediately below the organic layer at 0 cm depth, at 20–40 cm and at 40–80 cm (Nieminen et al. 2016). Sampling was carried out at a time interval of two weeks during frost-free period. Analysed chemical parameters and used methods are summarized in Table 1.

Sampling and analysis of litter in the Level II plots

Litter was sampled from 10 collectors placed randomly at each Level II plot under uniform forest canopy (Ukonmaanaho et al. 2016). Collector design was represented by a solid funnel (0.7 m deep) with a bag of inert material (polyethylene) with mesh size of 0.2 mm, the collecting area of individual traps were 0.25 m² in 2009–2018 and 0.42 m² since 2019 in Valgunde, and 0.50 m² in Taurene and Rucava. Within the observation period, the initial collectors were replaced with collectors having larger collecting area to obtain a larger amount of litter samples for chemical analysis. Litter was collected

Table 1. Analysed chemical parameters and used methods for deposition and soil solution samples

Parameter, unit	Method / Instrument	Standard
Conductivity, µS cm ⁻¹	Conductimetry at 25 °C	LVS EN 27888:1993
pH	Potentiometry	LVS EN ISO 10523:2012
Dissolved organic carbon (DOC), mg L ⁻¹	Infrared spectroscopy after oxidation to CO ₂	LVS EN 1484:2000
Total nitrogen (TN), mg L ⁻¹	Total N analyser with chemiluminescence detection	LVS EN 12260:2004
Nitrate nitrogen (NO ₃ -N), mg L ⁻¹	Ion chromatography	LVS EN ISO 10304-1:2009
Ammonium nitrogen (NH ₄ -N), mg L ⁻¹	Spectrophotometry	LVS ISO 7150-1:1984
Potassium (K), magnesium (Mg), calcium (Ca), sodium (Na), mg L ⁻¹	AAS Flame, AES Flame (for Na and K)	LVS EN ISO 7980:2000, LVS ISO 9964-3:2000
Sulphate sulphur (SO ₄ -S), mg L ⁻¹	Ion chromatography	LVS EN ISO 10304-1:2009
Phosphate phosphorus (PO ₄ -P), mg L ⁻¹	Spectrophotometry	LVS EN ISO 6878:2005

Table 2. Analysed chemical parameters and used methods for litter and needles

Parameter, unit	Method / Instrument	Standard
Carbon (C), g kg ⁻¹	Element-analyzer	LVS ISO 10694:2006
Nitrogen (N), g kg ⁻¹	Titration after Kjeldahl digestion	LVS ISO 11261:2002
Sulphur (S), g kg ⁻¹	Element-analyzer	ISO 15178:2000
Phosphorus (P), g kg ⁻¹	Microwave pressure digestion – closed system (nitric acid), spectrophotometry	ISO 16729:2013, LVS 398:2002
Potassium (K), magnesium (Mg), calcium (Ca), sodium (Na), g kg ⁻¹	Microwave pressure digestion – closed system (nitric acid), ICP-OES	ISO 16729:2013, LVS ISO 11047:1998, LVS EN 16170

on a monthly basis. In 2009–2017, litter collected from the Level II plots was sorted into the following fractions: (i) wood fraction (twigs < 2 cm diameter, branches, bark); (ii) foliar litter (*Pinus sylvestris* L.); (iii) fruits/seeds total (all species); (iv) other biomass (lichen, moss, etc.). Since 2018 litter was sorted into more divided fractions: (i) wood fraction (twigs < 2 cm diameter, branches, bark); (ii) foliar litter (*Pinus sylvestris* L.); (iii) foliar litter (other tree species); (iv) fruits/seeds total incl. green cones (*Pinus sylvestris* L.); (v) fruits/seeds total incl. green cones (other species); (vi) other biomass (lichen, moss, etc.). During the observation period, diversification of litter fraction was increased to obtain more detailed information, but, if necessary, the fractions can be pooled and compared within all observation periods. The analysed chemical parameters and methods used are summarized in Table 2.

Sampling and analysis of needles in the Level II plots

Sample trees (8–9 trees) of the main species were randomly selected at each Level II plot (Rautio et al. 2016). Both the current year needles and the second-year needles were taken from the upper third of the crown, but not from the very first whorls. Only needles developed in light (south-facing branches) were sampled, shaded parts of the canopy were excluded from sampling. Sampling was carried out after the end of vegetation season using climbing equipment and stainless-steel hand saw. Sampling and analysis were performed every second year. The analysed chemical parameters and methods used are summarized in Table 2.

Statistical analysis

Data processing and all statistical analyses were performed in the R environment (R Core Team 2017). Statistical differences of average values were analysed with the pairwise comparison using *t* test with pooled *SD* (function *pairwise.t.test()*). In the case of small number of repetitions in the data set (less than 30), a non-parametrical statistical method was used (the Wilcoxon rank sum test with continuity corrections or the Wilcoxon signed rank test). Trends during the selected period were evaluated using regression analysis. A 95% confidence level was used for all analyses.

Results

Crown condition and damaging agents

Defoliation of Scots pine at the Level I plots is shown in Figure 2. Both at the Level I and Level II monitoring plots, neither increasing nor decreasing trends in defoliation of Scots pine trees were detected within the observation period. Average defoliation of whole crown of Scots pine at the Level I plots was $20.1 \pm 0.1\%$ (2009–2019), but at the Level II plots defoliation of the upper part (upper third) of crown of Scots pine was $15.5 \pm 0.3\%$ in Valgunde (2009–2019), $22.8 \pm 0.7\%$ in Taurene (2015–2019), and $23.8 \pm 1.6\%$ in Rucava (2019). At the Level I plots, on average, 16% of all monitored trees were affected by damaging agents. The extent of damage (percentage of damaged trees from the total number of assessed trees) varied by year. It was the lowest in 2012 – 12.8%, but the highest in 2015 and 2016, 19% and 18.1%, respectively. 2015 and 2016 were also years with comparatively high defoliation rate for Scots pine – on average 20.5% and 20.3%, respectively. During these two years damage by insects was quite frequently recorded, as well, if compared to other observation years. This is related to an increase in the population and damages by European pine sawfly, *Neodiprion sertifer* (Michel and Seidling 2016). In general, the most frequently recorded damaging agent was the direct human influence (42% of all damaging cases on average, Figure 3), mostly harvesting. Comparatively high defoliation rate for Scots pine at the Level II plots in Taurene and Rucava is mostly related to bark beetle (*Tomicus piniperda*) damage

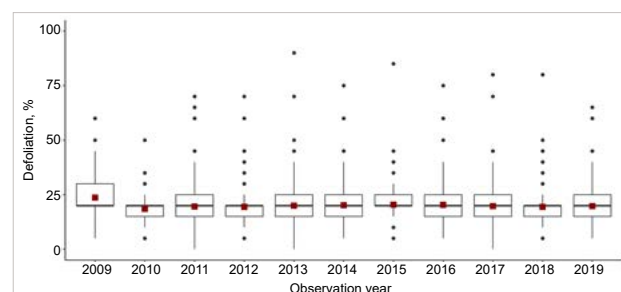


Figure 2. Defoliation of Scots pine at the Level I plots in Latvia. In the boxplots, the median is shown by the bold line, the mean is shown by the dark red square, the box corresponds to the lower and upper quartiles, whiskers show the minimal and maximal values (within 150% of the interquartile range from the median) and black dots represent outliers of the datasets

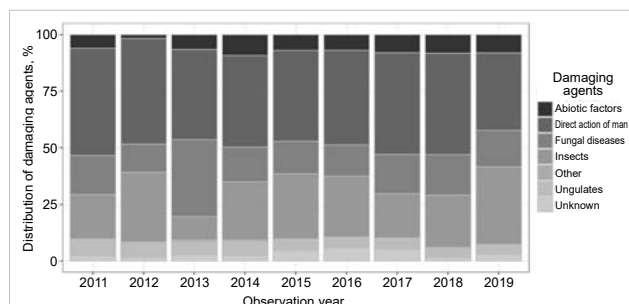


Figure 3. Distribution of damaging agents in Scots pine forests at the Level I plots in Latvia

(Laiviņš et al., 2019). It is especially evident in Rucava. There, surface fire in the nearby stand occurred in 2016 (Bārdule et al. 2017), and subsequent bark beetle outbreak and increase of the defoliation rate in the part of the plot of the closest to the damaged stand was observed in the following years. Mean defoliation rate in this subplot reached $27.3 \pm 2.6\%$ in 2019.

Tree growth

Increment of breast height diameter (BHD) of Scots pine at the Level II plots is shown in Figure 4. In Valgunde, average increment of BHD of Scots pine was $1.36 \pm 0.06 \text{ mm yr}^{-1}$ in 2009–2019 and $1.42 \pm 0.09 \text{ mm yr}^{-1}$ in 2016–2019. In Taurene, average increment of BHD of Scots pine was $1.04 \pm 0.10 \text{ mm yr}^{-1}$ (2016–2019), but $2.19 \pm 0.12 \text{ mm yr}^{-1}$ in Rucava (2016–2019). The results of analysis of variance showed statistically significant differences in the increment of BHD of Scots pine between the Le-

vel II plots ($p < 0.001$), but no clear impact of other environmental factors (e.g. annual average air temperature, input of nutrients through deposition or chemical composition of soil solution) on increment of BHD was detected when analysing each individual year separately. When analysing the Level II plot average values of increment of BHD of Scots pine for the period 2016–2019 and comparing between the

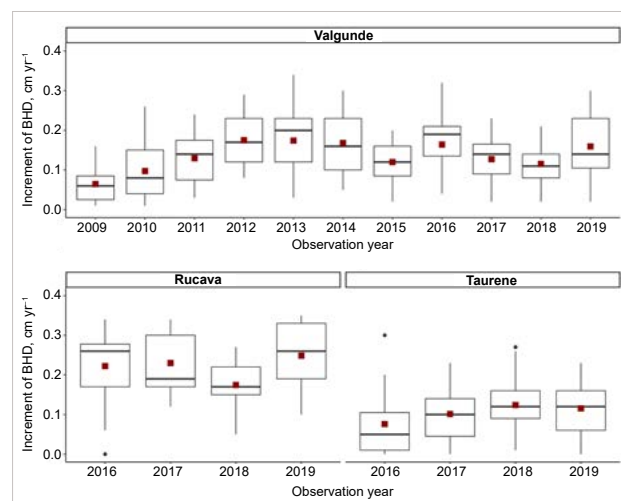
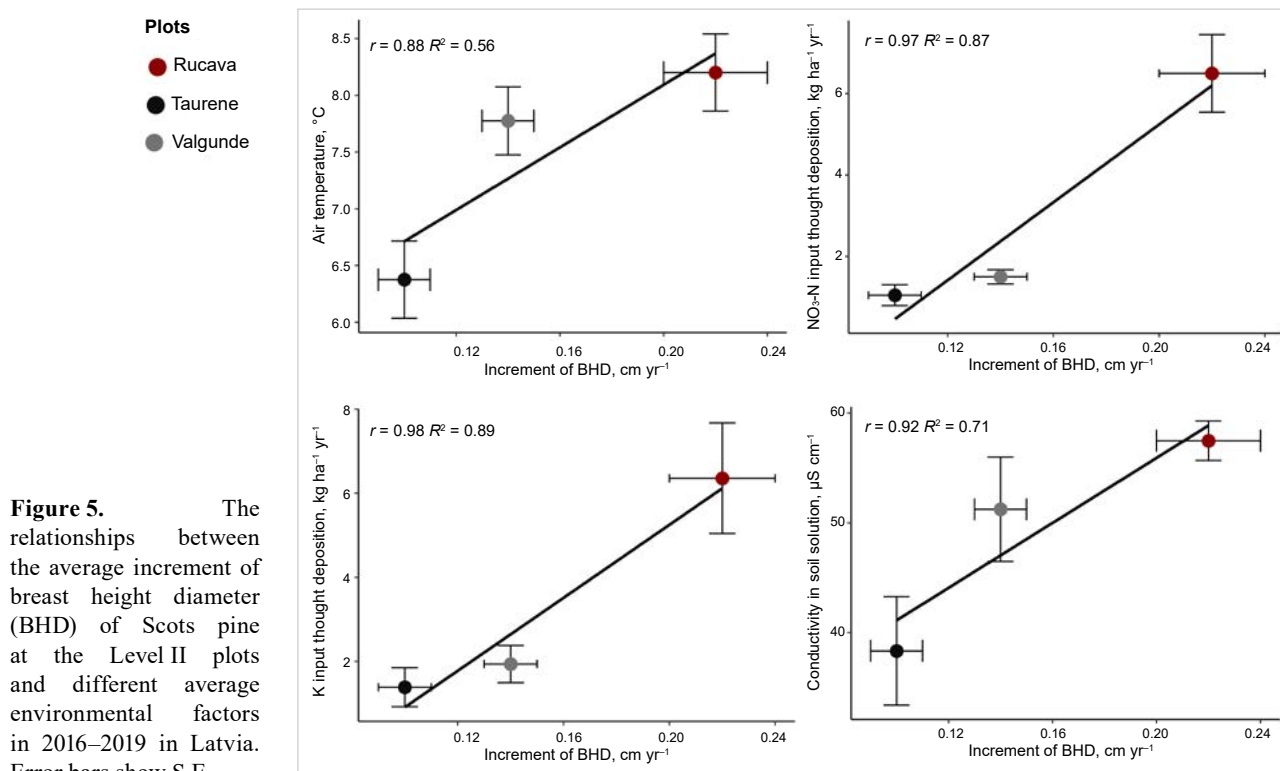


Figure 4. Increment of breast height diameter (BHD) of Scots pine at Level II plots in Latvia. In the boxplots, the median is shown by the bold line, the mean is shown by the dark red square, the box corresponds to the lower and upper quartiles, whiskers show the minimal and maximal values (within 150% of the interquartile range from the median) and black dots represent outliers of the datasets



Level II plots, several tendencies were highlighted – larger average increment of BHD was observed at Level II plots where average air temperature was higher and input of nutrients through deposition and concentrations in soil solution was larger (some of the relationships are shown in Figure 5).

Air quality

Figure 6 shows the vegetation season mean concentrations of NO_2 , SO_2 , O_3 , and NH_3 at the Level II plot in Valgunde in 2009–2019. No significant trends in the air quality parameters were observed during 2009–2019. Vegetation season mean NO_2 concentration was within the range from $1.4 \pm 0.1 \mu\text{g m}^{-3}$ to $2.5 \pm 0.2 \mu\text{g m}^{-3}$, but individual maximum NO_2 concentration ($3.6 \mu\text{g m}^{-3}$) was observed in October 2018. Vegetation season mean SO_2 concentration was within the range from $0.26 \pm 0.03 \mu\text{g m}^{-3}$ to $0.56 \pm 0.04 \mu\text{g m}^{-3}$, but individual maximum SO_2 concentration ($0.94 \mu\text{g m}^{-3}$) was observed in April 2009. Vegetation season mean NH_3 concentration was within the range from $1.2 \pm 0.1 \mu\text{g m}^{-3}$ to $12.8 \pm 3.4 \mu\text{g m}^{-3}$, but individual maximum NH_3 concentration ($30.4 \mu\text{g m}^{-3}$) was observed in November 2013. Vegetation seasons mean O_3 concentration was within the range from $36.7 \pm 2.4 \mu\text{g m}^{-3}$ to $46.9 \pm 3.1 \mu\text{g m}^{-3}$, but individual maximum O_3 concentration ($104.5 \mu\text{g m}^{-3}$) was observed in June 2011. Concentrations of gaseous air pollutants varied in a quite constant range with a few peaks (e.g. in 2013 for NH_3), but in general no increasing or decreasing trends during period of 2009–2019 were observed.

Deposition

Figure 7 shows chemical composition of BOF, THF, and STF at the Level II monitoring plot in Valgunde in 2009–2019. Average concentrations of DOC, TN, K, $\text{PO}_4\text{-P}$, $\text{SO}_4\text{-S}$, and conductivity were significantly higher ($p < 0.05$) in STF ($96.4 \pm 4.8 \text{ mg DOC L}^{-1}$, $5.08 \pm 0.53 \text{ mg TN L}^{-1}$, $6.16 \pm 0.34 \text{ mg K L}^{-1}$, $0.074 \pm 0.015 \text{ mg PO}_4\text{-P L}^{-1}$, $0.85 \pm 0.09 \text{ mg SO}_4\text{-S L}^{-1}$, $78.6 \pm 3.2 \mu\text{S cm}^{-1}$, respectively) than in BOF ($4.98 \pm 0.48 \text{ mg DOC L}^{-1}$, $1.51 \pm 0.16 \text{ mg TN L}^{-1}$, $0.43 \pm 0.06 \text{ mg K L}^{-1}$, $0.034 \pm 0.009 \text{ mg PO}_4\text{-P L}^{-1}$, $0.41 \pm 0.02 \text{ mg SO}_4\text{-S L}^{-1}$, $20.3 \pm 1.6 \mu\text{S cm}^{-1}$, respectively) and THF ($9.80 \pm 0.70 \text{ mg DOC L}^{-1}$, $2.28 \pm 0.21 \text{ mg TN L}^{-1}$, $1.24 \pm 0.11 \text{ mg K L}^{-1}$, $0.035 \pm 0.006 \text{ mg PO}_4\text{-P L}^{-1}$, $0.46 \pm 0.03 \text{ mg SO}_4\text{-S L}^{-1}$, $24.6 \pm 1.2 \mu\text{S cm}^{-1}$, respectively). Average pH values of deposition decreased

Table 3. Statistical data (correlation coefficient r , p -value and adjusted R^2 of linear regression) on the trends characterizing changes in concentration of sulphates and total nitrogen in deposition and input of total nitrogen through bulk deposition at the Level II monitoring plot in Valgunde (Latvia) for the period of 2009–2019

Dependent variable	Independent variable	r	p	Adjusted R^2 of linear regression
$\text{SO}_4\text{-S}$ concentration in STF	Time	-0.68	0.021	0.41
TN concentration in BOF	Time	-0.75	0.008	0.52
TN concentration in THF	Time	-0.75	0.008	0.51
TN concentration in STF	Time	-0.73	0.011	0.48
Input of TN through BOF	Time	-0.78	0.004	0.57

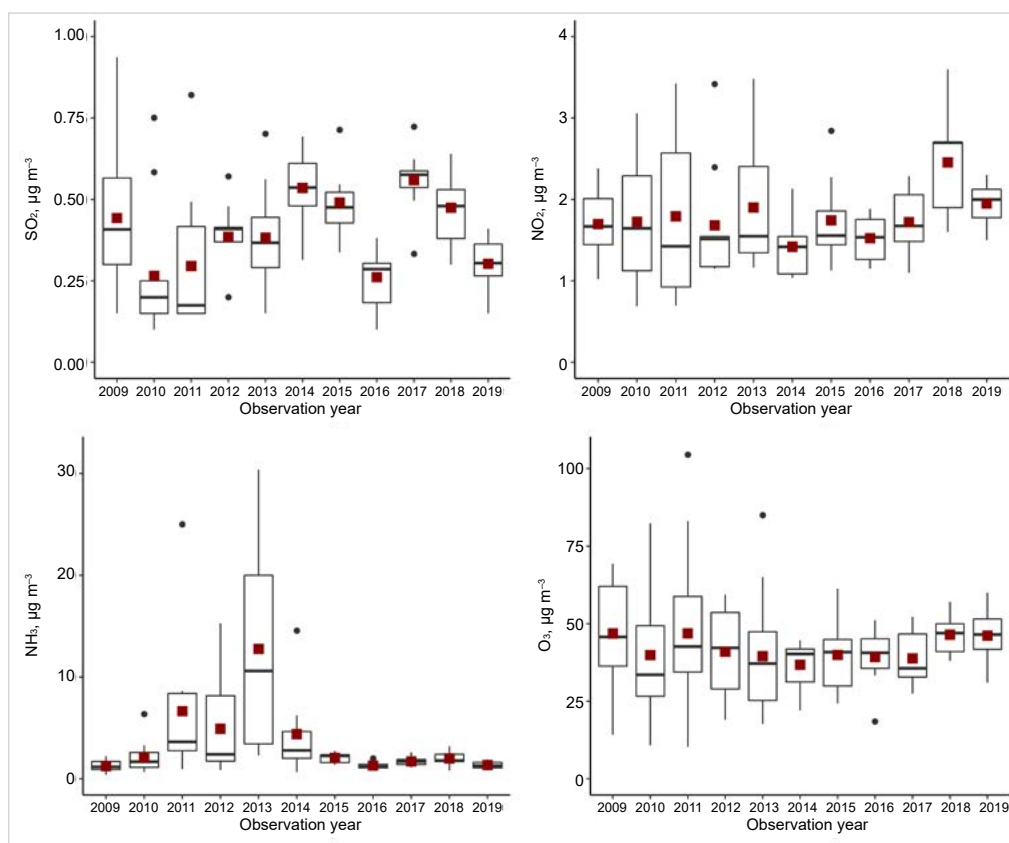


Figure 6. Air quality at the Level II monitoring plot in Valgunde (Latvia). In the boxplots, the median is shown by the bold line, the mean is shown by the dark red square, the box corresponds to the lower and upper quartiles, whiskers show the minimal and maximal values (within 150% of the interquartile range from the median) and black dots represent outliers of the datasets

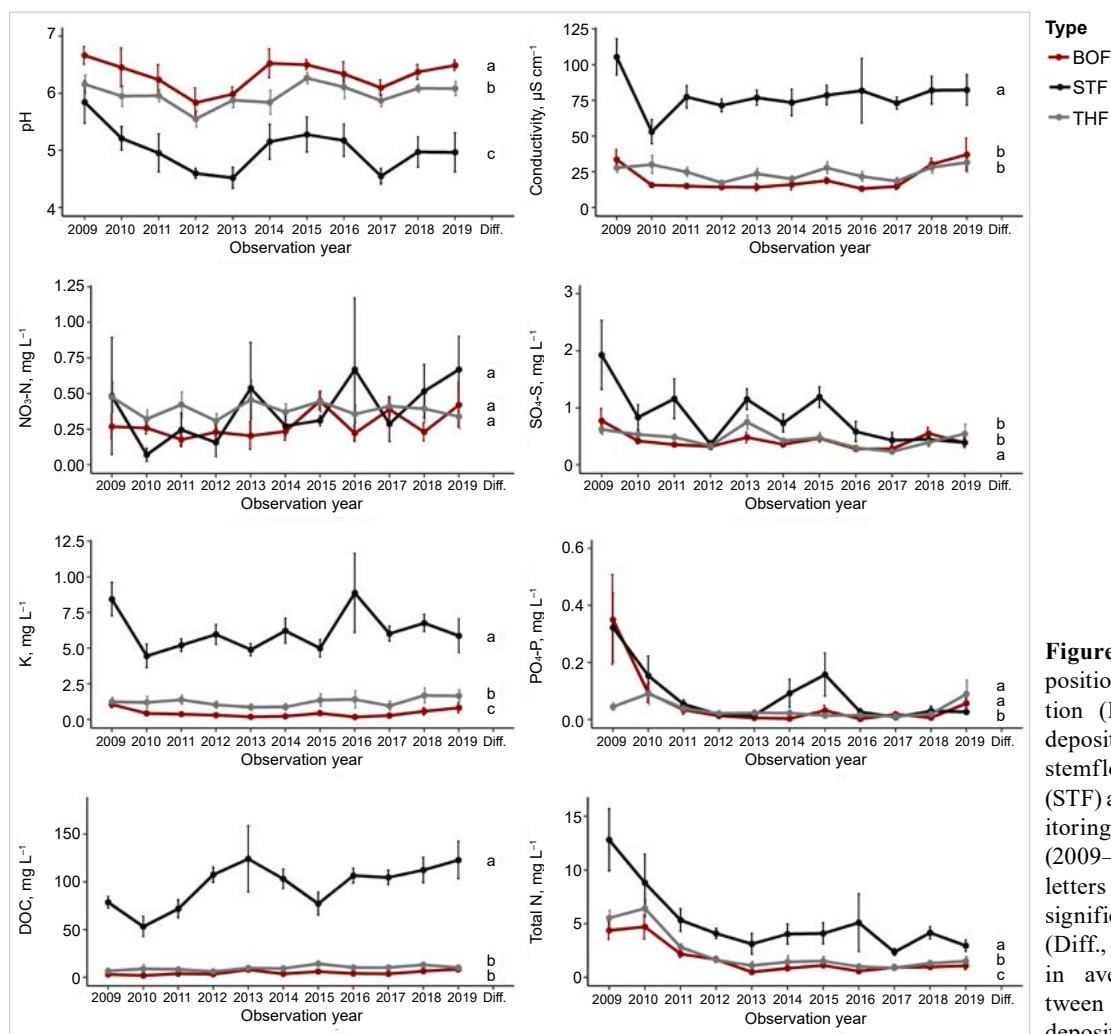
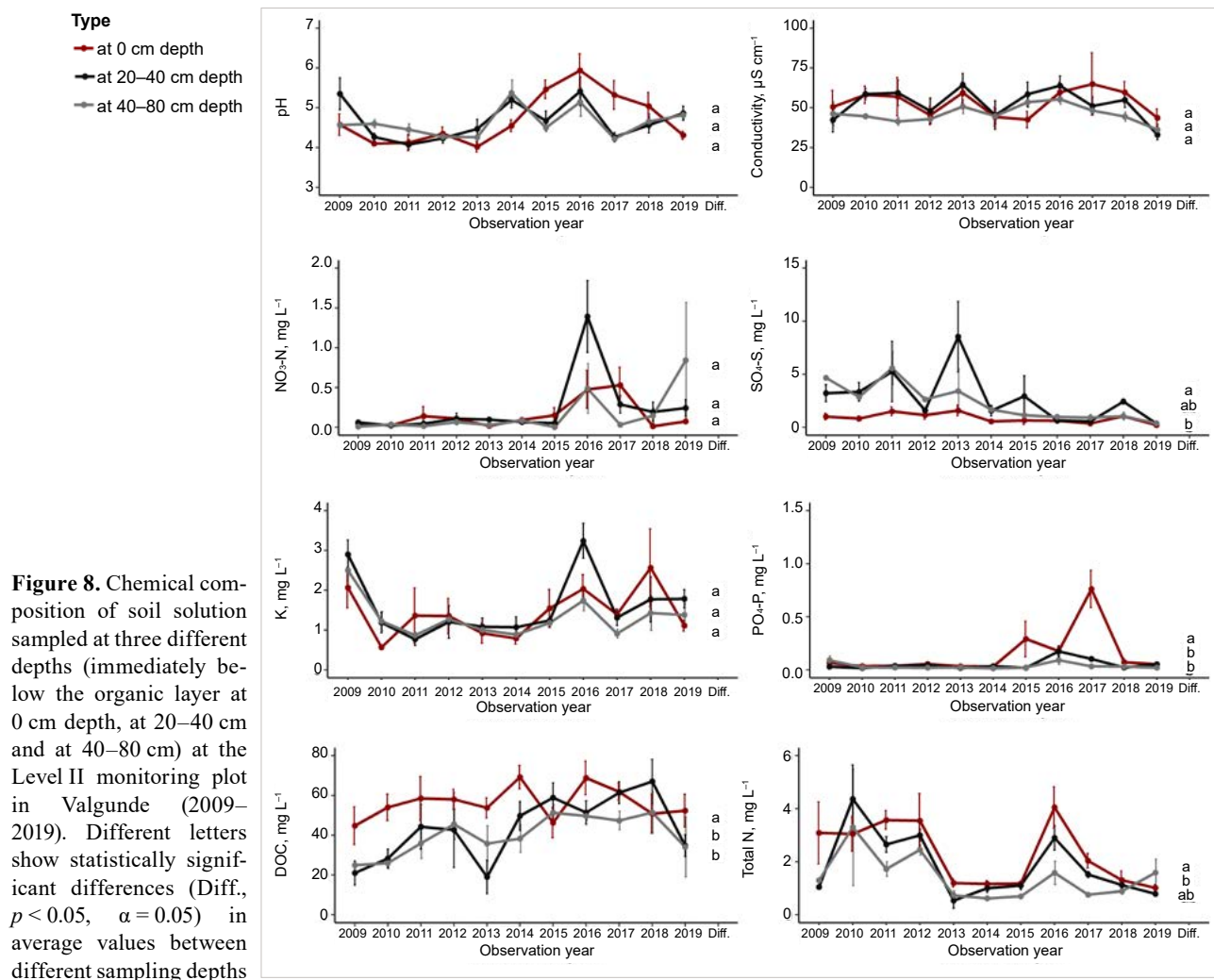


Figure 7. Chemical composition of bulk deposition (BOF), throughfall deposition (THF), and stemflow deposition (STF) at the Level II monitoring plot in Valgunde (2009–2019). Different letters show statistically significant differences (Diff., $p < 0.05$, $\alpha = 0.05$) in average values between different types of deposition

Table 4. Input of macroelements ($\text{kg ha}^{-1} \text{ yr}^{-1}$) through bulk deposition at Level II monitoring plots in Latvia. Different letters indicate statistically significant differences ($p < 0.05$, $\alpha = 0.05$) in average values between the different Level II monitoring plots for the period of 2016–2019

Element	Value	Level I monitoring plot			
		Valgunde		Taurene	Rucava
		2009–2019	2016–2019	2016–2019	2016–2019
TN	mean $\pm$ S.E.	12.3 $\pm$ 4.1	4.0 $\pm$ 0.3 ^a	3.4 $\pm$ 0.7 ^a	14.8 $\pm$ 4.0 ^b
	range (min...max)	3.2...44.3	3.4...4.8	1.8...5.3	6.2...25.4
NO ₃ -N	mean $\pm$ S.E.	1.7 $\pm$ 0.1	1.5 $\pm$ 0.2 ^a	1.0 $\pm$ 0.3 ^a	6.5 $\pm$ 1.0 ^b
	range (min...max)	1.2...2.3	1.2...1.9	0.5...1.6	3.8...8.1
NH ₄ -N	mean $\pm$ S.E.	2.7 $\pm$ 0.6	1.3 $\pm$ 0.2 ^a	1.3 $\pm$ 0.4 ^a	4.8 $\pm$ 1.2 ^b
	range (min...max)	0.8...7.6	0.8...1.6	0.4...2.0	1.6...7.3
PO ₄ -P	mean $\pm$ S.E.	0.4 $\pm$ 0.2	0.1 $\pm$ 0.1 ^a	0.3 $\pm$ 0.1 ^a	2.9 $\pm$ 1.1 ^b
	range (min...max)	< 0.1...1.8	< 0.1...0.4	< 0.1...0.7	0.4...5.9
SO ₄ -S	mean $\pm$ S.E.	2.6 $\pm$ 0.2	1.9 $\pm$ 0.2 ^a	2.0 $\pm$ 0.2 ^a	4.1 $\pm$ 0.8 ^b
	range (min...max)	1.6...3.8	1.6...2.3	1.7...2.5	1.8...5.6
K	mean $\pm$ S.E.	2.7 $\pm$ 0.5	1.9 $\pm$ 0.4 ^a	1.4 $\pm$ 0.5 ^a	6.4 $\pm$ 1.3 ^b
	range (min...max)	1.1...5.5	1.1...2.8	0.7...2.7	2.4...8.0
Ca	mean $\pm$ S.E.	7.4 $\pm$ 1.3	10.6 $\pm$ 2.9 ^a	6.9 $\pm$ 1.3 ^a	6.4 $\pm$ 1.5 ^a
	range (min...max)	3.3...16.5	4.1...14.1	4.8...10.5	3.1...10.2
Mg	mean $\pm$ S.E.	1.9 $\pm$ 0.1	1.7 $\pm$ 0.2 ^a	1.5 $\pm$ 0.2 ^a	2.6 $\pm$ 0.3 ^b
	range (min...max)	1.4 ... 2.5	1.4 ... 2.2	1.1 ... 1.9	1.9 ... 3.1
Na	mean $\pm$ S.E.	3.1 $\pm$ 0.2	3.6 $\pm$ 0.2 ^a	5.2 $\pm$ 1.2 ^a	11.9 $\pm$ 2.7 ^b
	range (min...max)	1.9 ... 4.0	3.4 ... 4.0	3.5 ... 8.7	6.6 ... 17.8
DOC	mean $\pm$ S.E.	30.0 $\pm$ 3.4	30.1 $\pm$ 0.9 ^a	25.3 $\pm$ 2.0 ^a	33.0 $\pm$ 7.0 ^a
	range (min...max)	14.8...59.7	22.4...36.5	21.1...29.1	13.4...45.9



significantly in the following order: BOF (6.31 ± 0.06) > THF (5.98 ± 0.05) > STF (5.04 ± 0.09). $\text{SO}_4\text{-S}$ concentration in stemflow deposition and TN concentration in all types of deposition in the Level II monitoring plot in Valgunde have substantially changed (decreased) in the period of 2009–2019. Table 3 shows the statistical data on the trends. Input of macroelements through bulk deposition at the different Level II monitoring plots in Latvia is shown in Table 4. If the different Level II monitoring plots were compared, the average input of macroelements, except Ca and DOC, was significantly higher in the Level II monitoring plot in Rucava.

Soil solution

Figure 8 shows chemical composition of soil solution sampled at three different depths (immediately below the organic layer at 0 cm depth, at 20–40 cm and at 40–80 cm) at the Level II monitoring plot in Valgunde in 2009–2019. Results show statistically significant differences in the average values of several parameters ($\text{SO}_4\text{-S}$, $\text{PO}_4\text{-P}$, DOC, and TN) between different sampling depths. Average concentrations of DOC, TN, and $\text{PO}_4\text{-P}$ were significantly

higher ($p < 0.05$) immediately below the organic layer at 0 cm depth (56.7 ± 2.5 mg DOC L^{-1} , 2.2 ± 0.2 mg TN L^{-1} , 0.27 ± 0.07 mg $\text{PO}_4\text{-P}$ L^{-1} , respectively) than at 20–80 cm depth (45.2 ± 1.9 mg DOC L^{-1} , 1.5 ± 0.1 mg TN L^{-1} , 0.10 ± 0.01 mg $\text{PO}_4\text{-P}$ L^{-1} , respectively). The average chemical composition of soil solution at the different Level II monitoring plots in Latvia is shown in Table 5. Statistically significant differences in average values between the plots were observed for several parameters (e.g. pH, conductivity, $\text{NO}_3\text{-N}$, DOC, Ca, Mg, Na).

Litter

Figure 9 shows dynamics of foliar litter biomass in Valgunde (2009–2019), as well as the comparison of variation of foliar litter biomass at the different Level II monitoring plots in Latvia. Total foliar litter biomass at Level II monitoring plots was within the range from $2198 \text{ kg ha}^{-1} \text{ yr}^{-1}$ to $6085 \text{ kg ha}^{-1} \text{ yr}^{-1}$. Comparing amounts of foliar litter biomass of different fractions between the Level II monitoring plots, a statistically larger fraction of Scots pine needles was observed in Rucava ($p < 0.01$), where the number of

Table 5. Average chemical composition of soil solution at Level II monitoring plots in Latvia. Different letters indicate statistically significant differences ($p < 0.05$, $\alpha = 0.05$) in average values between different Level II monitoring plots for the period of 2016–2019

Parameter, unit	Value	Level II monitoring plot			
		Valgunde		Taurene	Rucava
		2009–2019	2016–2019	2016–2019	2016–2019
pH	mean $\pm$ S.E.	4.72 $\pm$ 0.05	4.86 $\pm$ 0.09 ^a	5.33 $\pm$ 0.08 ^b	4.90 $\pm$ 0.10 ^a
	range (min...max)	3.70...7.94	3.85...7.94	3.74...6.81	3.97...6.43
Conductivity, $\mu\text{S cm}^{-1}$	mean $\pm$ S.E.	50.9 $\pm$ 1.3	51.1 $\pm$ 2.1 ^a	38.8 $\pm$ 2.2 ^b	58.3 $\pm$ 2.6 ^a
	range (min...max)	21.6...160.1	21.6...160.1	14.5...113.8	32.4...90.9
TN, mg L ⁻¹	mean $\pm$ S.E.	1.78 $\pm$ 0.12	1.60 $\pm$ 0.15 ^a	1.43 $\pm$ 0.15 ^a	1.40 $\pm$ 0.13 ^a
	range (min...max)	0.10...6.60	0.40...5.70	0.34...9.99	0.34...4.41
NO ₃ -N, mg L ⁻¹	mean $\pm$ S.E.	0.20 $\pm$ 0.04	0.39 $\pm$ 0.08 ^a	0.15 $\pm$ 0.03 ^b	0.04 $\pm$ 0.01 ^b
	range (min...max)	0.00...3.20	0.00...3.20	0.00...1.97	0.00...0.27
NH ₄ -N, mg L ⁻¹	mean $\pm$ S.E.	0.28 $\pm$ 0.07	0.42 $\pm$ 0.11 ^a	0.26 $\pm$ 0.03 ^a	0.14 $\pm$ 0.03 ^a
	range (min...max)	0.00...5.56	0.00...5.56	0.00...1.79	0.00...0.65
PO ₄ -P, mg L ⁻¹	mean $\pm$ S.E.	0.08 $\pm$ 0.01	0.12 $\pm$ 0.02 ^a	0.08 $\pm$ 0.02 ^a	0.03 $\pm$ 0.01 ^a
	range (min...max)	0.00...1.21	0.00...1.21	0.00...1.54	0.00...0.23
SO ₄ -S, mg L ⁻¹	mean $\pm$ S.E.	1.82 $\pm$ 0.24	0.69 $\pm$ 0.08 ^a	0.66 $\pm$ 0.07 ^a	0.78 $\pm$ 0.14 ^a
	range (min...max)	0.08...13.72	0.08...2.66	0.14...4.06	0.26...2.51
K, mg L ⁻¹	mean $\pm$ S.E.	1.46 $\pm$ 0.07	1.71 $\pm$ 0.13 ^a	1.62 $\pm$ 0.13 ^a	1.74 $\pm$ 0.16 ^a
	range (min...max)	0.15...5.14	0.50...5.14	0.14...5.98	0.64...4.24
Ca, mg L ⁻¹	mean $\pm$ S.E.	0.60 $\pm$ 0.06	0.79 $\pm$ 0.11 ^a	1.30 $\pm$ 0.14 ^b	1.63 $\pm$ 0.18 ^b
	range (min...max)	0.01...4.95	0.04...4.47	0.22...6.92	0.16...4.12
Mg, mg L ⁻¹	mean $\pm$ S.E.	0.90 $\pm$ 0.07	1.32 $\pm$ 0.14 ^a	0.70 $\pm$ 0.03 ^b	0.70 $\pm$ 0.06 ^b
	range (min...max)	0.25...4.30	0.37...4.30	0.15...2.09	0.00...1.54
Na, mg L ⁻¹	mean $\pm$ S.E.	1.89 $\pm$ 0.11	2.08 $\pm$ 0.10 ^a	1.62 $\pm$ 0.07 ^b	3.94 $\pm$ 0.18 ^c
	range (min...max)	0.24...11.91	0.75...4.30	0.65...4.98	2.27...6.38
DOC, mg L ⁻¹	mean $\pm$ S.E.	48.9 $\pm$ 1.6	53.0 $\pm$ 2.3 ^a	40.7 $\pm$ 3.4 ^b	54.7 $\pm$ 4.5 ^a
	range (min...max)	5.0...95.7	5.3...95.7	8.2...153.5	6.8...112.5

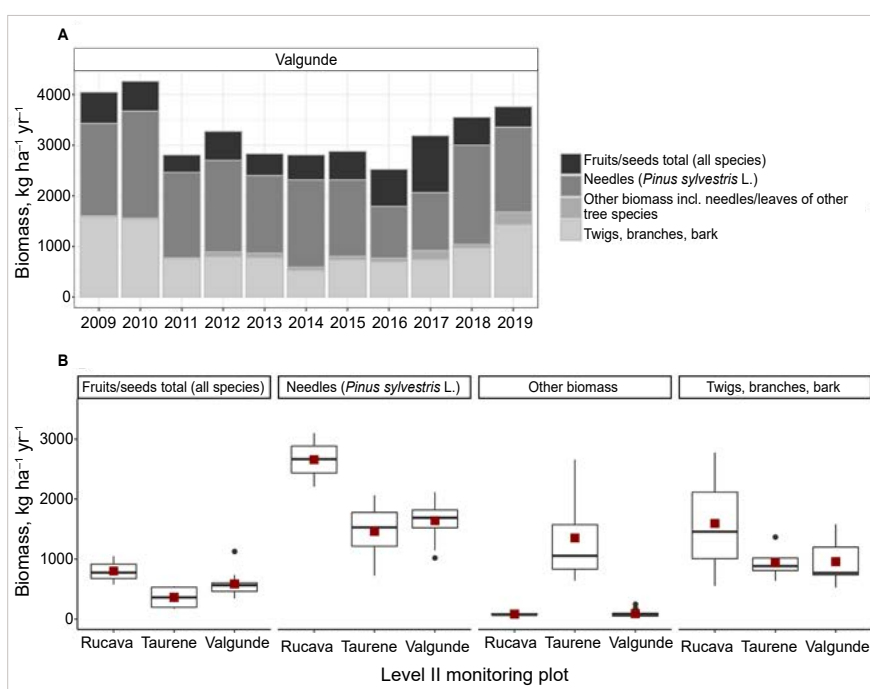


Figure 9. Foliar litter biomass at Level II monitoring plots in Latvia; A – Dynamic of foliar litter biomass for the period of 2016–2019 in Valgunde, B – Comparison of biomass of different foliar litter fractions between the Level II monitoring plots. In the boxplots, the median is shown by the bold line, the mean is shown by the dark red square, the box corresponds to the lower and upper quartiles, whiskers show the minimal and maximal values (within 150% of the interquartile range from the median) and black dots represent outliers of the datasets

Scots pine trees per ha is the lowest (Table S2 of supplemental material), but defoliation rate is the largest. Statistically larger fraction of other biomass (lichen, moss, insects, and faeces), including needles and leaves of other tree species, was observed in Taurene ($p < 0.01$), where admixture of other tree species (Norway spruce and silver birch) is the highest. Statistically

significant differences in biomass of fruits and seeds and in biomass of twigs, branches and bark between the different Level II monitoring plots were not observed.

Input of macroelements through foliar litter biomass at the different Level II monitoring plots in Latvia is summarized in Table 6. Statistically significant differences in input of macroelements through foliar litter biomass be-

Table 6. Input of macroelements ($\text{kg ha}^{-1} \text{ yr}^{-1}$) through litter biomass at Level II monitoring plots in Latvia. Different letters indicate statistically significant differences ($p < 0.05$, $\alpha = 0.05$) in average values between different Level II monitoring plots for the period of 2016–2019

Element	Value	Level II monitoring plot			
		Valgunde		Taurene	Rucava
		2009–2019	2016–2019	2016–2019	2016–2019
N	mean $\pm$ S.E.	24.4 ± 1.7	27.3 ± 1.2^a	32.9 ± 7.1^a	36.7 ± 3.7^a
	range (min...max)	16.2...32.6	23.9...29.4	17.7...52.0	29.3...40.5
P	mean $\pm$ S.E.	1.6 ± 0.2	1.7 ± 0.3^a	3.0 ± 0.4^a	2.4 ± 0.3^a
	range (min...max)	1.0...2.7	1.3...2.5	2.2...3.9	1.8...2.8
S	mean $\pm$ S.E.	1.7 ± 0.2	1.7 ± 0.2^a	3.1 ± 0.9^a	2.8 ± 0.3^a
	range (min...max)	0.5...3.0	1.2...2.2	2.1...4.9	2.2...3.3
K	mean $\pm$ S.E.	4.2 ± 0.2	4.6 ± 0.3^a	5.5 ± 1.1^a	5.9 ± 0.8^a
	range (min...max)	3.4...5.2	4.0...5.2	2.4...7.1	4.3...7.1
Ca	mean $\pm$ S.E.	20.9 ± 1.6	19.2 ± 1.9^a	21.1 ± 2.9^a	28.5 ± 2.6^a
	range (min...max)	14.6...29.4	14.9...24.0	13.8...26.0	23.4...31.7
Mg	mean $\pm$ S.E.	2.6 ± 0.2	2.6 ± 0.3^a	3.3 ± 0.5^a	2.9 ± 0.5^a
	range (min...max)	1.9...4.6	1.9...3.1	2.0...4.3	1.9...3.4
C	mean $\pm$ S.E.	1776 ± 90	1723 ± 135^a	2023 ± 365^a	2940 ± 453^a
	range (min...max)	1349...2389	1349...1995	1105...2892	2046...3511

tween the different Level II monitoring plots were not observed ($p > 0.05$).

Total C concentration in foliar litter was within the range from 463.4 g kg^{-1} (litter fraction of other biomass) to 593.5 g kg^{-1} (litter fraction of fruits and seeds). Total C input through foliar litter biomass at the Level II monitoring plots was within the range from $1,105 \text{ kg ha}^{-1} \text{ yr}^{-1}$ to $3,511 \text{ kg ha}^{-1} \text{ yr}^{-1}$, but average total C input through foliar litter was $2,025 \pm 150 \text{ kg ha}^{-1} \text{ yr}^{-1}$.

Needles

Statistically significant differences in concentrations of macroelements in Scots pine needles between the Level II monitoring plots in Latvia were not observed.

Variation of concentrations of macroelements in current and previous year needles of Scots pine at Level II monitoring plots in Latvia is shown in Figure 10. Statistically higher concentrations of K, P, and Mg were observed in current year needles of Scots pine if compared to previous year needles ($p = 0.02$, $p < 0.01$, $p < 0.01$, respectively), but statistically higher Ca concentration (Figure 10) was observed in previous year needles ($p < 0.001$). Statistically significant differences in concentrations of N and S between current and previous year needles of Scots pine was not observed.

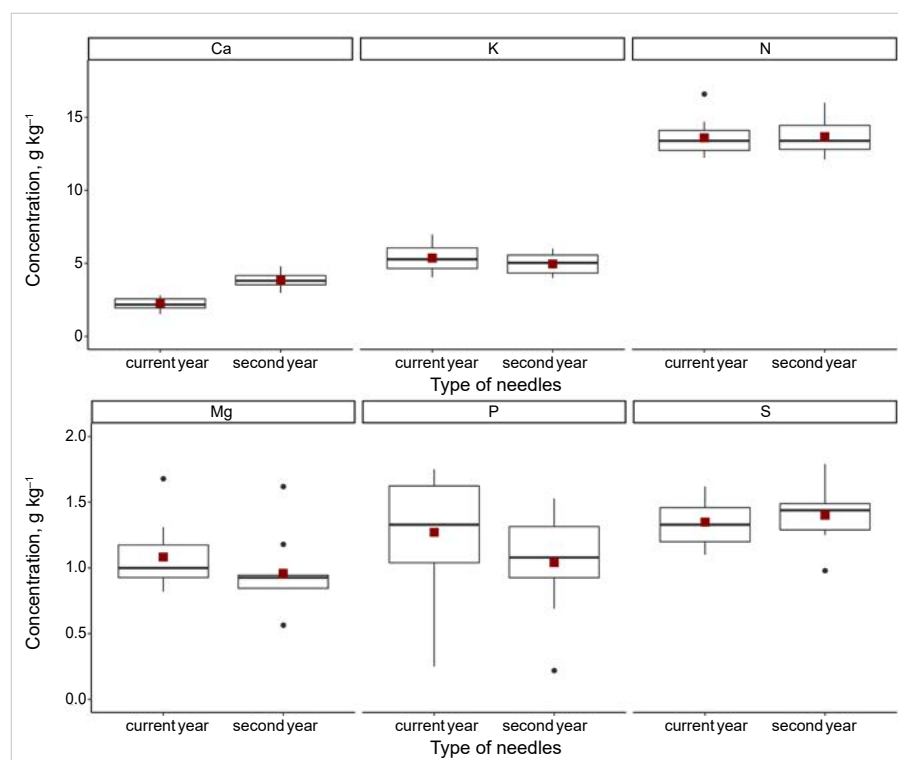


Figure 10. Average concentrations of macroelements in Scots pine needles (current and previous year) at Level II monitoring plots in Latvia. In the boxplots, the median is shown by the bold line, the mean is shown by the dark red square, the box corresponds to the lower and upper quartiles, whiskers show the minimal and maximal values (within 150% of the interquartile range from the median) and black dots represent outliers of the datasets

Discussion

Tree defoliation and occurrence of biotic and abiotic damage are important indicators of forest health and essential for the study of cause-effect mechanisms (De Marco et al. 2014, Eichhorn et al. 2016). Defoliation is easy to evaluate, and severe levels of it may serve as predictors for mortality rates (Dobbertin and Brang 2001). Defoliation affects tree transpiration, and in short periods of drought it can enhance tree water status by reducing canopy transpiration. If the drought is longer, however, the positive effect is temporary, as whole soil-plant-atmosphere continuum is affected (Balducci et al. 2020).

Results of the study performed in Lithuania confirm the importance of combined effects of climate variables, acid deposition and ozone pollution on the defoliation rates of Scots pine (Augustaitis and Bytnerowicz 2008). However, results from the Latvian sample plots show neither elevated values of acid deposition, nor ozone pollution. On the contrary, the concentration of sulphur and N in all deposition fractions has steadily decreased. The risk of environmental pollution to the tree health in Latvia currently is low, but at the same time it needs to be emphasized that further climate change-induced processes may have substantial impact on nutrient cycling in forest ecosystems and plant physiological responses to the stressors.

According to Oliva et al. (2016), pathogen-caused defoliation of Scots pine significantly increases tree susceptibility to secondary pests and reduces resilience to environmental stressors, thus increasing the probability of tree death. Our results demonstrate connection between more pronounced occurrence of tree damage and somewhat higher defoliation rates of Scots pine, even though in our case the link is not particularly clear.

The growth of trees is a key ecological parameter of forests and thus of high importance as an indicator of forest condition (Dobbertin and Neumann 2016). However, according to Seidl et al. (2012), stem increment of Scots pine is only weakly correlated with crown transparency. Our results from the Level II monitoring plot in Valgunde with the longest time series of monitoring demonstrated a weak negative correlation ($r = -0.17$) between annual average growth of Scots pine and defoliation rate as well. Several studies have pointed that the increased atmospheric N deposition (Seftigen et al. 2013, Kosonen et al. 2019) and drought stress shown by the negative correlations between diameter increment and average temperatures (Vacek et al. 2016) are among the main environmental stressors affecting Scots pine growth. Our results indicated that higher air temperature and input of nutrients including N have a positive effect on increment of Scots pine BHD even outside a low N deposition area ($< 2 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), where positive effects of N deposition on tree growth are usually observed (e.g. From et al. 2016, Högberg et al. 2021). However, these relationships should not be generalized, as our results are based on data from only three sites.

Measurements of air pollutant concentrations are necessary to evaluate risks for forest ecosystems, including vegetation, and to document spatial and temporal variability of ambient air quality (Sanders et al. 2016). Gaseous air pollutants such as SO_2 , NO_2 , NH_3 and O_3 may damage forest vegetation by influencing physiological processes such as photosynthesis respiration, stomata regulation and phloem loading, or by leaf or needle damage (Mohren et al. 1992) causing effects ranging from visible injury to reduced carbon sink strength of forest trees (Sanders et al. 2016). Furthermore, SO_2 and NO_2 are known to cause acid deposition with subsequent negative impacts on soil and water quality (Neirynck et al. 2011), but NO_2 and NH_3 contribute to eutrophication (an excess of N).

Critical levels (air quality standards) for protecting vegetation from gaseous air pollutants set by the Ambient Air Quality Directive (EU 2008) are $20 \mu\text{g m}^{-3}$ for SO_2 and $30 \mu\text{g m}^{-3}$ for NO_x ($\text{NO} + \text{NO}_2$) (averaging period – calendar year), but provisional NH_3 critical level for higher plants is $23 \mu\text{g m}^{-3}$ (averaging period – month) (ICP Vegetation 2017). According to Zimny (2006, as quoted by Likus-Ciešlik 2020), the lowest SO_2 level that adversely affects Scots pine is $22 \mu\text{g m}^{-3}$ per day. Both vegetation season mean SO_2 and NO_2 concentrations ($0.39 \pm 0.02 \mu\text{g SO}_2 \text{ m}^{-3}$, $1.78 \pm 0.06 \mu\text{g NO}_2 \text{ m}^{-3}$) in 2009–2019 and individual maximum SO_2 and NO_2 concentrations ($0.94 \mu\text{g SO}_2 \text{ m}^{-3}$, $3.6 \mu\text{g NO}_2 \text{ m}^{-3}$) at the Level II monitoring plot in Valgunde, Latvia, were significantly below critical levels. Also vegetation season mean NH_3 concentration ($3.73 \pm 0.53 \mu\text{g NH}_3 \text{ m}^{-3}$) in 2009–2019 was significantly below critical level, but there were a few peaks when individual maximum NH_3 concentration (up to $30.4 \mu\text{g NH}_3 \text{ m}^{-3}$) exceed critical level at the Level II monitoring plot in Valgunde. In Latvia, the main origin of NH_3 emissions is the agricultural sector, but it was not possible to trace exact source of several identified peaks at the Level II monitoring plot in Valgunde.

Tropospheric O_3 is an important stressor in natural ecosystems – extremely reactive and strong oxidant, which, by reacting with organic compounds, may disrupt various physiological processes (Mohren et al. 1992, Grulke and Heath 2020). Typical symptoms of large doses of O_3 are leaves mottling with decreased photosynthesis and increased dark respiration (Mohren et al. 1992). High levels of O_3 reduce ozone-sensitive forest growth and biodiversity through reducing resistance to environmental stresses, damage plant cells, impairing plants' reproduction and growth (Grulke and Heath 2020). O_3 is thus a priority for the UNECE Convention on Long-range Transboundary Air Pollution (Sanders et al. 2016). Furthermore, Scots pine is ozone-sensitive forest tree species (Augustaitis et al. 2018). The European Community legislative standard is AOT40, i.e. the accumulation of O_3 concentrations exceeding 40 ppb ($80 \mu\text{g m}^{-3}$) over daylight hours from May to July (EU target value: $18,000 \mu\text{g m}^{-3}$

hours) and from April to September (critical level for the protection of forests: $10,000 \mu\text{g m}^{-3}$ hours) (EU Directive 2008/50/EC). Both vegetation season mean O_3 concentration ($42.4 \pm 1.2 \mu\text{g m}^{-3}$) in 2009–2019 and individual peaks of O_3 concentration (up to $104.5 \mu\text{g m}^{-3}$) at the Level II monitoring plot in Valgunde, Latvia, may be characterized as low. Also, visual assessment of trees in the sample plot did not indicate any O_3 damage on tree needles and ground vegetation.

Deposition is one of the key factors in the causal chain between emission of air pollutants and effects in forest ecosystems (Clarke et al. 2016). The adverse effects of increased N deposition include altered soil chemistry and nutrient imbalances followed by soil acidification and base cation losses, disruptions of tree nutrition and productivity decline, lower tree resistance to abiotic and biotic stress factors, changes in the composition of understory vegetation and ectomycorrhizal fungal communities, increased leaching of N from forest soils to surface and ground waters and eutrophication (Porter et al. 2012, De Marco et al. 2014, Kosonen et al. 2019, Schmitz et al. 2019). The thresholds for TN deposition to coniferous forest ecosystems, including *Pinus sylvestris* forests, below which adverse effects are not expected – the ‘critical load’ – is $5\text{--}15 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ (CLRTAP 2017). Measurements show that these thresholds are exceeded at many forest sites in Europe (ICP Forests 2018), including several cases at Level II monitoring plots in Latvia during the observation period (in 2009–2011 in Valgunde and in 2016 in Rucava). But it must be emphasised that for time series with a length of 10 years (2009–2019) decreasing trends of N compounds were observed in Valgunde, especially in the 2009–2014 period, when linear regression slope for TN input through bulk deposition was $-7.3 \text{ kg ha}^{-1} \text{ yr}^{-1}$. During 2015–2019, the TN input through bulk deposition has decreased to $-0.32 \text{ kg ha}^{-1} \text{ yr}^{-1}$, and it agrees well with the findings of earlier studies in Europe (Waldner et al. 2014). Non-significant changes in other parameters of chemical composition of deposition were observed during 2009–2019 in Valgunde. Nevertheless, there are statistically significant differences in the input of macroelements between the Level II monitoring plots in Latvia – the highest input was observed in Rucava. It can likely be explained by the spatial location of the plot (coastal area in the southwest of Latvia) affected by the general trends in Europe (e.g. N deposition gradient from northern Scandinavia to Central Europe) as well as natural causes (e.g. elevated concentrations of elements originating from sea salt).

Soil solution is the matrix linking the solid phase of soil and the plant roots because both macroelements and microelements including nutrients are taken up via the soil solution (Nieminen et al. 2016). Thus, soil solution chemistry is an important indicator for evaluating not only the availability of nutrients and potentially toxic substances to plant roots and mycorrhizas (Iost et al. 2012), but also the effects of air pollution and other stress factors on

forest ecosystems (Nieminen et al. 2016). In Europe, recent research focus was mainly on decreasing acidifying deposition as an explanatory factor for DOC increases in surface waters (Camino-Serrano et al. 2016), as well as on decreasing N deposition impact on soil solution chemistry and soil N stock (Verstraeten et al. 2012, Schmitz et al. 2019). In Latvia, at the Level II monitoring plot in Valgunde, we found significantly increasing trends in soil solution DOC at 20–40 cm (linear regression slope was $+3.02 \text{ mg L}^{-1} \text{ yr}^{-1}$ and $p = 0.04$) and at 40–80 cm depth (linear regression slope was $+1.87 \text{ mg L}^{-1} \text{ yr}^{-1}$ and $p = 0.03$) between 2009 and 2019. For $\text{SO}_4\text{-S}$ concentration in soil solution we found significantly decreasing trend at 40–80 cm depth (linear regression slope was $-0.46 \text{ mg L}^{-1} \text{ yr}^{-1}$ and $p < 0.01$). No indications for increasing or decreasing trends of other soil solution parameters in different depths of the soil profile were found during the period of 2009–2019. Both the mean and individual maximum values of $\text{NO}_3\text{-N}$ concentrations in the soil solution at Level II monitoring plots were significantly lower than the quality threshold value ($11.3 \text{ mg NO}_3\text{-N L}^{-1}$) stated in the national legislation, Water Framework Directive and Nitrate Directive.

In the biogeochemical cycle of forest ecosystems, litterfall (annual return of elements and organic matter to the soil) is a key process linking the tree part to the water and soil part (Ukonmaanaho et al. 2016), furthermore, the importance of forest litter dynamics to site productivity is well known (Kavvadias et al. 2001). To measure, model and predict soil C stocks and their changes due to, for instance, climate change or management activities, it is necessary to have accurate data on litter production (Wutzler and Mund 2007, Cao et al. 2019). In Latvia, during the period of 1997–2006, average amount of foliar litter biomass was $3,621 \pm 462 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in Rucava and $2,869 \pm 388 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in Taurene (Tērauda 2008). Comparing the data of this period (1997–2006) to our time series, during the 2016–2019 the average amount of foliar litter biomass has increased in both plots to $5,136 \pm 535 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in Rucava and $3,801 \pm 620 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in Taurene. In Valgunde, the average amount of foliar litter biomass during 2009–2019 was $3,260 \pm 172 \text{ kg ha}^{-1} \text{ yr}^{-1}$ and no increasing trend was observed. Comparing the different Level II monitoring plots in Latvia, the largest average amount of foliar litter total biomass was observed in Rucava, but statistically significant differences in input of total C through foliar litter biomass between the different Level II monitoring plots were not observed (average total C input was $2,025 \pm 150 \text{ kg ha}^{-1} \text{ yr}^{-1}$). Statistically significant differences in amount of foliar litter biomass between the different Level II monitoring plots may be interpreted by a combination of environmental factors, such as lower latitude (Vucetich et al. 2000), proximity to the Baltic Sea and coastal climate conditions, higher input of macronutrients, as well as slightly higher average defoliation rate that may

be caused by bark beetle outbreak nearby. The assumption that forest litter is a principal pathway for the return of nutrients to the soil (Kavvadias et al. 2001) was confirmed by the results of this study as well: the input of macroelements through litter biomass significantly ($p < 0.05$) exceeded the input through bulk deposition.

Sampling and analysis of needles and leaves is essential to characterize current state of environmental pollution and subsequent tree response (e.g. Yilmaz and Zengin 2004, Pietrzykowski et al. 2014, Likus-Cieślak et al. 2020), nutritional status of trees (Rautio et al. 2016) and general relationships between foliar chemistry and chemistry of soil and soil solution in forest ecosystems (e.g. Jandl and Herzberger 2001, Aitkenhead-Peterson et al. 2006, Merilä and Derome 2008). Results of this study highlighted trends of increasing Ca accumulation with needle age, but the needle mobile macroelement (K, Mg, P) concentrations decreased significantly with increasing needle age. It is in line with previous findings about *Pinus sylvestris* L. needles as bioindicators in the environment (e.g. Kochian 1991, Kurczyńska et al. 1997, Rautio et al. 1998, Lamppu and Huttunen 2003, Rautio 2003). We compared average macroelement concentrations in needles at the Level II monitoring plots in Rucava and Taurene in 2001–2006 (Tērauda 2008) with average concentrations in 2009–2019. In general, significant differences were not detected, only slight decreases in N and K concentrations both in current year and previous year needles were observed.

Conclusions

In Latvia, during the period of 2009–2019 the health of Scots pine forests characterized by defoliation may be considered as rather stable and favourable. The mean defoliation and damage intensity somewhat vary between years, reflecting temporal changes in environmental conditions and activity of damaging agents. Most of the damages to the trees are caused by direct human influence, mainly taking the form of mechanical damage during forest management operations. Forest health indicators reflect also regional insect outbreaks.

Although during the last decade climate change in Latvia has manifested itself as increase in average air temperature and longer periods of drought, element flows in Scots pine forests at Level II monitoring plots have been relatively stable except for the decreasing trend in total N concentration in deposition and $\text{SO}_4\text{-S}$ concentration in soil solution and increasing trends in DOC concentration in soil solution that is in line with common trends in Europe.

To evaluate, model and predict C turnover and SOC stock and its changes in forest ecosystems, information on C input through foliar litter is crucial. Although inter-annual variation of foliar litter biomass was relatively wide, C input with above-ground litter was rather stable during the whole period.

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Supplements

Table S1. Characteristics of Scots pine forests in Level I monitoring plots in Latvia (2019 year data)

Parameter	Description
Dominant tree specie	Scots pine (<i>Pinus sylvestris</i> L.)
Distribution of forest site types	<i>Hylocomiosa</i> – 40%; <i>Myrtillosa</i> – 18%; <i>Vacciniosa</i> – 16%; <i>Sphagnosa</i> – 7%; <i>Myrtillosa mel.</i> – 5%; <i>Myrtillosa turf. mel.</i> – 5%
Age variation	47–152 years
Average age	89 years
Site index variation	I ^a –V ^b
Shape of monitoring plot	circular

Table S2. Characteristics of Scots pine forests in Level II monitoring plots in Latvia

Parameter	Level II monitoring plot		
	Valgunde	Taurene	Rucava
Dominant tree specie	Scots pine (<i>Pinus sylvestris</i> L.)	Scots pine (<i>Pinus sylvestris</i> L.)	Scots pine (<i>Pinus sylvestris</i> L.)
Admixture	<i>Picea abies</i> (L.) H. Karst. in the second floor	<i>Picea abies</i> (L.) H. Karst. in the second floor	<i>Picea abies</i> (L.) H. Karst. in the second floor
Forest site types	<i>Myrtillosa</i>	<i>Myrtillosa</i>	<i>Myrtillosa</i>
Dominant species in forest floor	Moss layer – <i>Hylocomium splendens</i> , <i>Pleurozium schreberi</i> Herb layer – <i>Vaccinium myrtillus</i>	Moss layer – <i>Hylocomium splendens</i> , <i>Pleurozium schreberi</i> Herb layer – <i>Vaccinium myrtillus</i>	Moss layer – <i>Hylocomium splendens</i> , <i>Pleurozium schreberi</i> Herb layer – <i>Vaccinium myrtillus</i>
Site index	I	I	I
Age in 2019 (years)	92	79–92	72
Number of dominant tree species (Scots pine) per ha	863	821	764
Year of establishment of ICP Forests monitoring plot	2004	2015	2015
Shape of monitoring plot	One continuous rectangle	Two individual circle	Three individual circle
Total area of monitoring plot	2400.0 m ²	1413.0 m ²	2119.5 m ²
Soil type (WRB 2014)	<i>Folic Arenosol (dystric)</i> , <i>Ortsteinic Albic Folic Podzol (dystric)</i>	<i>Albic Podzols (Novic)</i>	<i>Haplic Podzols (Novic)</i>

Table S3. Meteorological conditions in 2009–2019 in Latvia

Parameter	Site	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
Mean precipitation amount, mm	Valgunde *	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	731	491	602
	Taurene **	n.a.	n.a.	n.a.	n.a.	522	756	617	854	897	n.a.	869
	Rucava ***	549	946	789	857	757	742	723	455	998	n.a.	692
	Average in Latvia	753	856	690	832	622	725	606	740	810	473	629
Mean air temperature, °C	Valgunde *	n.a.	n.a.	n.a.	n.a.	+7.0	+7.5	+7.8	+7.4	+7.2	+8.0	+8.5
	Taurene **	n.a.	n.a.	n.a.	n.a.	+5.9	+6.4	+6.7	+5.9	+5.7	+6.9	+7.0
	Rucava ***	+7.3	+6.0	+7.9	+7.0	+7.7	+8.3	+8.4	+7.7	+7.8	+8.1	+9.2
	Average in Latvia	+6.5	+5.6	+7.3	+6.1	+7.0	+7.4	+7.8	+7.1	+6.9	+7.6	+8.2

Note: * – data from meteorological observation station in Jelgava; ** – data from meteorological observation station in Zosēni; *** – data from meteorological observation station in Rucava; n.a. – data not available. Data source: State limited Liability Company “Latvian Environment, Geology and Meteorology Centre”.

Table S4. Frequency of assessment, measurement and sampling in Level I and Level II plots in 2009–2019 included in the study

Type of observations	Level I	Level II		
		Valgunde	Taurene	Rucava
Defoliation	2009–2019, continuously	2009–2019, continuously	2015–2019, continuously	Since 2019
Damaging agents	2011–2019, continuously	2009–2019, continuously	2015–2019, continuously	Since 2019
Tree growth	-	2009–2019, continuously	2016–2019, continuously	2016–2019, continuously
Air quality	-	2009–2019, continuously during vegetation season	-	-
Deposition	-	2009–2019, continuously	2016–2019, continuously	2016–2019, continuously
Soil solution	-	2009–2019, continuously	2016–2019, continuously	2016–2019, continuously
Litterfall	-	2009–2019, continuously	2016–2019, continuously	2016–2019, continuously
Needles	-	2009–2019, every second years	2015–2019, every second years	2017–2019, every second years