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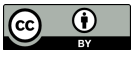
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Assessment of Atmospheric Deposition and Vitality Indicators in Mediterranean Forest Ecosystems

Tamara Jakovljević [1], Aldo Marchetto [2], Lucija Lovreškov [1], Nenad Potočić [1], Ivan Seletković [1], Krunoslav Indir [1], Goran Jelić [3], Lukrecija Butorac [3,*], Željko Zgrablić [1], Alessandra De Marco [4,*], Guillaume Simioni [5], Mladen Ognjenović [1] and Ana Jurinjak Tušek [6]

1 Croatian Forest Research Institute, Cvjetno naselje 41, 10450 Jastrebarsko, Croatia; tamaraj@sumins.hr (T.J.); lucijal@sumins.hr (L.L.); nenadp@sumins.hr (N.P.); ivans@sumins.hr (I.S.); kindir@sumins.hr (K.I.); zeljkoz@sumins.hr (Ž.Z.); mladeno@sumins.hr (M.O.)
2 Water Research Institute, National Research Council (CNR-IRSA), Viale Tonolli 50, 28922 Verbania, Italy; aldo.marchetto@cnr.it
3 Institute for Adriatic Crops and Karst Reclamation, Put Duilova 11, 21000 Split, Croatia; goran.jelic@krs.hr
4 Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA), CR Casaccia, Viale Anguillarese 301, 00123 Roma, Italy
5 INRA, Ecology of Mediterranean Forests Unit (URFM) Domaine Saint Paul, Site Agroparc, 84914 Avignon CEDEX 9, France; guillaume.simioni@inra.fr
6 Faculty of Food Technology and Biotechnology, University of Zagreb, Pierottijeva 6, 10000 Zagreb, Croatia; atusek@pbf.hr
* Correspondence: lukrecija.butorac@krs.hr (L.B.); alessandra.demarco@enea.it (A.D.M.); Tel.: +385-21434432 (L.B.)

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Abstract: Considering the fragility of the Mediterranean environment, there is an increasing need to improve the knowledge of this forest environment. The aim of this study was to examine the effects of air pollution on the forest ecosystem's condition by analyzing tree vitality. The study area was chosen to represent the most important and the most common species in Mediterranean forest ecosystems of the Eastern Adriatic coast. *Quercus pubescens*, *Quercus ilex*, *Pinus halepensis*, and *Pinus nigra* plots were equipped with rain collectors and dendrometer bands. Sampling, measurements, and analyses of atmospheric deposition, foliar nutrient, defoliation, and growth were all carried out. Results showed that actual N deposition loads were the lowest in Aleppo pine forest and the highest in holm oak forests. This, however, did not have an effect on the concentrations of N in foliage. Most elements' concentrations were in the plausible range. No relevant differences in mean defoliation between the plots were observed. The plots with a lower percentage of basal area increment (BAI%) were found to have lower defoliation. The research was conducted to bridge the gap in the knowledge of air pollutants and vitality indicators in different forest types. These findings are a valuable contribution to the sustainable forest management of Mediterranean forest.

Keywords: atmospheric inputs; crown; defoliation; foliage; growth; Mediterranean forest types

1. Introduction

Mediterranean forests provide a wide array of environmental services and products. Despite their important role as primary green infrastructure of the region, Mediterranean forests are subject to numerous threats, such as forest fires, intense droughts, over-exploitation, deforestation, and degradation. Among all bioclimatic regions, the Mediterranean region appears to be the most

vulnerable. The vulnerability to climate change is related to the trend of increasing temperature, degradation of water resources and to the increased water demand and impacts of air pollution [1].

According to the National Emission Ceilings Directive reporting status 2018 [2], to achieve adequate air quality levels and avoid significant negative impacts on ecosystems in Europe, the revised National Emission Ceilings Directive [2] sets emission reduction commitments for each Member State to be reached by 2030. Four Member States exceeded ceilings in 2016 for one of four important air pollutants—nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), sulphur dioxide (SO_2), and ammonia (NH_3). Two of them were Mediterranean countries, Croatia and Spain. They exceeded ceilings for NH_3 (Spain 39% and Croatia 17%). The same countries persistently exceeded their ceiling for this pollutant over the period 2013–2016 [2].

However, the potential ecological effects of N deposition in Mediterranean-type climates have been less investigated [3–8], even though they are usually recognized as hot-spots of biodiversity [3]. That is why it is important to understand how systems that are seasonally water limited, where dry deposition is cumulative, and where fires occur naturally and are frequent, will respond to enhanced reactive N deposition. Consequently, more research is needed for each region and vegetation type [9].

Advancing scientific knowledge and fostering innovation is also essential for sustainable forest management and to guide policymakers in the Mediterranean region. Countries of the Mediterranean basin, as well as other regions with a Mediterranean climate, face similar challenges regarding forest ecosystems and the provision of crucial goods and services in the context of climate change [10].

Mediterranean forest ecosystems along the Eastern Adriatic coast, which mostly belong to Croatia and cover 24% of its total forest area, are probably the most endangered forest ecosystems [11]. They provide multiple goods and services, such as water-related services, soil protection, and an exceptional richness in terms of biodiversity and unique non-wood forest products, which is highly significant from an ecological and economical point of view. This region is ecologically very sensitive, because of its geographic position, complex orography, specific meteorological conditions, and number of pollution sources [12]. The impacts of air pollution, erosion, landslides, and floods are very high in this region. These risk factors combined with climate change are likely to affect nutrient turnover and nutrient availability, soil moisture, and, ultimately, growth and primary productivity. Atmospheric deposition and its transformation in contact with vegetation are of great importance in understanding its effects on forests. It has an impact on forest ecosystems through eutrophication by nitrogen and soil acidification, thus altering soil properties and processes [13]. Soil acidification by sulphur and nitrogen deposition could cause a loss of base cations (i.e., calcium, magnesium, and potassium) from the soil, inhibit base cation uptake, and decrease pH that may increase the mobility of heavy metals. In this stage, a decrease in forest condition and growth is assumed to occur [14]. Changes in the soil chemistry may lead to imbalances in nutrient supply and subsequently to unbalanced nutrition of the trees. Nutrient imbalance will affect canopy photosynthesis and in turn decrease forest vitality. The chemical composition of the foliage of forest trees is an important indicator for tree nutrition, providing information on deficiency or an excess of nutrients [14,15].

Mediterranean forest ecosystem damage due to air pollution was determined only on the basis of visual induces based on foliage color, degree of leaf damage (necrotic spots, fraction of area removed by herbivores), and the degree of crown defoliation. Field-based visual observations of defoliation showed that the situation is alarming not only because of the percentage of severely damaged trees but also the trend of increases in the percentage of damage, as for all types of total and individual types (except holm oak) [16].

Considering the described peculiarity and fragility of the Mediterranean environment, there is an increasing need to improve the knowledge of this forest environment. Consequently, the aim of this study was to examine the effects of air pollution and climate change on forest ecosystems' condition, mainly by the observation of tree vitality. However, tree vitality cannot be measured directly and indicators, such as tree growth or crown transparency, defoliation, and foliar nutrient, may instead be used [17]. Furthermore, forest monitoring and data collection activities will directly contribute

to sustainable forest management and enhance the protection of EU forests because atmospheric pollution has a sizeable influence on the ecological condition and productive capacity of forests. As the protection of forests against biotic and abiotic agents is one of the main priorities of forest policy, it is essential to have up-to-date information about the state of forests in the EU [18]. Collected data could be used to assess the sustainable forest management on the basis of Criterion 2: Maintenance of Forest Health and Vitality, according to Pan-European Criteria and Indicators for Sustainable Forest Management [19]. For the purpose of this study, two regions along the Adriatic coast were considered, Istria and Dalmatia, with different vegetation-type coverage. The eu-Mediterranean and sub-Mediterranean area of Dalmatia is mostly covered with Aleppo pine and black pine forests while broadleaf forests of holm oak and pubescent oak cover smaller areas and are present in different stages of degradation. In Istria, broadleaf forests of holm and pubescent oak cover the main area of the region, dominating over Aleppo and black pine forests.

Atmospheric deposition was measured together with vitality indicators, including those related to foliar nutrient, growth, and defoliation, at selected plots in Mediterranean forest ecosystems.

The main objectives of the study were:

(1) To estimate the atmospheric inputs and identify differences in N deposition between the plots;
(2) To estimate actual N deposition loads and compare them to critical loads; and
(3) To identify the status of nutrients in foliage to evaluate tree growth and defoliation.

The novelty of this study was to bridge the gap in the knowledge of typical Mediterranean forest ecosystems and to contribute to a wider overview of the impacts of air pollution in the Mediterranean forest ecosystem of the Eastern Adriatic coast. Furthermore, the provision of relevant information from the monitoring of these indicators for policy makers in forestry contributes to sustainable forest management.

2. Materials and Methods

2.1. Locations and Experimental Site

Four experimental sites (plots) were chosen, which were square with an area of 0.25 ha. (Figure 1). Two plots are located in Istria. The Šišan plot (44°51′41.2″ N; 13°59′24.4″ E, 3 m a. s. l.) is located in southern Istria. The most common species is *Quercus ilex* L. (holm oak). The Poreč plot (45°14′59.2″ N; 13°43′52.9″ E, 264 m a. s. l.) is located in Western Istria. Vegetation in this plot is dominated by *Quercus pubescens* Wild. (pubescent oak). The terra rossa soil type is present on plots located in Istria [20]. Two other plots are in Dalmatia. The Vrana plot (43°53′23″ N; 15°33′47″ E, 20 m a. s. l.) is located in Northern Dalmatia. The main tree species in this plot is *Pinus halepensis* Mill. (Aleppo pine). The Split plot (43°41′59″ N; 16°26′34″ E, 550 m a. s. l.) is located in Middle Dalmatia. The dominating species is *Pinus nigra* L. (black pine). Plots in Dalmatia have a calcocambisol soil type, which is the dominant soil in the Croatian Mediterranean area [20].

According to the Köppen climate classification, the plots are distributed in the hot-summer Mediterranean climate subtype (Csa) [21]. Frosty days ($t_{min} < 0°C$) are limited to winter months and range from 12 (Split plot) to 4 days (Poreč plot). Average annual precipitation ranges from 879 (Vrana plot) to 1277 mm (Split plot), while the mean air temperature varies from 12.4 (Split plot) to 13.4 °C (Poreč plot) [22].

2.2. Sampling and Analysis

The four plots were fully equipped for the measurement of atmospheric deposition and tree growth. Sampling, measurements, and analyses on the plots were all carried out according to the ICP Forests (International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests) manuals. The study period was from April 2017 to December 2018. Annual deposition was calculated from January to December 2018.

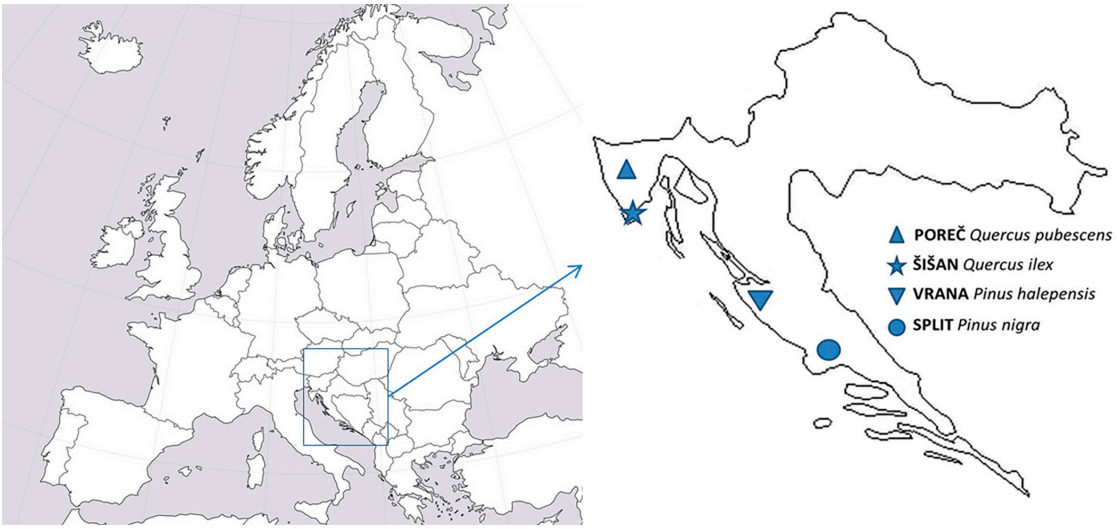


Figure 1. Location of four experimental sites in the Eastern Adriatic coast and dominant forest type at each plot.

Deposition was sampled using continuously exposed, randomly placed collectors, comprising a 2-L graduated polyethylene bottle with a funnel with a 14-cm diameter. Nine collectors were placed beneath the forest canopy to collect throughfall deposition (THR) samples. Bulk open field deposition (BOF) was sampled using three collectors, continuously exposed. Samples were collected bi-weekly for the whole period from January to December 2018. The analyses of ion concentrations and alkalinity were performed on filtered samples (0.45 µm), except for the measurements of pH and conductivity [23]. Ion chromatography was used to determine the concentrations of ions, i.e., chloride, nitrate, sulphate, phosphate, ammonium, sodium, potassium, calcium, and magnesium [23]. Validation of the analytical results was performed [24]. Actual N deposition loads were calculated for THR deposition [25].

In each plot, foliar samples of 10 dominant trees, assumed to be representative of the mean health status, were collected during the second half of the growing season [26]. The concentrations of nutrients were determined as follows: N by an elemental analyzer [27]; and Ca, P, Mg, and K by an atomic absorption spectrophotometer [26].

Stand dendrometric parameters (tree species, mean diameter and height of trees, top height, tree density, total basal area, tree volume per hectare, stand age, mortality) were measured and determined on all trees within plots. The cut-off tree diemeter DBH was 5 cm. Measurements of actual basal area increment BAI were performed bi-weekly during the vegetation period, on 40 trees per plot equipped with dendrometer bands. Those trees were spread across the 0.25-ha plots, and their DBH distribution was similar to the stand DBH distribution. To determine the height of the treetop, a hypsometer was used [28]. Crown condition variables—defoliation and discoloration for all sample trees (45 trees per plot)—were assessed annually [16]. Mean annual values of crown defoliation as well as stem diameter were computed for every plot. The frequency of trees with crown defoliation (F > 25) and the frequency of trees with observed foliage discoloration (F_damage) were used as additional indicators of tree vitality on the plots [29].

2.3. Statistical Analysis

All experimental results were statistically analyzed using the program Statsoft Statistica 13.0. Data in the text, tables, and figures are expressed as the mean ± standard deviation (± SD). The annual mean concentrations of measured ions were calculated as volume-weighted means. The Wilcoxon signed test was used to test whether BOF and THR depositions of ions were significantly different among collector types. A significant difference was considered at the level of $p < 0.05$. Data normality was tested using the Kolmogorof–Smirnov test implemented in the Statistica 13.0. software, where the

data was considered normally distributed if the D value was insignificant at $p < 0.05$. Based on normally distributed data, Pearson correlation was used to compare BOF and THR depositions of the main chemical species at the selected locations.

The cumulative diameter increment, average annual values of the basal area increment (BAI), and the average percentage of basal area increment (BAI%) were calculated across all trees with dendrometer bands for each plot:

$$BAI = BA_{Y_END} - BA_{Y_BEGIN} \quad (1)$$

$$BAI\% = (BAI/BA_{Y_BEGIN}) \cdot 100 \quad (2)$$

Deposition of nitrogen was calculated from deposition fluxes as follows [30]:

$$DEP_N = N\text{-}NH4^{+} + N\text{-}NO_3^{-} \quad (3)$$

The deposition of nitrogen was than compared with the critical load for nitrogen taken from the literature [31]:

$$DEP_N \leq CL\ (N) \quad (4)$$

R statistical environment [32] was used to produce plots displaying the results of the foliar analysis.

3. Results

3.1. Deposition

The annual amount of precipitation in 2018 in the open field collectors ranged between 900 (Aleppo pine plot) and 1417 mm (black pine plot) (Table 1). In general, the literature reports a higher precipitation volume collected by bulk samplers than by wet-only samplers, with the collection efficiency of wet compared to bulk ranging between 71% and 99% [4]. The location of the sampling plots clearly influenced the content of the major ions. By comparing the mean annual BOF and THR ion concentrations at the analyzed plots, it can be noticed that the highest values for Mg^{+}, Na^{+}, K^{+}, $N\text{-}NH_4^{+}$, SO_4^{2-}, Cl^{-}, and total alkalinity were measured for THR deposition in the holm oak forest. The throughfall rainfall volumes at all four sites were significantly lower than the volumes collected in the open field by the same sampler (bulk). The highest value was at the black pine plot (1222 mm) and the lowest at the Aleppo pine plot (749 mm).

Table 1. Amount of precipitation (V) and mean annual bulk (BOF) and throughfall (THR) pH, conductivity, and concentrations of measured ions and alkalinity at each plot. Results are presented as mean ± st.dev.

Forest Type		V	pH	Cond.	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	$N\text{-}NH_4^{+}$	$S\text{-}SO_4^{2-}$	$N\text{-}NO_3^{-}$	Cl^{-}	Total Alkalinity
		mm		μScm^{-1}				$\mu eq\ m^{-2}$					$\mu eq\ L^{-1}$
Black pine	BOF	1417	5.78 ± 0.58	13.40 ± 5.27	16,446.32 ± 10,416.89	2449.04 ± 1618.25	1534.73 ± 1751.95	1222.05 ± 1406.84	6515.08 ± 9274.82	4769.93 ± 3310.24	1732.60 ± 2428.61	3881.62 ± 4757.61	0.03 ± 0.02
	THR	1222	5.83 ± 0.33	26.19 ± 18.31	18,163.97 ± 11,608.10	2735.88 ± 2186.17	2229.31 ± 2510.78	1983.38 ± 1849.48	2415.61 ± 5428.74	5185.37 ± 4294.81	1596.40 ± 2973.45	2259.73 ± 2111.27	0.05 ± 0.04
Aleppo pine	BOF	900	6.01 ± 0.47	15.31 ± 7.81	1519.24 ± 7308.08	2769.67 ± 1356.26	5291.51 ± 11107.20	2046.88 ± 1323.77	5355.71 ± 1246.29	5383.76 ± 5173.81	1710.65 ± 2218.92	8793.18 ± 1761.47	0.02 ± 0.01
	THR	749	5.86 ± 0.37	48.30 ± 29.07	11,887.30 ± 8025.95	3030.25 ± 1976.73	3837.16 ± 3484.15	2280.89 ± 1582.49	3199.788 ± 5550.11	5223.28 ± 3830.76	1392.11 ± 1029.45	5294.39 ± 4978.78	0.03 ± 0.01
Pubescent oak	BOF	1068	5.84 ± 0.53	21.90 ± 17.99	12,925.22 ± 25,598.87	2376.20 ± 4629.77	2669.42 ± 5252.71	2613.54 ± 5036.87	1666.15 ± 3239.78	4634.22 ± 9132.84	1178.68 ± 2344.53	3092.10 ± 6078.87	0.03 ± 0.06
	THR	999	6.00 ± 0.29	36.99 ± 22.28	11,674.10 ± 7812.99	2200.24 ± 1808.33	2888.62 ± 2670.54	2766.59 ± 2752.75	2014.23 ± 2793.92	4376.92 ± 3613.12	1439.99 ± 1098. 86	3266.12 ± 3401.15	0.05 ± 0.03
Holm oak	BOF	1073	5.92 ± 0.49	42.07 ± 23.45	10,864.84 ± 7084.86	6172.85 ± 5831.87	16,170.32 ± 22,222.82	2458.92 ± 21,112.703	10,117.74 ± 29,134.63	8093.55 ± 6199.94	1156.96 ± 1040.91	27,412.98 ± 32,260.71	0.03 ± 0.02
	THR	968	5.94 ± 0.29	62.18 ± 33.75	12,332.84 ± 7011.47	7659.34 ± 6101.29	19,545.58 ± 21,461.21	5046.59 ± 4749.18	12,332.84 ± 7011.47	11,316.76 ± 8769.40	1553.41 ± 1213.60	34,156.20 ± 38,285.65	0.06 ± 0.05

According to the Wilcoxon test applied to the bi-weekly data (Table 2), significant differences between BOF and THR depositions of ions can be observed at the black pine plot, while there were no significant differences in the Aleppo pine plot. In the black pine plot, statistical analysis revealed a significant difference in depositions between BOF and THR for Na^+ ($p = 0.037$), K^+ ($p = 0.039$), N-NH_4^+ ($p < 0.001$), and total alkalinity ($p = 0.006$).

Table 2. Significance level of the differences between bulk and throughfall fluxes according to the Wilcoxon signed test (marked as bold as significant for $p < 0.05$).

Forest Type	Ca^{2+}	Mg^{2+}	Na^+	K^+	N-NH_4^+	S-SO_4^{2-}	N-NO_3^-	Cl^-	Total Alkalinity
Black pine	0.423	0.407	**0.037**	**0.039**	**<0.001**	0.086	0.253	0.333	**0.006**
Aleppo pine	0.272	0.683	0.729	0.594	0.470	0.826	0.778	0.875	0.115
Pubescent oak	0.300	0.759	0.911	0.509	**0.011**	0.427	0.191	0.840	**0.016**
Holm oak	0.405	0.238	0.294	**0.023**	0.308	0.106	0.208	0.289	**<0.001**

Pearson correlation was used to compare BOF and THR depositions of the main chemical species at the analyzed plots (Table 3). In the case of the black pine plot, there were strong positive relationships for SO_4^{2-} and NO_3^-. Also, there was a strong positive relationship for K^+, NH_4^+, and SO_4^{2-} in the pubescent oak plot. On the other hand, there were no strong correlations between BOF and THR depositions of the main chemical species in the Aleppo pine and holm oak plots.

Table 3. Pearson correlation coefficients between bi-weekly bulk and throughfall fluxes at all analyzed plots (marked as bold as significant for $p < 0.05$).

Forest Type	Ca^{2+}	Mg^{2+}	Na^+	K^+	N-NH_4^+	S-SO_4^{2-}	N-NO_3^-	Cl^-
Black pine	**0.497**	**0.429**	**0.474**	0.316	0.236	**0.609**	**0.865**	0.164
Aleppo pine	−0.174	−0.155	0.127	0.094	0.258	−0.093	0.075	0.139
Pubescent oak	0.276	**0.459**	0.291	**0.541**	**0.639**	**0.685**	0.227	0.361
Holm oak	−0.170	−0.136	−0.116	−0.123	0.217	−0.125	−0.104	0.117

Actual throughfall N deposition loads ranged between 3.41 kg N ha^{-1} y^{-1} in the Aleppo pine plot to 19.70 kg N ha^{-1} y^{-1} in the holm oak plot (Table 4).

Table 4. Actual N deposition loads and empirical N critical loads.

Forest Type	DEP_N	DEP_N Critical
	kg ha^{-1} y^{-1}	
Black pine	8.19	15
Aleppo pine	3.41	5–15
Pubescent oak	17.64	10–20
Holm oak	19.7	10–20

3.2. Foliar Nutrition

To evaluate the foliar nutrient concentrations, we compared them to the nutrient concentration classes derived from the plausible range of element concentrations in foliage [33]. Foliar nutrient concentrations are expressed in mg g^{-1} of dry weight (Figure 2). For most elements and species, concentrations were in the plausible range. Low values of nitrogen were recorded for holm oak and black pine. High values of Ca were found for pubescent oak, black pine, and Aleppo pine. Only in holm oak was the concentration of K in the low range.

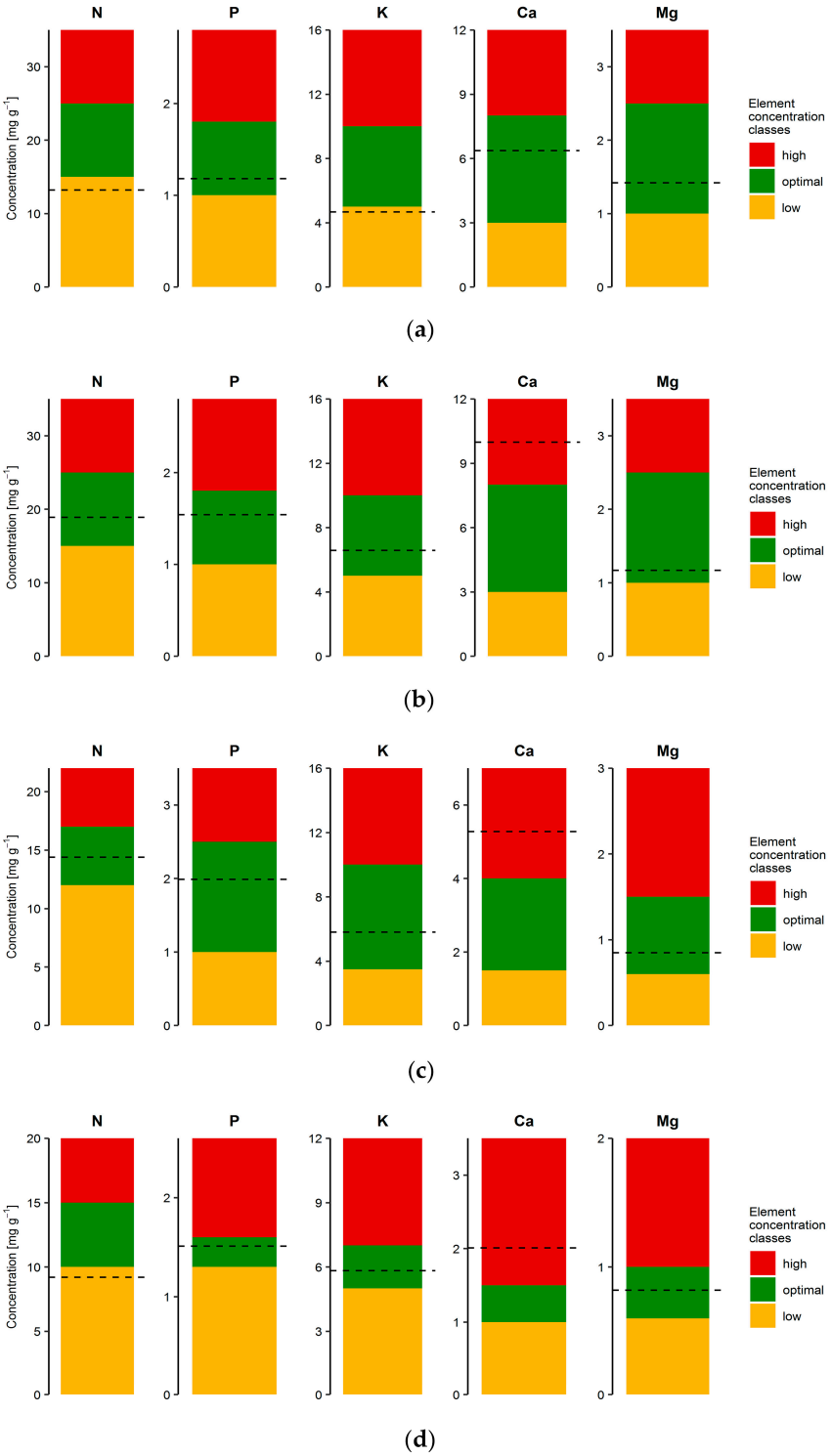


Figure 2. Concentrations of mineral elements in leaves and needles (dashed lines): (**a**) holm oak leaves; (**b**) pubescent oak leaves; (**c**) Aleppo pine needles; and (**d**) black pine needles.

3.3. Defoliation

The results of defoliation monitoring are given in Figure 3 for 2017 and 2018, according to the defoliation class and plot location. There are no pronounced differences in the mean defoliation between plots; nevertheless, we observed differences in the percentage of trees with defoliation higher than 25%. The highest percentage of significantly defoliated trees (share of trees with defoliation higher than 25%) was established in the Aleppo pine plot, (66.67% in the year 2017 and 60.0% in 2018). A high

percentage of significantly defoliated trees was also observed in the pubescent oak plot (48.89% in 2017, 33.33% in 2018) and black pine plot (33.33% in 2017, 43.6% in 2018). Holm oak forest was the most vital plot in our study, with 2.78% and 8.3% significantly defoliated trees in 2017 and 2018, respectively.

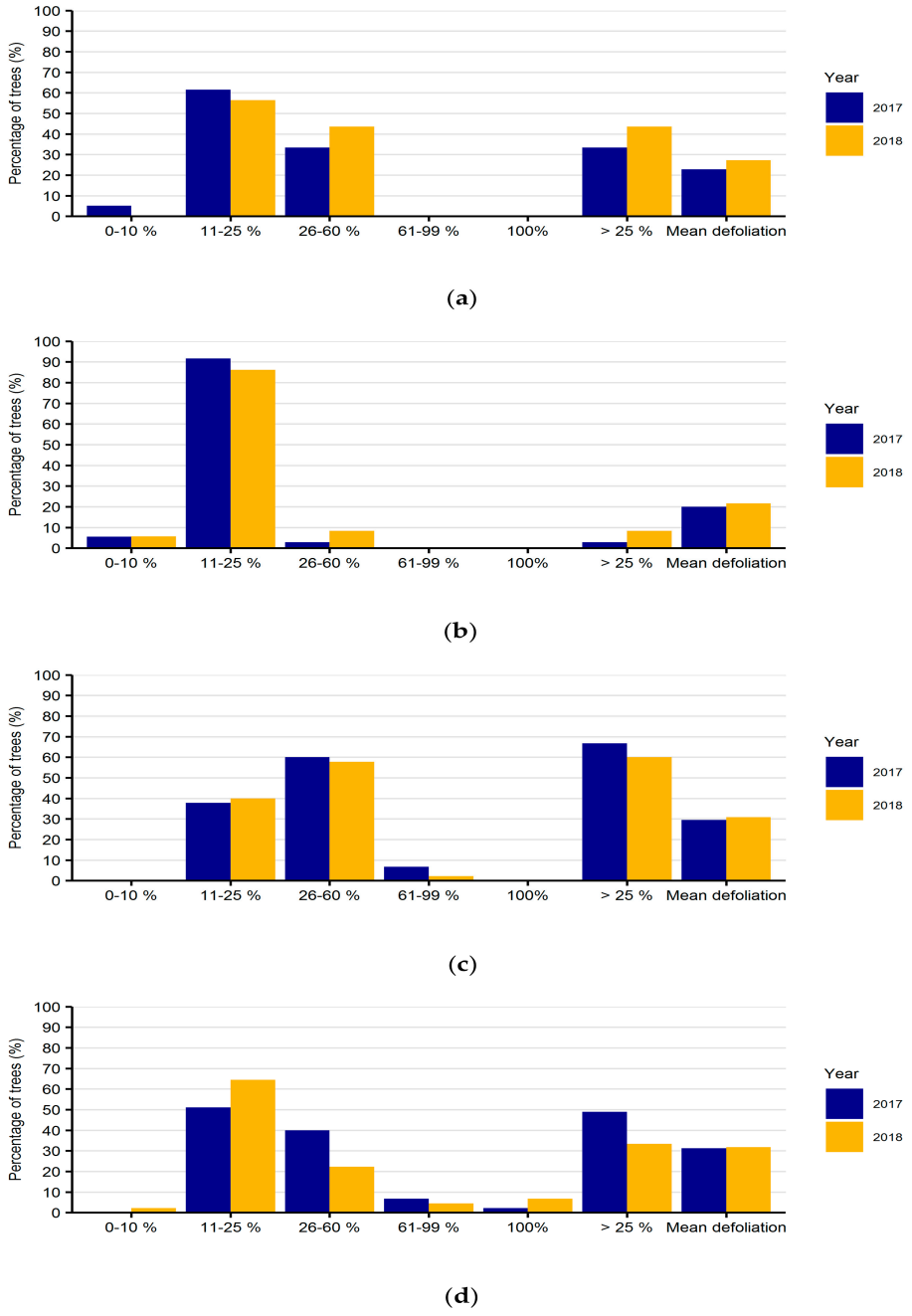
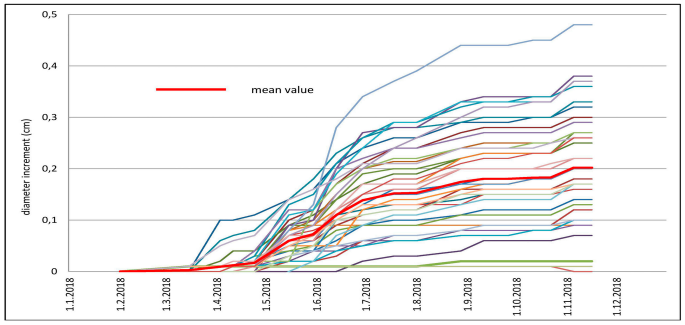


Figure 3. Defoliation of trees, years 2017 and 2018: (**a**) black pine plot; (**b**) holm oak plot; (**c**) Aleppo pine plot; and (**d**) pubescent oak plot.

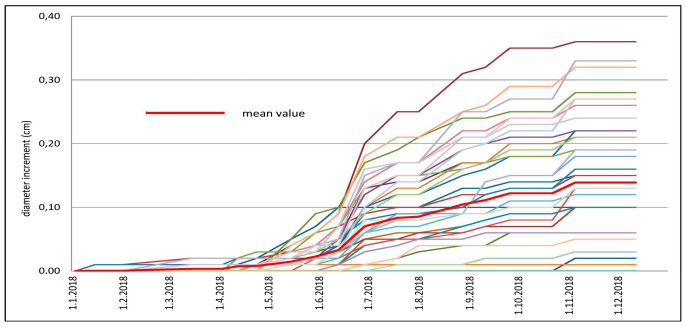
3.4. Growth

The result of the growth track on dendrometer bands is the intra-annual dynamic of tree growth (Figure 4). The cumulative diameter increment shows the seasonality of growth. The bold red line in the figures represents an average value of the cumulative diameter increment in a particular year, ranging from 0.13 cm year^{-1} for black pine, 0.26 cm year^{-1} for Aleppo pine, and 0.13 cm year^{-1}

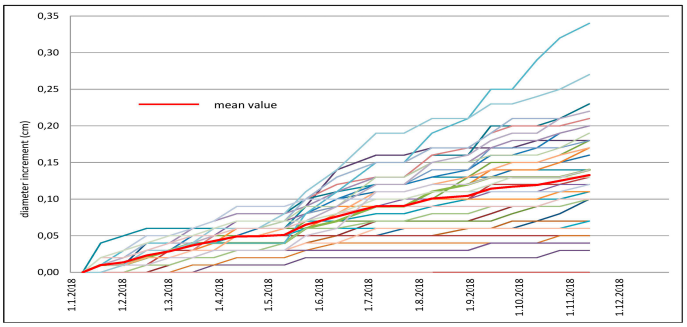
holm oak, to 0.20 for pubescent oak. As expected, higher values of increment can be observed in the spring months.



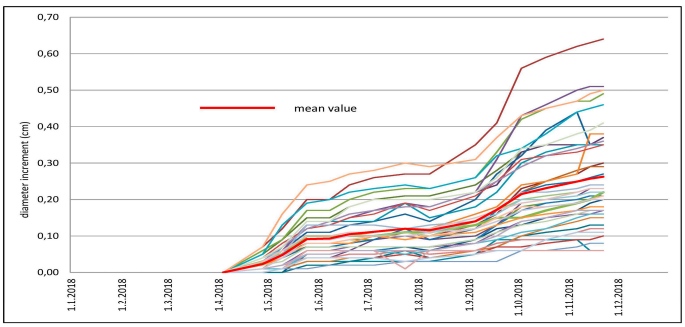
(**a**)



(**b**)



(**c**)



(**d**)

Figure 4. Cumulative diameter increments in 2018: (**a**) pubescent oak trees; (**b**) holm oak trees; (**c**) black pine trees; and (**d**) Aleppo pine trees.

Average annual values of the basal area increment (BAI) and the average percentage of basal area increment (BAI%) were calculated for all trees with dendrometer bands (40 trees per plot). The

percentage of basal area increment ranged from 0.621 to 2.582 in 2017 and from 0.717 to 2.544 in 2018. In all four plots, the BAI% was higher in 2018 than 2017 (Table 5).

Table 5. Basal area increment (BAI), percentage of basal area increment (BAI%), and mean defoliation on all plots.

Forest Type	Tree Density	2017			2018		
		BAI	BAI %	Mean Defoliation	BAI	BAI %	Mean Defoliation
	n/plot	$mm^2\ y^{-1}$	%	%	$mm^2\ y^{-1}$	%	%
Black pine	86	820	0.671	22.78	861	0.731	27.17
Aleppo pine	169	867	2.582	29.52	897	2.544	30.78
Pubescent oak	365	523	2.173	31.27	607	2.420	31.78
Holm oak	354	247	0.621	20.00	562	1.589	21.60

BAI, BAI%—mean value for 40 trees per plot.

4. Discussion

4.1. Estimation of Atmospheric Inputs and Identification of Differences in N Deposition

Several factors may have influenced rain collection efficiency, such as the evaporation of collected rainwater, wind speed during rain events, and the geometry of the collectors [4]. Results of atmospheric deposition chemistry, pH, conductivity, alkalinity, and volume weighted mean concentration of each anion and cation for BOF and THR deposition are presented in Table 1. A smaller amount of precipitation was expected in THR samples due to rainfall interception by the forest canopy [34]. THR samples contained bulk + leached + dry deposition-absorbed ions [25]. The important aspect evident from the comparison of the collected data is that the mean values of pH between two different typologies of samples do not differ significantly. In all plots, the average pH values were higher than 5.70, indicating that no events of acid rain occurred [35].

The difference between THR and the rain collected in an unforested area (BOF) reflects both the wash-off of the dry deposited particles and the exchange with the leaf surfaces (absorption and leaching) [4]. Correlations between the BOF and THR fluxes indicate differences between the plots and typology of the samples (Table 3). Comparing all the results obtained by Wilcoxon test (Table 2), it can be noticed that in three (black pine, holm oak, and pubescent oak plots) out of the four plots, there was a significant difference in deposition between BOF and THR for total alkalinity, suggesting the washing of calcareous soil dust deposited on the canopy at this site that may derive from the calcareous nature of the soils [7].

Furthermore, Na^+ and Cl^- ions are the main constituents of sea salt, being considered the main marker elements of a sea spray source. Ca^{2+} is a significant component of the soil dust and Mg^{2+} is grouped with the crustal elements in Southern Europe [36]. Ca^{2+} and Mg^{2+} can also have marine origins, together with K^+ and $SO_4{}^{2-}$. Nevertheless, ion $SO_4{}^{2-}$ as a marine component was well correlated with anthropogenic origin ions, such as $NO_3{}^-$, in the THR samples and BOF (Table 3). They can also be found in the plots located close to the coastline (distance < 50 m), such as the holm oak plot of the present study (Table 1). The plot is also influenced by the intense agricultural activities in its surroundings, and this is highlighted by the high amount of ammonium. Alkalinity fluxes in THR were about double those in BOF. Higher enrichment in the holm oak plot suggests the washing of calcareous soil dust deposited on the canopy at this site that may derive from the calcareous nature of the soils [7]. High concentrations of potassium are probably due to leakage from leaves. Furthermore, as holm oak is evergreen, the influence of dry deposition of particles from tree canopies can be expected over the whole year. The highest value for Ca^{2+} deposition was measured for THR deposition in the black pine forest (Table 1), while in the case of N-$NO_3{}^-$ deposition, the highest deposition was measured for THR deposition in the Aleppo pine forest. Although Ca^{2+} ion derives from the calcareous nature of soils in all plots, the highest value of Ca^{2+} was measured in the black pine

plot, which is on the hill (550 m above sea level), due to the long-range transport of Saharan dust, which was more evident in our southeast site. Calcium THR deposition is generally high across southern Europe, likely related to contributions from Saharan dust [37]. During its atmospheric transport, it is partly dissolved, transformed, and can be deposited mainly through wet processes, causing red rain events [38]. The spatial pattern of magnesium deposition is mainly dominated by marine sources. Both Ca^{2+} and Mg^{2+} are macronutrients and act as buffers against acidification [37]. Considering N-NO_3^- deposition at the Aleppo pine plot (Table 1), it originates from wet air masses coming from the Adriatic Sea, and the fact that most of this land area is suitable for agricultural production also contributes to its concentration. The accumulation of dust in BOF samples was evident for Mg^{2+} and Ca^{2+} ions in the pubescent oak plot (Table 1). These components were from the soil dust. For BOF samples, the interesting fact was that concentrations of these ions were higher than in THR samples, due to the soil being very rich of those elements. Potassium was derived from the soil dust and from the canopy leaching in the THR samples (Table 1).

Considering throughfall N deposition, it is expected to be higher than in the open field, because the former also accounts for the dry deposition of particles on tree canopies [6]. This was evident in the oak plots. However, throughfall of both N-NO_3^- and N-NH_4^+ was lower than open field fluxes in the black pine and Aleppo pine plots, indicating possible retention from the canopy (Table 1). Furthermore, since atmospheric ammonia is principally involved in neutralizing SO_2 to form ammonium sulphate aerosols, the strong decrease of SO_2 may have resulted in a reduced formation of ammonium aerosols and may have facilitated NH_3 dry deposition close to sources [7]. The canopy might take up NH_4^+ from wet and dry depositions. The exchange of NH_3 with the leaf surface can take place through stomata or physical adsorption to epicuticular waxes or dissolution in water films on the leaves [39]. Other gases, such as SO_2 and HNO_3, interact with NH_3, enhancing the deposition process. NO_3^-, NH_4^+, and SO_4^{2-} ions can be deposited from ammonium sulphate and nitrate aerosols formed by gas phase reaction of ammonia with sulphuric and nitric acids in throughfall deposition [25].

4.2. Actual N Deposition Loads in Mediterranean Forests Compared to Critical Loads

Actual N deposition loads ranged between 3.41 to 19.70 kg N ha^{-1} y^{-1}, with the lowest value in Aleppo pine forest and the highest value in holm oak forest (Table 4). According to Bobbink et al. [31], empirical N critical loads are range from 3 to 15 kg N ha^{-1} y^{-1} for Mediterranean *Pinus* woodland, 10 to 20 kg N ha^{-1} y^{-1} for Mediterranean evergreen (*Quercus*) woodland, and 15 to 20 kg N ha^{-1} y^{-1} for *Quercus*-dominated woodland. When considering our study plots (Table 4), these overlapping values of 10 to 15 kg N ha^{-1} y^{-1} for deciduous and coniferous trees could have an impact on nutrient imbalance, increased N, and decreased concentration of P, K, and Mg in the foliage [31].

In general, only a few geographically scattered N deposition loads are available for the Mediterranean Basin [6,7,40,41].

In Greece, bulk N deposition values of 15 kg N ha^{-1} y^{-1} were found in Aleppo pine stands close to Athens [40]. Throughfall deposition was much higher, indicative of dry deposition capture, reaching values of 38 kg N ha^{-1} y^{-1}. Similar N deposition values (15 kg N ha^{-1} y^{-1}) were measured at an urban site in the center of Thessaloniki, Greece, with dry forms accounting for 70% to 90% of total N inputs [40]. The N load in Spain (15–17 kg ha^{-1} y^{-1}) was within the critical load range proposed for Mediterranean sclerophyllous forests (15–17.5 kg ha^{-1} y^{-1}) [7]. In Italy, the average nitrogen throughfall, in terms of NO_3/NH_4^+, measured using a network of permanent monitoring stations, ranged between 4 and 29 kg N ha^{-1} y^{-1} [6], and the critical loads indicated for Mediterranean forest ecosystems that fall within the range of 10 to 15 kg N ha^{-1} y^{-1} have low reliability owing to the lack of experimental evidence in the Mediterranean region [5].

4.3. The Status of Nutrients in Foliage

For nutrient availability, there are two major sources of variability: Soil chemical composition and climate conditions. In Europe, increased tree productivity, probably caused by high N deposition

and atmospheric CO_2 enrichment, has led to a higher nutrient demand by trees. The consequential deterioration of tree mineral nutrition can at least be partly explained by the limited soil nutrient supply. It is thus crucial to account for nutrient limitation when studying forest response to climate change [42], especially since it could counteract the fertilization effect of higher CO_2 [43]. Nutrient contents of tree foliage, which reflect atmospheric- and soil-related influences, are an important part of UN-ECE ICP's forests monitoring scheme. The basic principle of mineral nutrition can be applied to all plants, where growth can be achieved only if all growth factors are present and in a proper relation. The concentration of elements (nutrients) and element ratios in foliage provides insight in the nutritional status of the tree in terms of deficiency or excess, either in an absolute sense or relative to the concentration of other elements [44].

The cycling and uptake of nutrients have been shown to be an important process for tree vitality. Loss of nutrients from the system, disruption of nutrient cycling and uptake, or imbalances in nutrient status may be associated with declines in tree condition. The lack of a certain element will result in the hindering of dependent physiological functions, depending on the severity of the deficiency.

Compared to chemical analysis of soil, foliar analysis is a direct indicator of the availability of soil nutrients and plant nutrition. Leaf nutrient concentrations depend on several factors: The amount and distribution of precipitation during the growing season, the growing season duration, the soil nutrient amount, the sampling time, the ion antagonism and mobility, etc. By combining soil and foliar nutrient analysis, a complete picture of soil nutrient supply and nutrient accessibility to plants is obtained. On our plots, most elements and species concentrations fall into the plausible range. Although, some deviations can be noticed: Lower N and K in holm oak, lower N in black pine, and higher Ca in all plots except holm oak (Figure 2). The high values of Ca did not, however, have a detrimental effect on the concentrations of K or Mg. Only in holm oak was the concentration of K in the low range, probably due to the fixation of potassium in the heavy clay soil present in the plot. Potassium uptake strongly depends on its availability [45]. Because potassium deficiency reduces plant resistance to abiotic and biotic factors, it is considered within the forest decline hypothesis [46]. Although it is perceived as an unspecific indicator of tree vitality, defoliation seems to be related to the nutritional status of the trees. Ferretti et al. [47] found that the proportion of beech trees with defoliation over 25% increases with increasing ratios of foliar N with Ca and K, indicating that defoliation is related to an imbalance in foliar nutrients.

4.4. Evaluation of Tree Growth and Defoliation

According to Lempereur et al. [48] and Campelo et al. [49], yearly BAI for holm oak shows bi-phasic growth patterns over the year (spring growing period and early autumn growing period). This pattern was also evident on Aleppo pine [50]. However, in our study, only thee spring growing period was the main driver of the annual basal area growth for holm oak (Figure 4b) and pubescent oak (Figure 4a). On the other hand, the Aleppo pine plot showed the mentioned biphasic growth pattern (Figure 4d), and black pine showed continuous intra-annual growth in 2018 (Figure 4c). A study in Spain suggests that drought was the key climatic factor explaining variations in growth patterns of Aleppo pine and black pine [51]. For Mediterranean *Pinus* species, it is known that under favorable growth conditions, the vascular cambium may be active throughout the entire year [52,53]. Tree vitality can be defined as the ability of a tree to assimilate, to survive stressful conditions, to react to a changing environment, and to reproduce [54]. According to the ICP Forests Manual [29], defoliation is defined as leaf loss in the assessable crown as compared to a reference tree. Crown defoliation is an indicator of tree vitality often used in forest practice [55]. At the same time, defoliation is a product of the tree crown status from the past several years of growth, which can be misleading if used as a stress indicator when assessing current vitality [55]. Carnicer et al. [56] showed that in water-limited forests, the main driver of defoliation is drought occurrence. Data on soil, foliar composition, and defoliation from the ICP Forests Level I network, paired with modelled climate data, have shown the substantial importance of climatic parameters as predictors of defoliation in several tree species [57,58]. Holm oak forest was

the most vital plot in our study (Figure 3b). A high percentage of significantly defoliated trees was observed in the Aleppo pine plot and black pine plot. It should be noted that we did not observe zero-class trees (defoliation 0–10%) in the Aleppo pine plot (Figure 3c) while in the black pine plot, we did not observe three- and four-class trees (defoliation 61–99% and 100%) (Figure 3a). The pubescent oak plot was the only plot where dead trees (defoliation 100%) were observed (Figure 3d), which is not surprising considering the greater age of the stand. The cause of the high percentage of significantly defoliated trees in the Aleppo pine plot and black pine plot is a fungus from the *Lophodermium* genus.

Compared to the average estimated defoliation of the crown, it is evident that the plots with lower BAI% have lower defoliation levels that can be due to the different tree species. On the other hand, the increase in the BAI% in 2018 was followed by a slight increase in the average defoliation of the same year (Table 5). Lower BAI% values in 2017 could be a result of the reported drought during the growing season [59]. The observed rise of defoliation one year after the reported drought is consistent with previous findings [60].

Tree growth is an important indicator of forest health, productivity, and demography [61]. Tree diameter growth correlates with biomass, and therefore carbon uptake, as well as with pathogen damage, nutrient availability, and the influence of climate on photosynthesis, among others [62]. For central Europe, growth studies have shown considerable increases in growth during the past decades, with the fertilizing effect of N as one of the possible causes [63]. As shown in the literature, an increase of 1 kg N ha^{-1} y^{-1} corresponds to an increase of the basal area increment between 1.20% and 1.49% depending on the species [64].

5. Conclusions

The results of this research indicate the need for in-depth studies in Mediterranean forest ecosystems. The research was also conducted to bridge the gap in the knowledge and to contribute to a wider overview of Mediterranean forest ecosystems of the Eastern Adriatic coast. This especially relevant for some species, such as pubescent oak and black pine, that are not investigated enough in the European Mediterranean region. The atmospheric inputs and present deposition loads for nutrient N were estimated. Results showed the highest ion concentrations for throughfall deposition in holm oak forest. Throughfall N deposition was expected to be higher than in the open field because it accounts for the dry deposition of particles on tree canopies as well. This was evident in oak forests. However, throughfall of N deposition was lower than open field fluxes in pine forests, indicating possible retention from the canopy. Actual N deposition loads ranged between 3.41 and 19.70 kg N ha^{-1} y^{-1}. This, however, did not have an effect on the concentrations in needles/leaves. On two out of four plots, N was in the low range, while Ca was mostly high, reflecting the chemistry of the predominantly calcareous soils of the region. No relevant differences in mean defoliation between the plots were observed. Compared to the average estimated defoliation of the crown, it is apparent that the plots with a lower BAI% (black pine forest and holm oak forest) have lower defoliation, which can be due to the different tree species accounted for. It is not yet possible to make any conclusion on the relationship between N-deposition and tree growth. With more years of growth observation and using multiannual deposition loads, this evaluation will be done.

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References

1. Sicard, P.; De Marco, A.; Troussier, F.; Renou, C.; Vas, N.; Paoletti, E. Decrease in Surface Ozone Concentrations at Mediterranean Remote Sites and Increase in the Cities. *Atmos. Environ.* **2013**, *79*, 705–715. [CrossRef]
2. EEA (2019) NEC Directive Reporting Status 2018. Available online: https://www.eea.europa.eu/themes/air/national-emission-ceilings/nec-directive-reporting-status-2018 (accessed on 18 August 2019).
3. Myers, N.; Mittermeier, R.A.; Mittermeier, C.G.; da Fonseca, G.A.B.; Kent, J. Biodiversity Hotspots for Conservation Priorities. *Nature* **2000**, *403*, 853–858. [CrossRef] [PubMed]
4. Balestrini, R.; Arisci, S.; Brizzio, M.C.; Mosello, R.; Rogora, M.; Tagliaferri, A. Dry Deposition of Particles and Canopy Exchange: Comparison of Wet, Bulk and Throughfall Deposition at Five Forest Sites in Italy. *Atmos. Environ.* **2007**, *41*, 745–756. [CrossRef]
5. Bobbink, R.; Hicks, K.; Galloway, J.; Spranger, T.; Alkemade, R.; Ashmore, M.; Bustamante, M.; Cinderby, S.; Davidson, E.; Dentener, F.; et al. Global Assessment of Nitrogen Deposition Effects on Terrestrial Plant Diversity: A Synthesis. *Ecol. Appl.* **2010**, *20*, 30–59. [CrossRef]
6. Ferretti, M.; Marchetto, A.; Arisci, S.; Bussotti, F.; Calderisi, M.; Carnicelli, S.; Cecchini, G.; Fabbio, G.; Bertini, G.; Matteucci, G.; et al. On the Tracks of Nitrogen Deposition Effects on Temperate Forests at Their Southern European Range—An Observational Study from Italy. *Glob. Chang. Biol.* **2014**, *20*, 3423–3438. [CrossRef] [PubMed]
7. Aguillaume, L.; Izquieta-Rojano, S.; García-Gómez, H.; Elustondo, D.; Santamaría, J.M. Dry deposition and canopy uptake in Mediterranean holm-oak forests estimated with a canopy budget model: A focus on N estimations. *Atmos. Environ.* **2017**, *152*, 191–200. [CrossRef]
8. García-Gómez, H.; Izquieta-Rojano, S.; Aguillaume, L.; González-Fernández, I.; Valiño, F.; Elustondo, D.; Santamaría, J.M.; Àvila, A.; Bytnerowicz, A.; Bermejo, V.; et al. Joining Empirical and Modelling Approaches to Estimate Dry Deposition of Nitrogen in Mediterranean Forests. *Environ. Pollut.* **2018**, *243*, 427–436. [CrossRef] [PubMed]
9. Ochoa-Hueso, R.; Allen, E.B.; Branquinho, C.; Cruz, C.; Dias, T.; Fenn, M.E.; Manrique, E.; Pérez-Corona, M.E.; Sheppard, L.J.; Stock, W.D. Nitrogen Deposition Effects on Mediterranean-Type Ecosystems: An Ecological Assessment. *Environ. Pollut.* **2011**, *159*, 2265–2279. [CrossRef]
10. Food and Agriculture Organization of the United Nations. State of Mediterranean Forests 2018. Available online: http://www.fao.org/3/CA2081EN/ca2081en.PDF (accessed on 15 September 2019).
11. Matić, S. Proslov. In *Šume Hrvatskog Sredozemlja*; Matić, S., Prpić, B., Jakovac, H., Vukelić, J., Anić, I., Delač, D., Eds.; Akademija Šumarskih Znanosti: Zagreb, Croatia, 2011; pp. 5–7. ISBN 978-953-985715-6.
12. Škevin-Sović, J.; Djuričić, V.; Kosanović, C. Major ions wet deposition and trends during the last decade on the eastern Adriatic coast. In *Air Pollution XX, Proceedings of the 20th International Conference on the Modelling, Monitoring and Management of Air Pollution, Coruña, Spain, 16–18 May 2012*; Longhurst, J.W.S., Brebbia, C.A., Eds.; WIT Press: Southampton, UK, 2012; Volume 157, pp. 339–359. [CrossRef]
13. Clark, C.M.; Cleland, E.E.; Collins, S.L.; Fargione, J.E.; Gough, L.; Gross, K.L.; Pennings, S.C.; Suding, K.N.; Grace, J.B. Environmental and Plant Community Determinants of Species Loss Following Nitrogen Enrichment. *Ecol. Lett.* **2007**, *10*, 596–607. [CrossRef]
14. de Vries, W.; Dobbertin, M.H.; Solberg, S.; van Dobben, H.F.; Schaub, M. Impacts of Acid Deposition, Ozone Exposure and Weather Conditions on Forest Ecosystems in Europe: An Overview. *Plant Soil* **2014**, *380*, 1–45. [CrossRef]
15. Lu, X.; Mo, J.; Dong, S. Effects of Nitrogen Deposition on Forest Biodiversity. *Acta Ecol. Sin.* **2008**, *28*, 5532–5548. [CrossRef]
16. Potočić, N.; Seletković, I. Defoliation of forest trees in Croatia in the period 2006–2009. *Šumarski List* **2011**, *135*, 149–158.
17. Dobbertin, M. Tree growth as indicator of tree vitality and of tree reaction to environmental stress: A review. *Eur. J. For. Res.* **2006**, *125*, 89. [CrossRef]

18. EC. *Communication from the Commission to the Council and the European Parliament on an EU Forest Action Plan*; European Commision: Bruxelles, Belgium, 2006; p. 13. Available online: https://ec.europa.eu/agriculture/sites/agriculture/files/fore/action_plan/com_en.pdf (accessed on 18 September 2019).
19. Larrubia, C.J.; Kane, K.R.; Wolfslehner, B.; Guldin, R.; Rametsteiner, E. *Using Criteria and Indicators for Sustainable Forest Management. A Way to Strengthen Results-Based Management of National Forest Programmes*; Forestry Policy and Institutions Working Paper; Food and Agriculture Organization of the United Nations FAO: Rome, Italy, 2017; p. 85. Available online: http://www.fao.org/3/a-i6883e.pdf (accessed on 15 September 2019).
20. Pernar, N.; Vrbek, B.; Bakšić, D. Soil. In *Šume hrvatskog Sredozemlja/Forests of the Croatian Mediterranean*; Matić, S., Prpić, B., Jakovac, H., Vukelić, J., Anić, I., Delač, D., Eds.; Akademija šumarskih znanosti: Zagreb, Croatia, 2011; pp. 130–141. ISBN 978-953-985715-6.
21. Köpen, W.; Volken, E.; Brönnimann, S. The thermal zones of the earth according to the duration of hot, moderate and cold periods and to the impact of heat on the organic world. *Meteorol. Z.* **2003**, *20*, 351–360. [CrossRef] [PubMed]
22. Zaninović, K.; Gajić-Čapka, M.; Perčec Tadić, M.; Vučetić, M.; Milković, J.; Bajić, A.; Cindrić, K.; Cvitan, L.; Katušin, Z.; Kaučić, D.; et al. *Klimatski atlas Hrvatske/Climate atlas of Croatia 1961–1990.: 1971–2000*; Državni Hidrometeorološki Zavod: Zagreb, Croatia, 2000; p. 200. ISBN 978-953-7526-01-6.
23. Clarke, N.; Zlindra, D.; Ulrich, E.; Mosello, R.; Derome, J.; Derome, K.; König, N.; Lövblad, G.; Draaijers, G.P.J.; Hansen, K.; et al. Part XIV: Sampling and Analysis of Deposition. In *Manual on Methods and Criteria for Harmonized Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests*; UNECE ICP Forests Programme Coordinating Centre, Ed.; Thünen Institute of Forest Ecosystems: Eberswalde, Germany, 2016; p. 66. ISBN 978-3-86576-162-0.
24. König, N.; Kowalska, A.; Brunialti, G.; Ferretti, M.; Clarke, N.; Cools, N.; Derome, J.; Derome, K.; de Vos, B.; Fuerst, A.; et al. Part XVI: Quality Assurance and Control in Laboratories. In *Manual on Methods and Criteria for Harmonized Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests*; UNECE ICP Forests Programme Co-ordinating Centre, Ed.; Thünen Institute of Forest Ecosystems: Eberswalde, Germany, 2016; p. 46. ISBN 978-3-86576-162-0.
25. Jakovljević, T.; Marchetto, A.; Berković, K.; Rosa, J.; Potočki, A. Atmospheric deposition measurement in the lowland forest ecosystem of Pokupsko basin in Croatia. *Period. Biol.* **2013**, *115*, 363–370.
26. Rautio, P.; Fürst, A.; Stefan, K.; Raitio, H.; Bartels, U. Part XII: Sampling and Analysis of Needles and Leaves. In *Manual on Methods and Criteria for Harmonized Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests*; UNECE ICP Forests Programme Co-ordinating Centre, Ed.; Thünen Institute of Forest Ecosystems: Eberswalde, Germany, 2016; p. 19. ISBN 978-3-86576-162-0.
27. Leco Corporation. Carbon, Nitrogen, and Sulphur in Plant Tissue. Organic Application Note, 203-821-172. Available online: https://uk.leco-europe.com/wp-content/uploads/leco_docs/product-files/TRUMAC_CNS_PLANT_TISSUE_203-821-539.pdf (accessed on 9 September 2017).
28. Dobbertin, M.; Neumann, M. Part V: Tree Growth. In *Manual on Methods and Criteria for Harmonized Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests*; UNECE ICP Forests Programme Coordinating Centre, Ed.; Thünen Institute of Forest Ecosystems: Eberswalde, Germany, 2016; p. 17. ISBN 978-3-86576-162-0.
29. Eichhorn, J.; Roskams, P.; Potočić, N.; Timmermann, V.; Ferretti, M.; Mues, V.; Szepesi, A.; Durrant, D.; Seletković, I.; Schröck, H.W.; et al. Part IV: Visual Assessment of Crown Condition and Damaging Agents. In *Manual on Methods and Criteria for Harmonized Sampling, Assessment, Monitoring and Analysis of the Effects of Air Pollution on Forests*; UNECE ICP Forests Programme Coordinating Centre, Ed.; Thünen Institute of Forest Ecosystems: Eberswalde, Germany, 2016; p. 49. ISBN 978-3-86576-162-0.
30. Mosello, R.; Brizzio, M.C.; Kotzias, D.; Marchetto, A.; Rembges, D.; Tartari, G. The Chemistry of Atmospheric Deposition in Italy in the Framework of the National Programme for Forest Ecosystems Control (CONECOFOR). *J. Limnol.* **2002**, *61*, 77–92. [CrossRef]
31. Bobbink, R.; Braun, S.; Nordin, A.; Power, S.; Schütz, K.; Strengbom, J.; Weijters, M.; Tomassen, H. Effects of nitrogen deposition on woodland, forest and other wooded land (eunis class G). In *Review and Revision of Empirical Critical Loads and Dose-Response Relationships, Proceedings of an Expert Workshop, Noordwijkerhout, The Netherlands, 23–25 June 2010*; Bobbink, R., Hettelingh, J.P., Eds.; National Institute for Public Health and the Environment: Bilthoven, The Netherlands, 2011; pp. 135–160.

32. R Core Team. A Language and Environment for Statistical Computing. In *R Foundation for Statistical Computing*; R Core Team: Vienna, Austria, 2016; Available online: https://www.R-project.org/ (accessed on 10 February 2019).
33. Clarke, N.; Cools, N.; Derome, J.; Derome, K.; De Vos, B.; Fuerst, A.; Koenig, N.; Kowalska, A.; Mosello, R.; Tartari, G.A.; et al. *Quality Assurance and Control in Laboratories. A Review of Possible Quality Checks and Other Forms of Assistance*; ICP Forests Working Group on QA/QC in Laboratories, Ed.; Thünen Institute of Forest Ecosystems: Eberswalde, Germany, 2008; p. 56.
34. Llorens, P.; Domingo, F. Rainfall Partitioning by Vegetation under Mediterranean Conditions. A Review of Studies in Europe. *J. Hydrol.* **2007**, *335*, 37–54. [CrossRef]
35. Karavoltsos, S.; Fotiadis, F.; Michopoulos, P.; Sakellari, A.; Plavšić, M.; Bourletsikas, A.; Kaoukis, K.; Thomaidis, N.S.; Dassenakis, M.; Scoullos, M. Organic complexation of copper in throughfall and open field bulk deposition: Influence of the tree canopy of Mediterranean forest ecosystems. *Chemosphere* **2017**, *167*, 28–35. [CrossRef]
36. Viana, M.; Kuhlbusch, T.A.J.; Querol, X.; Alastuey, A.; Harrison, R.M.; Hopke, P.K.; Winiwarter, W.; Vallius, M.; Szidat, S.; Prévôt, A.S.H.; et al. Source Apportionment of Particulate Matter in Europe: A Review of Methods and Results. *J. Aerosol Sci.* **2008**, *39*, 827–849. [CrossRef]
37. Michel, A.; Seidling, W.; Prescher, A.K. Forest condition in Europe. In *2018 Technical Report of ICP Forests. Report under the UN-ECE Convention on Long-Range Transboundary Air Pollution (Air Convention), BFW, Dokumentation 25/2018*; Michel, A., Seidling, W., Prescher, A.K., Eds.; BFW Austrian Research Center of Forest: Vienna, Austria, 2018; p. 92. ISBN 978-3-903258-08-2.
38. Díaz-Hernández, J.L.; Párraga, J. The Nature and Tropospheric Formation of Iberulites: Pinkish Mineral Microspherulites. *Geochimic. Cosmochim. Ac.* **2008**, *72*, 3883–3906. [CrossRef]
39. Burkhardt, J.; Kaiser, H.; Kappen, L.; Goldbach, H.E. The Possible Role of Aerosols on Stomatal Conductivity for Water Vapour. *Basic Appl. Ecol.* **2001**, *2*, 351–364. [CrossRef]
40. Anatolaki, C.h.; Tsitouridou, R. Atmospheric Deposition of Nitrogen, Sulfur and Chloride in Thessaloniki, Greece. *Atmos. Res.* **2007**, *85*, 413–428. [CrossRef]
41. Avila, A.; Molowny-Horas, R.; Gimeno, B.S.; Peñuelas, J. Analysis of decadal time series in wet N concentrations at five rural sites in NE Spain. *Water Air Soil Pollut.* **2010**, *207*, 123–138. [CrossRef]
42. Jonard, M.; Fürst, A.; Verstraeten, A.; Thimonier, A.; Timmermann, V.; Potočić, N.; Waldner, P.; Benham, S.; Hansen, K.; Merilä, P.; et al. Tree Mineral Nutrition Is Deteriorating in Europe. *Glob. Chang. Biol.* **2015**, *21*, 418–430. [CrossRef]
43. Körner, C. Plant CO_2 Responses: An Issue of Definition, Time and Resource Supply. *New Phytol.* **2006**, *172*, 393–411. [CrossRef]
44. de Vries, W.; Reinds, G.J.; van Keerkvoorde, M.S.; Hendriks, C.M.A.; Leeters, E.E.J.M.; Gross, C.P.; Voogd, J.C.H.; Vel, E.M. *Intensive Monitoring of Forest Ecosystems in Europe Technical Report 2000*; FIMCI, EC-UN/ECE: Brussels, Geneva, 2000; p. 191.
45. Mengel, K.; Kirkby, E.A. *Principles of Plant Nutrition*, 5th ed.; Kluwer Academic Publishers: Dordrecht, The Netherlands, 2001; p. 849. ISBN 978-0-7923-7150-2.
46. Bergmann, W. Buchbesprechungen. In *Nutritional Disorders of Plants*, 3rd ed.; Bergmann, W., Ed.; Gustav Fischer Verlag: Jena, Germany, 1992; pp. 377–379. ISBN 3334604225.
47. Ferretti, M.; Calderisi, M.; Marchetto, A.; Waldner, P.; Thimonier, A.; Jonard, M.; Cools, N.; Rautio, P.; Clarke, N.; Hansen, K.; et al. Variables Related to Nitrogen Deposition Improve Defoliation Models for European Forests. *Ann. For. Sci.* **2015**, *72*, 897–906. [CrossRef]
48. Lempereur, M.; Martin-St.Paul, N.K.; Damesin, C.; Joffre, R.; Ourcival, J.-M.; Rocheteau, A.; Rambal, S. Growth duration is a better predictor of stem increment than carbon supply in a Mediterranean oak forest: Implications for assessing forest productivity under climate change. *New Phytol.* **2015**, *207*, 579–590. [CrossRef]
49. Campelo, F.; Gutierrez, E.; Ribas, M.; Nabais, C.; Freitas, H. Relationships between climate and double rings in *Quercus ilex* from northeast Spain. *Can. J. For. Res.* **2007**, *37*, 1915–1923. [CrossRef]
50. Camarero, J.J.; Olano, J.M.; Parras, A. Plastic Bimodal Xylogenesis in Conifers from Continental Mediterranean Climates. *New Phytol.* **2010**, *185*, 471–480. [CrossRef]

51. Martinez del Castillo, E.; Prislan, P.; Gričar, J.; Gryc, V.; Merela, M.; Giagli, K.; de Luis, M.; Vavrčík, H.; Čufar, K. Challenges for Growth of Beech and Co-Occurring Conifers in a Changing Climate Context. *Dendrochronologia* **2018**, *52*, 1–10. [CrossRef]
52. Liphschitz, N.; Lev-Yaddun, S.; Rosen, E.; Waisel, Y. The annual rhythm of activity of the lateral meristems (cambium and phellogen) in Pinus halepensis Mill. and P. pinea L. *IAWA J.* **1984**, *5*, 263–274. [CrossRef]
53. Prislan, P.; Gričar, J.; de Luis, M.; Novak, K.; Martinez del Castillo, E.; Schmitt, U.; Koch, G.; Štrus, J.; Mrak, P.; Žnidarič, M.T.; et al. Annual Cambial Rhythm in Pinus halepensis and Pinus sylvestris as Indicator for Climate Adaptation. *Front. Plant Sci.* **2016**, *7*, 1923. [CrossRef] [PubMed]
54. Brang, P. *Sanasilva-Bericht 1997. Zustand und Gefährdung des Schweizer Waldes—Eine Zwischenbilanz Nach 15 Jahren Waldschadenforschung*; Ber. Eidgenöss. Forsch.anst; Wald Schnee Landsch: Birmensdorf, Schweiz, 1998; p. 345. ISBN 3-905620-67-7.
55. Polak, T.; Rock, B.N.; Campbell, P.E.; Soukupova, J.; Solcova, B.; Zvara, K.; Albrechtova, J. Shoot Growth Processes, Assessed by Bud Development Types, Reflect Norway Spruce Vitality and Sink Prioritization. *For. Ecol. Manag.* **2006**, *225*, 337–348. [CrossRef]
56. Carnicer, J.; Coll, M.; Ninyerola, M.; Pons, X.; Sánchez, G.; Peñuelas, J.; Mooney, H.A. Widespread Crown Condition Decline, Food Web Disruption, and Amplified Tree Mortality with Increased Climate Change-Type Drought. *Proc. Natl. Acad. Sci. USA* **2011**, *108*, 1474–1478. [CrossRef] [PubMed]
57. Vitale, M.; Proietti, C.; Cionni, I.; Fischer, R.; De Marco, A. Random forests analysis: A useful tool for defining the relative importance of environmental conditions on crown defoliation. *Water Air Soil Pollut.* **2014**, *225*, 1992. [CrossRef]
58. de Marco, A.; Proietti, C.; Cionni, I.; Fischer, R.; Screpanti, A.; Vitale, M. Future Impacts of Nitrogen Deposition and Climate Change Scenarios on Forest Crown Defoliation. *Environ. Pollut.* **2014**, *194*, 171–180. [CrossRef]
59. Strelec Mahović, N. *Meteorološki i Hidrološki Bilten 07/2017*; Državni Hidrometeorološki Zavod: Zagreb, Croatia, 2017; p. 51. ISSN 1334-3017.
60. Graf Pannatier, E.; Dobbertin, M.; Schmitt, M.; Thimonier, A.; Waldner, P. Effects of the drought 2003 on forests in Swiss Level II plots. In *Schriften aus der Forstlichen Fakultät der Universität Göttingen und der Nordwestdeutschen Forstlichen Versuchsanstalt, Proceedings of the Forests in a Changing Environment—Results of 20 Years ICP Forests Monitoring, Göttingen, Germany, 25–28 October 2006*; Eichhorn, J., Ed.; Sauerländer: Frankfurt am Main, Germany, 2007; Volume 142, pp. 128–135. ISBN 9783793951421.
61. McMahon, S.M.; Parker, G.G. A general model of intra-annual tree growth using dendrometer bands. *Ecol. Evol.* **2015**, *5*, 243–254. [CrossRef]
62. Barford, C.C.; Wofsy, S.C.; Goulden, M.L.; Munger, J.W.; Pyle, E.H.; Urbanski, S.P.; Hutyra, L.; Saleska, S.R.; Fitzjarrald, D.; Moore, K. Factors Controlling Long- and Short-Term Sequestration of Atmospheric CO_2 in a Mid-Latitude. *Science* **2001**, *294*, 1688–1691. [CrossRef]
63. Spiecker, H.; Mielikäinen, R.; Köhl, M.; Skorgsgaard, J.P. Growth Trends and site productivity. Has site productivity changed? In *Growth Trends in European Forests*, 1st ed.; Spiecker, H., Mielikäinen, R., Köhl, M., Skorgsgaard, J.P., Eds.; Springer: Berlin/Heidelberg, Germany, 1996; p. 372, ISBN 13: 978-3-642-64734-5.
64. Laubhann, D.; Sterba, H.; Reinds, G.J. The Impact of Atmospheric Deposition and Climate on Forest Growth in European Monitoring Plots: An Individual Tree Growth Model. *For. Ecol. Manag.* **2009**, *258*, 1751–1761. [CrossRef]



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