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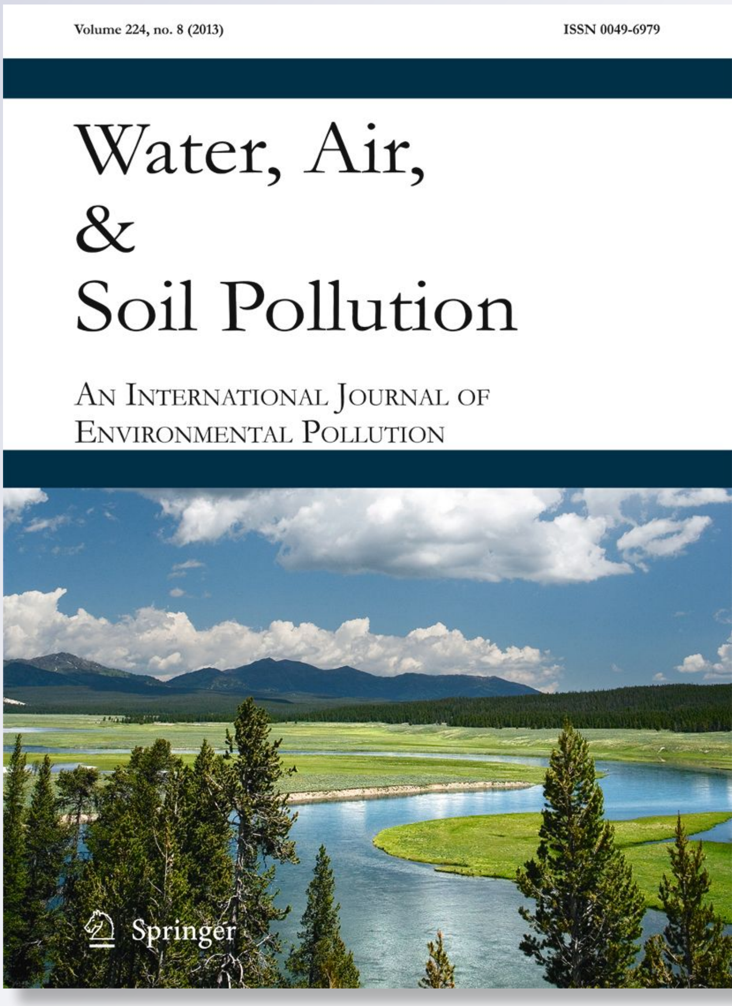

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1 Introduction

For more than a century, a trend of increasing tropospheric ozone concentration has been confirmed in the Northern hemisphere. Nowadays, the current average of tropospheric ozone is between 35 and 50 ppb (IPCC 2013; Cooper et al. 2012; Lefohn et al. 2017). This increase is explained by the formation of anthropogenic tropospheric ozone. Naturally, without human intervention, the tropospheric ozone represents a small part of atmospheric ozone. The formation of ozone results from complex photooxidative reactions involving chemical precursors and UV radiations. Temperature plays also a major role in the formation of ozone. Since the average global ambient temperature increased by 1.5° between 1850 and 2012 (IPCC 2013), and ozone formation is enhanced by solar radiation, the peaks of pollution are generally observed during spring and summer when temperatures are higher (The Royal Society 2008). Ozone is a common pollutant (UNECE 2016), a powerful greenhouse gas with very high radiative forcing (Stevenson et al. 2013; Kulkarni et al. 2015), involved in climate change (Kampa and Castanas 2008; Screpanti and De Marco 2009; Bytnerowicz et al. 2013). Increasing human activity, particularly road traffic and industrial development (Vestreng et al. 2009), is one of the main factors responsible for the presence of this photooxidant in large quantities, and this trend is expected to continue in the coming years (Bytnerowicz et al. 2004).

The Mediterranean region in France is prone to photooxidant formation. Indeed, it is a dense populated a sunny area where important air masses are highly contaminated by chemical precursors. These factors enhance ozone formation and the Mediterranean perimeter presents the highest critical levels regarding forests' health (EEA 2017; Sicard et al. 2016) in comparison to national data. The average annual ozone concentration is over 40 ppb and maximum hourly concentration over 120 ppb.

The countryside is directly affected at a large scale, by the diffusion of the chemical precursors which are produced in close urban areas. In the south of France, some studies have showed that in rural alpine areas, and more precisely in the Mercantour National Park, high concentrations of atmospheric ozone are explained by the transfer of air masses from the French Riviera and the plain of the Po in Italy. In these urbanized areas, ozone and its precursors are generated by industrial activities and road traffic (Sicard et al. 2011). Forests are then the most affected areas by contamination produced in urban conurbations (Sandermann 1996), and this situation will get worse in the future (Matyssek and Innes 1999).

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The negative effects of ozone on ecosystems are well known (Bytnerowicz et al. 2013; Cristofori et al. 2014; Feng et al. 2015; Sicard and Dalstein-Richier 2015). Adverse effects of ozone on forests were first identified in the USA, in The San Bernardino Mountains, which are located downwind of the Los Angeles metropolitan area pollution source. Specific symptoms as foliage yellowing followed by premature fall and tree growth retardation were described in response to high ozone concentrations (Parmeter et al. 1962; Miller 1973; Skelly et al. 1997; Novak et al. 2005; Paoletti 2006). These damages are visible on herbaceous and shrub or tree vegetation (Gottardini et al. 2016; Gottardini et al. 2017), and a list of sensitive plants to ozone has been consolidated considering 19 countries at a global scale. In every European countries, the ICP-Forests Network (International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests) reports every year, the damages due to ozone fluxes observed in forests areas (ICP Annual Vegetation Report 2015/2016).

Among the ozone damages reported, pine species are more reactive and sensitive that other conifers like fir or spruce (Dalstein and Vas 2005; Sicard et al. 2010; Sicard and Dalstein-Richier 2015). Aleppo pine (*Pinus halepensis*), black pine (*Pinus laricio*), and especially Arolla pine (*Pinus cembra*) are the most affected species (Sicard and Dalstein-Richier 2015). In the case of broadleaved trees, the beech, *Fagus sylvatica,* is one of the most ozone-sensitive (Schaub et al. 2007).

Ozone is an oxidizing gas that penetrates via stomata into foliar tissues causing cellular damage, responsible for visible leaf symptoms on shrub and tree vegetation (Dalstein et al. 2002; Contran and Paoletti 2007; Tiwariet et al. 2016). On the French forest plots of the National Forestry Office, the foliar deficit has also been monitored for some 20 years because it is known to be a good indicator of trees' health. Defoliation is directly related to aging, droughts, or nutrient deficiencies and can be used to monitor trees health status over the years (Ferretti and Fischer 2013). Ozone can act indirectly by weakening the defenses of individuals; it indirectly contributes to the general poor state of tree species recognized to be sensitive to ozone (Dalstein et al. 2002; Ulrich et al. 2006; Vollenweider and Günthardt-Goerg 2006; Fares et al. 2013). Generally, ozone damage increases parasitic pathologies on trees (Dalstein et al. 2002; Karnosky et al. 2007; Paoletti et al. 2009; Sicard et al. 2011). Meteorological parameters such as temperature and rainfall, associated to ozone concentration, seem to influence defoliation, but studies are sometimes contradictive one to another about the importance of each parameter. In the Mediterranean area, it has been proved that the most important defoliation factors for *Pinus halepensis* were the atmospheric temperature and the soil water content. In the case of *Pinus cembra,* another factor was the annual amount of precipitations (Sicard and Dalstein-Richier 2015).

Similarly to defoliation factors, depending on the studies carried out, visible foliar symptoms are more or less correlated with the concentration of ozone. In the south of France, where pine defoliation (*Pinus cembra*) has been monitored since the 2000s, a significant upward trend has been observed (Sicard and Dalstein-Richier 2015).

The GIEFS (Groupe International d'Etudes des Forêts Sud-Européennes) and the National Forestry Office (ONF) have been working together monitoring ozone concentrations and their effects on coniferous and deciduous forest plots of the RENECOFOR network (National Long-Term Monitoring Network of Forest ecosystems). These are distributed all over the French national territory. Within the framework of European regulations no. 3528/86 and 2152/2003 (Forest Focus), the collaboration between the GIEFS and the ONF has allowed researchers to test and improve the methods proposed by the ICP "Forest" (Geneva Convention on long-distance trans-boundary pollution) on a selection of RENECOFOR network sites. These results were summarized by Ulrich et al. (2006).

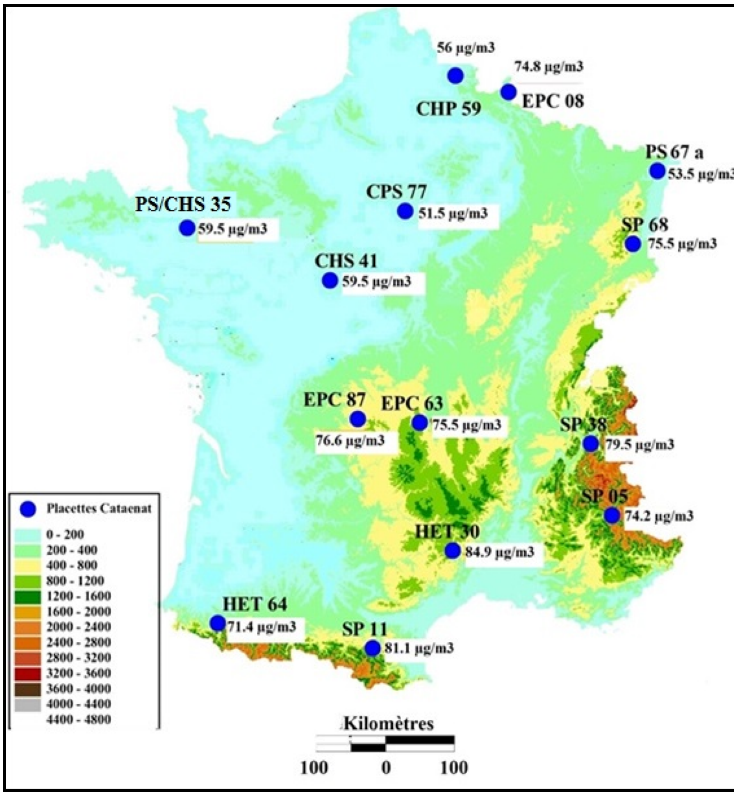
Observations were also made on clearings (LESS) located near the plots in order to detect damages specifically induced by ozone on the vegetation outside the forest.

This study presents the results of the monitoring of foliar ozone damages and ozone concentration from 2013 to 2015, on a selection of 15 permanent forest plots (Fig. 1).

The tree species chosen for their sensitiveness to ozone were:

- Conifers: fir (*Abies alba*), Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*).
- Broadleaves: beech (*Fagus sylvatica*), sessile oak (*Quercus petraea*), and pedunculate oak (*Quercus robur*).

To know the evolution of trees' health status, defoliation measures of the past 15 years on these species, has been analyzed in order to identify patterns for each plot.



Plot	Specie	Altitude (m)	County name
CHP 59	*Quercus robur*	149	Nord
CHS 35	*Quercus petraea*	80	Ile-et-Vilaine
CHS 41	*Quercus petraea*	127	Loir-et-Cher
CPS 77	*Quercus robur/ petraea*	8	Seine-et-Marne
HET 30	*Fagus sylvatica*	1400	Gard
HET 64	*Fagus sylvatica*	400	Pyrénées-Atlantique
EPC 08	*Picea abies-*	480	Ardennes
EPC 63	*Picea abies-*	950	Puy-de-Dôme
EPC 87	*Picea abies-*	650	Haute-Vienne
PS 67a	*Pinus sylvestris*	175	Bas-Rhin
PS 35	*Pinus sylvestris*	80	Ile-et-Vilaine
SP 05	*Abies alba-*	1360	Hautes-Alpes
SP 11	*Abies alba-*	950	Aude
SP 38	*Abies alba-*	1100	Isère
SP 68	*Abies alba-*	680	Haut-Rhin

Fig. 1 Location, species studied, altitude, and geographical area of each RENECOFOR forest plot selected for ozone damage monitoring—National Forestry Office (ONF) from 2000 to 2015 and average ozone concentrations (µg m^{3}) from 2013 to 2015

2 Materials and Methods

2.1 The RENECOFOR Monitoring Plots

The RENECOFOR network, launched in 1992, is built on a selection of 102 permanent monitoring plots. The choice of the sites was based on regional characteristics and main species of each location, making sure to choose healthy trees in high forest.

The RENECOFOR plots are upheld under the same management operations and planned forest thinning have been carried out on most of the sites. The plots are located all over the country; they cover the range of French environmental conditions and include the main species of the territory. *Fagus sylvatica, Quercus petraea*, and *Quercus robur* in the case of broadleaved trees, and *Pinus sylvestris, Abies alba*, and *Picea abies* for conifers, are the most represented. Each plot has a surface area of 0.5 ha.

2.2 Study Area and Sampling Sites

Figure 1 shows the distribution of the 15 forest plots of the RENECOFOR network in France. They were selected at different altitudes for this study, between 2013 and 2015.

2.3 Sampling and Scoring of Visible Ozone Symptoms

Many plant species react to ambient levels of ozone pollution with specific visible foliar injuries. The assessment of these symptoms is realized in:

2.3.1 IMP

Seventy-five trees were monitored on each of the 15 plots from 2013 to 2015, according to the European protocol defined by the ICP-Forests Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests; Ferretti et al. 2014; Harmens et al. 2016), to determine the percentage of crown defoliation and the percentage of needle or leaf surface area affected by ozone-induced symptoms for the main tree species (MTS). The ozone damage measures were conducted on leaves or needles of the upper fully sun exposed crown, at the end of July and August, which is the period when ozone levels are highest. These records were carried out before the month of September because leaf senescence tends to make the determination of ozone symptoms more difficult, especially for broadleaves (Tagliaferro et al. 2005; Schaub et al. 2010).

Assessment was based on the ICP-Forests Manual on Assessment of Ozone Injury (Innes et al. 2001). On each plot, every year, the evaluation of specific ozone symptoms is performed on the same five trees. On each one of these five, a minimum of three branches of the sunned upper part of the crown are removed. For the broadleaves, the percentage of symptomatic leaves (on a minimum of 30 leaves) per branch is estimated and recorded. In the case of conifers, the different needle age classes are identified. Only current year (C), 1-year-old (C + 1), and 2-year-old needles (C + 2) are assessed. The chlorotic mottling is recorded for each needle age class in percentage of the total surface affected.

Leaves or needles with ozone symptoms are photographed. If the observed ozone injury is masked by yellowing due to another cause, the sample is excluded from the evaluation.

2.3.2 LESS

Since most of the intensive monitoring plots (IMP) are situated in closed forests, and visible ozone injury is usually restricted to the sunlight exposed upper most crown part, a special light exposed sampling site (LESS) is installed in the vicinity of the IMP.

The LESS was established close to the meteorological station where a passive ozone sensor is installed. The aim of vegetation assessment inside the LESS was to estimate foliar damages due to ozone in forest border areas close to the studied plots, and more particularly, within a radius of 500 m (European protocol defined by the ICP-Forests Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests; Ferretti at al. 2014).

The most common trees, shrubs, and perennial species in the LESS area were assessed during summer. It is important to highlight that when the observations were realized, a taxonomy expert was present to identify plant species.

Visible ozone symptoms on the listed species in the LESS area were photographed and sampled in order to be preserved in an herbarium. The classification was realized according to Flora Europaea rules.

This list was then compared to the European ozone sensitive species list to determine which of them were present in the LESS. This site permitted to control visible ozone injuries at an extended number of species including, if present, the main tree species.

2.4 Crown Defoliation

Studies in Europe have shown that leaf deficiency of trees reflects their health status (De Marco et al. 2014). These are based on a comparison between the crown condition of observed trees and a "reference" tree at the same stage of development and under the same station conditions. In France, the Forest Health Department (DSF-Département Santé des Forêts) recommends to evaluate defoliation on a minimum of 20 trees. In this study, the ONF chose to assess crown defoliation on 52 selected trees on each plot.

The crown condition was estimated in classes of 5%, between 0 and 100%. This method was used to reduce the margin of error. The two extreme values indicate that a tree is perfectly healthy (0%) or is dead (100%).

The "warning" stage is reached when a foliar deficit is between 10 and 25%. Tree damage is established when the percentage of defoliation is greater than 25% (Michel et al. 2014). The average percentage of defoliation was determined on the 52 trees of each of the 15 plots; thus, a total of 780 trees were evaluated every year.

In addition to the visible symptoms that have been identified as specific to ozone gas, several factors influence crown condition such as microphylla, branching damages, premature drop of leaves/needles, or branches mortality.

2.5 Meteorological Data and Ozone Data

PULSIA meteorological stations developed by PULSONIC company were used for this study because they can be network managed and because of their conformity to the standards established by the World Meteorological Organization (WMO). Météo-France, the national weather agency, undertook this kind of station after a series of test realized by the service in charge of Equipment and Meteorological Instrumental

Techniques (SETIM). These stations are powered by solar energy and are equipped with a memory card. They can support until seven different sensors.

The meteorological stations were installed within a distance lower than 2500 m from the permanent observation plots. The locations were chosen to ensure the optimal exposition of measuring instruments. All the stations were installed on flat ground or slopes covered with short grass. The surface area of each one was 6 × 6 m.

The characteristics of the sensors used in this study are listed in Table 1. The measuring heights for air temperature and relative humidity (1.5 m) considered the range of values recommended by the WMO (between 1.25 and 2 m). The 1 m height for precipitation measurement was chosen to avoid contribution from water which could bounce on the soil. The choice of parameters and data's acquisitions rates were already justified (Q. Ponette et al. 1996). A daily data is established when the number of data is a least equal to 80% of the maximum expected during a 24 h period. In other terms, to establish a daily data, 39 half-hourly data or 1152 min data must be validated in hourly stations. In the case of half-hourly stations, daily data are calculated using half-hourly values after their transmission. For hourly stations, daily data are calculated by the microprocessor using measures acquired every 0.5 s or every minute. The air relative humidity is expressed in percentage (%), precipitations in millimeters (mm), and temperatures in Celsius degrees (°C).

The ONF provided the following daily meteorological data for each plot from 2013 to 2015: total precipitation amount (mm), maximum precipitation (mm / 6mn), mean temperature (°C), maximum temperature (°C), minimum temperature (°C), mean relative humidity (%), minimum relative humidity (%), maximum relative humidity (%), and relative humidity >90% (hours).

Annual averages were calculated using these daily data for each parameter. In addition, for every year and every plot, the number of days and hours when temperatures were above 30 °C (temperature > 30 °C) were calculated during the period 2013–2015. Finally, it is important to highlight that a meteorological station was installed on/nearby each plot.

On one hand, each meteorological station was equipped with passive sensors to measure the average monthly ozone concentration. These permitted measurements in mountainous areas, far away from any electrical access (Dalstein et al. 2004). These sensors allowed then to record the periods of high ozone concentrations and also indicate its spatial distribution in these zones sometimes difficult to reach.

On the other hand, the foliar symptoms which can be observed are caused by a long-term exposition to ozone. Thus, it is impossible to establish a correlation between high temperatures or recorded ozone concentrations peaks, with the exact time of appearance of these foliar symptoms. These were recorded once a year, usually at the end of summer, and were the result of an accumulation of ozone effects on a long term, corresponding to a chronic more than to an acute pollution.

The passive ozone sensors chosen for this study were developed in Sweden by IVL (IVL Laboratories Sweden), an environmental monitoring institute which is recognized at European level. The ozone sensors were placed at 3 m height above the ground and were protected from the rain by a metal plate. There were always located outside the forest cover, nearby the studied plots. Each sensor contained a filter soaked in a solution that absorbs atmospheric ozone by molecular diffusion. The sensors were then sent to the laboratory IVL in Sweden to determine the ozone concentration by ionic chromatography.

These passive ozone sensors were used to measure the average monthly ozone concentration on the RENECOFOR plots (Fig. 1) during the growing season (April-September), every year, from 2000 to 2015.

2.6 Statistics and Estimation of Annual Trends

Time series of environmental data often contain missing or extreme values or do not have a normal distribution. For these, non-parametric tests must be used; another advantage is that the number of data can be reduced. Among the non-parametric tests, the Mann-Kendall test allows to detect the presence of increasing or decreasing trends in a time series. Numerous studies have proved the effectiveness of this test (Sen 1968; Ruoho-Airola et al. 2004; Lehmann et al. 2005). The confidence intervals used are 0.01 (99%) and 0.05 (95%).

This study applied the Mann-Kendall test to the foliar deficit data obtained from 2000 to 2015, to determine whether or not there was a significant defoliation trend in each of the 15 study plots. A Spearman test was then carried out to understand the contribution of ozone concentrations and meteorological parameters (mean temperature, relative humidity, and precipitation from April to September) to crown defoliation. Meteorological data for each year, from 2013 to 2015, were provided

Table 1 Sensor specifications

Parameter	Sensor	Manufacturer	Operating range	Precision	Resolution	Measuring height
Air temperature (T)	Resistance thermometer	Engelhard	$40 < T < +60$ °C	±0.07 to 0 °C	±0.1 °C	1.5 m (under shelter)
Air relative humidity (H)	Capacitive sensor	Pulsonic	$0 < H < 100\%$	±2% (10 < H < 96%)	1%	1.5 m (under shelter)
Precipitation (P)	Tipping bucket rain gauge (receiving area 400 cm^2)	Précis mécanique	$0 < P < 7.5$ mm min^{-1}	±0.2 mm	0.2 mm	1 m

by the existing ONF's equipment for each plot, that is to say, average temperature, relative humidity, precipitation, and ozone concentrations from April to September.

3 Results

3.1 Defoliation

The results of the Mann-Kendall test indicate that foliar deficit for the conifers *Picea Abies* and *Abies alba* has increased significantly over the past 15 years. Broadleaved species have not suffered such a clear trend (Table 2). Monitoring of Air Pollution Effects on Forests shows average leaf deficit between 7.69 and 27.15% (Table 2) that means warning stage of health conditions of the main species in France.

The decrease in rainfall is responsible for an increase in the average percentage of defoliation of *Pinus Sylvestris* and *Picea abies* (Table 3). *Abies alba* is the only conifer where no correlation between defoliation and meteorological parameters were founded (Table 3), as in the case of all other broadleaves studied (*Quercus robur*, *Quercus petraea*, and *Fagus sylvatica*).

3.2 Ozone Symptoms

In 2015, ozone foliar damages were globally more important than during precedent years considering all the studies forest plots (Fig. 2). It is the only time that leaf injuries were recorded on pine *Abies alba* and oak *Quercus robur* (Fig. 2). Greater injuries were observed on the older needles in all species (Fig. 2). Broadleaves have always shown more resistance to ozone damage than conifers.

3.3 Sensitivity of Species to Ozone

Among conifers, *Pinus sylvestris* was more affected by ozone than fir and spruce (Figs. 3 and 4). Considering broadleaves, beech trees were more sensitive to ozone than the two species of oak (Fig. 3). The symptoms observed on *Pinus sylvestris* needles are small pale yellow points, which are called *mottling* (Fig. 4a). In general, the symptoms observed on beech leaves (bronzing followed by stippling) were quite characteristic (Fig. 4b). The damages were much less visible on oaks, where only a slight bronzing could be seen (Fig. 4c).

Table 2 Mann-Kendall test showing the average leaf deficit for each species and the presence or absence of a trend from 2000 to 2015 for each plot (*p* value = 0.001***, 0.01**, 0.05*, 0.1+, > 0.1, not significant = ns)

Plot	Species	Average leaf deficit (%) per plot	Average leaf deficit (%) for each specie	Trend (% year^{-1})
CPS 77	*Quercus petraea/robur*	41.2 ± 9.9	–	+1.33^{+}
CHS 41	*Quercus petraea*	23.5 ± 3.9	21.75 ± 6	ns
CHS 35	*Quercus petraea*	19.9 ± 7.2		1.01*
CHP 59	*Quercus robur*	27.1 ± 9.1	27.15 ± 9.15	ns
HET 64	*Fagus sylvatica*	24.1 ± 10.1	23.25 ± 10.95	+1.20^{+}
HET 30	*Fagus sylvatica*	22.4 ± 12.1		+1.18*
SP 68	*Abies alba*	5.8 ± 3.4	18.56 ± 15.7	+0.47***
SP 38	*Abies alba*	29.1 ± 22.9		+4.98***
SP 11	*Abies alba*	18.6 ± 13.3		+2.53*
SP 05	*Abies alba*	21.6 ± 5.1		ns
EPC 63	*Picea abies*	7.7 ± 4.9	7.69 ± 4.4	+1.01***
EPC 08	*Picea abies*	9.9 ± 4.4		+0.98***
EPC 87	*Picea abies*	5.3 ± 2.3		+0.34*
PS 67 a	*Pinus sylvestris*	17.7 ± 7.3	17.7 ± 7.3	ns
PS 35	*Pinus sylvestris*	12.6 ± 2.4		ns

3.4 With regard to LESS plots: vegetation in clearings

In 2015, ozone damages were much more important on vegetation situated in clearings than during the two precedent years (Figs. 5 and 6). In these areas, the most sensitive species was *Corylus avellana* (Fig. 7). The hornbeam *Carpinus betulus*, the beech *Fagus sylvatica*, and brambles (Rubus sp.) are also sensitive to ozone (Fig. 7).

3.5 Ozone Concentrations and Meteorological Parameters

Figure 1 shows average levels of ozone concentrations across all the RENECOFOR plots considered in this study. The highest values were generally recorded in the southern half of the national territory, which is known to be sunnier and more affected by high concentrations of ozone. In the northern half of the country, only the plots in the Ardennes (EPC 08) and Alsace (SP 68), both of which are mountainous zones, had high values of ozone concentration.

During the 3 years of monitoring (Fig. 1), mountain sites presented the highest ozone values. Figure 7 shows a good correlation between ozone pollution levels and the altitude of these plots (R^2 = 0.7409). Lowland sites, at lower altitudes (between 0 and 200 m above sea level), contain mostly oak trees which are less subject to high concentrations of ozone. Indeed, the altitudinal gradient is an important parameter to consider as ozone concentration increases with altitude in mountainous areas

Table 3 Spearman correlation and *p* value (*p* value = 0.01**, 0.05*, not significant = ns) between leaf deficit of each species and average of temperature, precipitation, relative humidity, and ozone concentration from April to September over the period 2000–2015 on all plots

	Annual temperature (°C)	Precipitation (mm)	Relative humidity (%)	Ozone concentration (μg m^3)
Pinus sylvestris leaf deficit (%)	ns	− 0.758*	ns	ns
Picea abies leaf deficit (%)	ns	− 0.370*	0.540**	ns
Abies alba leaf deficit (%)	ns	ns	ns	ns
Quercus petraea leaf deficit (%)	ns	ns	ns	ns
Fagus sylvatica leaf deficit (%)	ns	ns	ns	ns
Quercus robur leaf deficit (%)	ns	ns	ns	ns

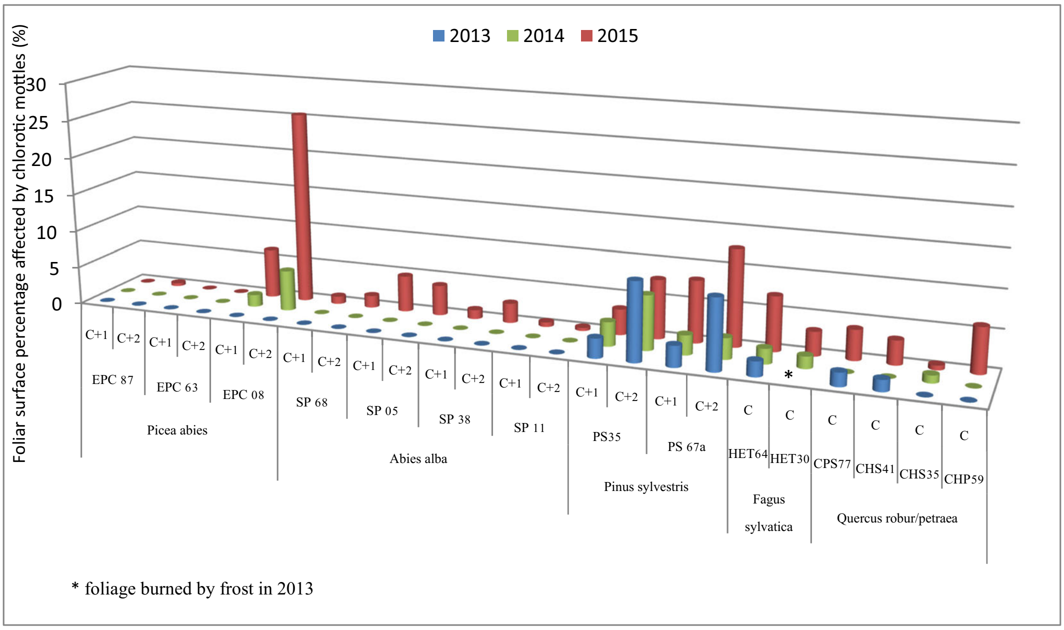


Fig. 2 Average percentage of foliar symptoms on 1-year-old (C + 1) and 2-year-old (C + 2) needles of conifers plots and on current year (C) leaves of hardwood plots between 2013 and 2015

(Vingarzan 2004) (Fig. 8). In urban zones, there is more destruction of ozone by primary pollutants such as NOx and COV, because these are more concentrated (Sicard et al. 2016).

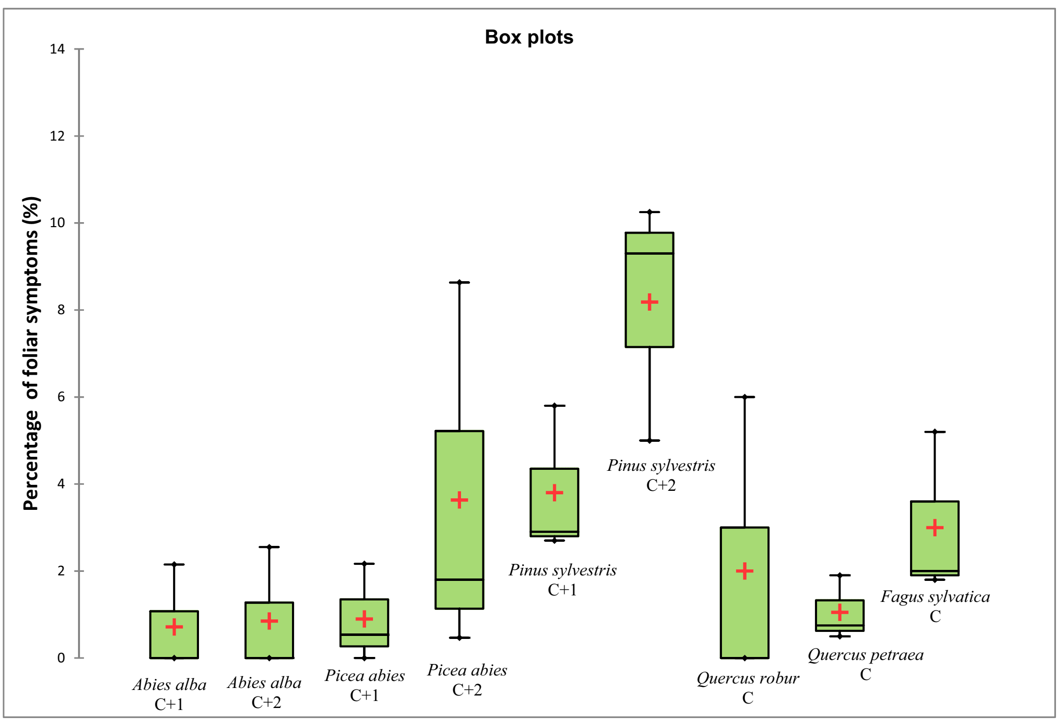


Fig. 3 Tukey box plots of the average percentage of foliar symptoms on 1-year-old (C + 1) and 2-year-old (C + 2) needles of each studied conifer species and on current year (C) leaves of each studied hardwood species throughout the national territory from 2013 to 2015

Fig. 4 Pictures of ozone symptoms. **a** Mottling on the needle of *Pinus sylvestris* observed in PS35 intensive monitoring plot in 2013. **b** Interveinal, upper surface bronzing, and light brown stippling on *Fagus sylvatica* observed in the LESS of EPC 08 in 2014. **c** Upper surface light bronzing on *Quercus petraea* observed in CHS35 intensive monitoring plot in 2014

The average ozone concentration measured by every forest monitoring stations increased during 2015 (74.3 μg m^3), in comparison to the levels recorded during 2013 and 2014 (67 μg m^3); to increased by 10% (Table 4). Year 2015 was the only year considered in this study, when damages appeared on fir trees which are very little sensitive to ozone. Observing meteorological parameters (Table 4), it is possible to note that during 2015 when recorded specific ozone damages had been the most important, the average temperature was 1.3 °C higher than during 2013 and 2014. Furthermore, the number of days and hours when the temperature exceeded 30 °C was greater (Table 4).

4 Discussion and Conclusion

4.1 Ozone Concentration

Year 2015 was characterized by the World Meteorological Organization (WMO), as a historically warm year at a global level. Considering average temperatures in Europe, 2015 was the warmest year at this point (EEA 2016); the consequence of a series of heatwaves, affecting the continent from May to September 2015, was high tropospheric ozone levels (CAMS 2016). The largest ozone episode in 2015 occurred between the 1st and the 5th of July, over central Europe and northern Italy, and was mainly due to traffic and industrial emissions to a lesser extent (CAMS 2016). The formation of ozone is directly correlated to high temperatures which permit its generation by photochemistry from the precursors VOC or NOX; it requires sunlight for complex photooxidative reactions. Weather conditions during summer heatwave events (sunlight in warm and stagnant high pressure air) permit a greater transformation of traffic emissions to ozone ("Air quality in Europe," 2017 report). Therefore, these extreme average temperatures appear to be at the origin of the higher ozone concentrations recorded in the majority of plots during 2015. This may explain why vegetation was more impacted by ozone during this year.

4.2 Visible Foliar Damages

The sensibility of tree species to this gas was characterized by foliar damages such as diffuse chlorotic mottles and bronzing or photobleaching on the upper top of the leaf more exposed to direct sunlight. Among all the forest plots, *Pinus sylvestris* reacted more to ozone than other studied species and is well known to be very sensitive to this gas (Dalstein-Richier et al. 2005; Sicard et Dalstein-Richier 2015). Sunlight can easily passes through its crown which enhances further visible ozone damages; meanwhile, oaks and beeches usually form closed areas as they have thicker foliage and higher crown. However, in clearings, beeches appeared to be significantly sensitive to ozone.

At national scale, important foliar damages associated to this gas were observed upon most of the studied sites. However, in a few ones (plots EPC 63, EPC 87 and SP11), foliar damages were very low. This fact can be explained by the choice of the sites for this study. Indeed, as it has been described, the

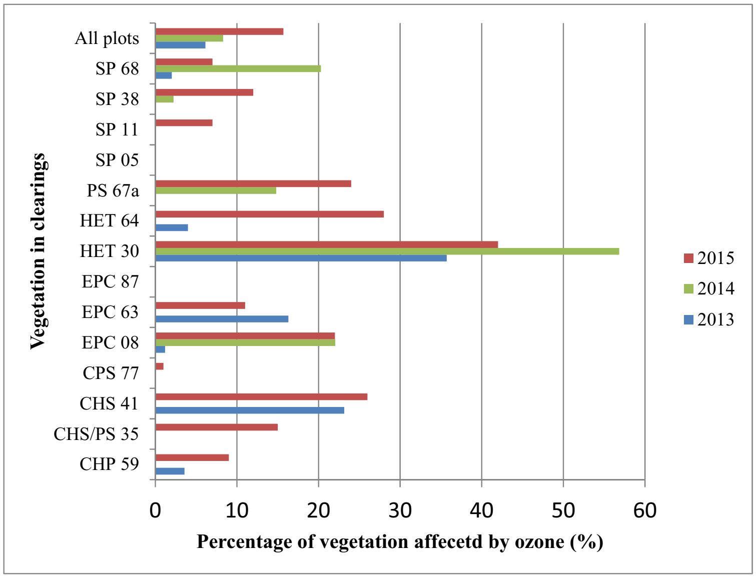


Fig. 5 Average percentage of vegetation (%) affected by ozone in each clearing (LESS) from 2013 to 2015

results depend on the proper conditions of the stations (geographical, topographical, meteorological, and altitudinal conditions) and to the sensitiveness of the chosen species to ozone.

4.3 Defoliation

This study could not establish a correlation between ozone and defoliation. However, average concentrations of this gas, among French territory, remained relatively low between 2013 and 2015 (67 μg m^3 in 2013 and 2014, 74 μg m^3 in 2015). The study made by Sicard and Dalstein-Richier (2015), and carried out in the Alpes-Maritimes Department where ozone concentrations are among the highest of the French territory, could not establish a correlation between this gas and defoliation either. Thus, as ozone concentrations remained quite low over all the plots followed by the National Forestry Office (ONF) in France, this lack of correlation seems to be coherent. The water deficit resulting from climatic conditions is known to be one of the most important causes of defoliation especially in the Mediterranean region. It is therefore possible that soil water content, which reflects the precipitation state of the medium, influences foliar deficit, as it has been shown in several studies (Sicard et al. 2011; Sicard and Dalstein-Richier

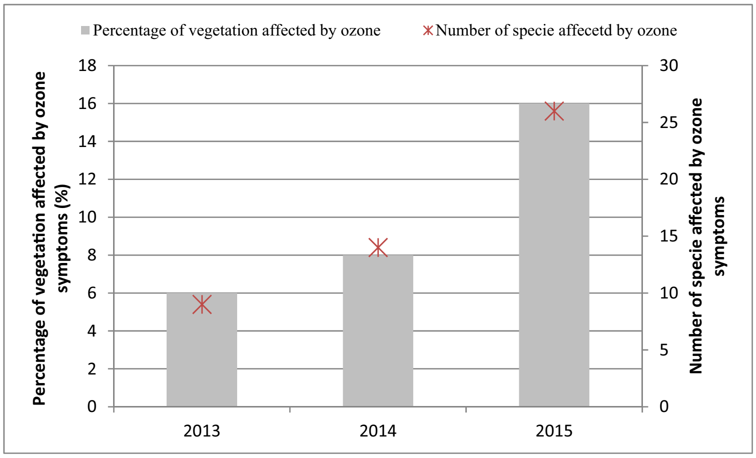


Fig. 6 Percentage of vegetation (%) and number of species of all clearing areas affected by ozone each year from 2013 to 2015

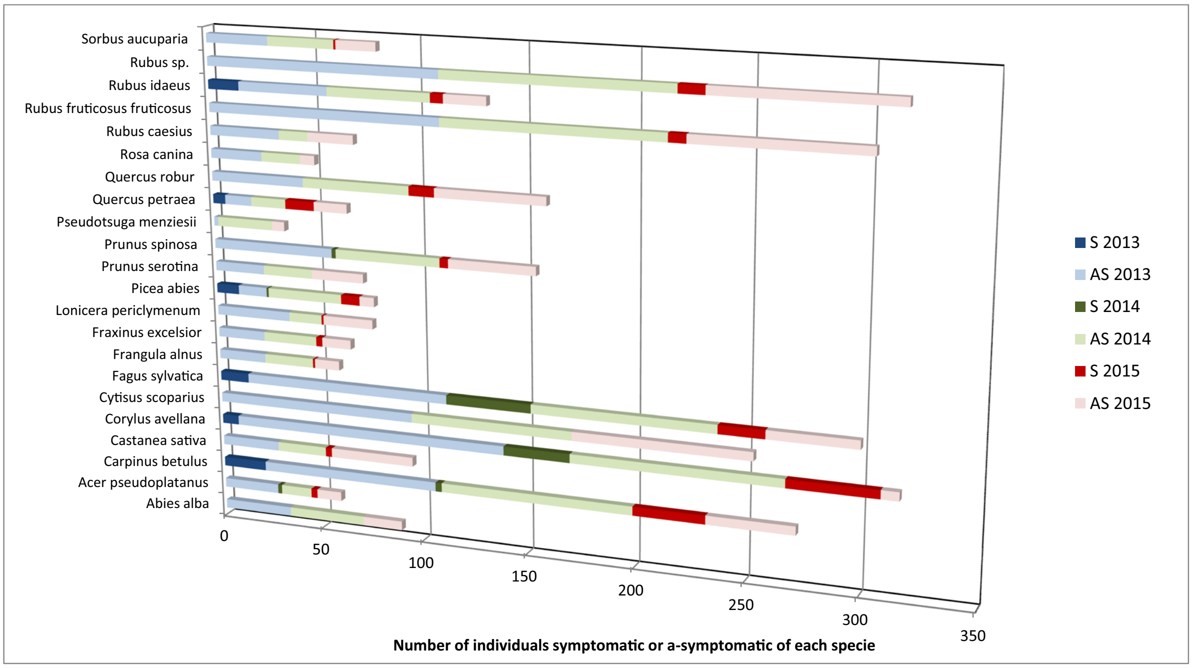


Fig. 7 Number of symptomatic (S) and a-symptomatic (AS) individuals of each species (a species is represented when it is at least observed 20 times a year) on all clearings each year from 2013 to 2015

2015). Unfortunately, data of soil water content were not available for this study.

4.4 Strategy of Adaptation to Climate Change

Climate change will probably increase the number and intensity of extreme events (droughts, heavy rainfalls, heatwaves), and higher ozone concentrations are to be expected. To mitigate the impact on forest which plays an important social, economical, and environmental paper, the following actions/recommendation could be done:

To diversify trees plantations promoting pioneer species (*Rubus* sp.) which are more resistant to ozone.

To set up monitoring on pine plots in the south of France particularly in the region Provence Alpes Côte d'Azur which suffers the highest levels of ozone pollution of the territory because of strong sunshine and the closeness of great conurbations in

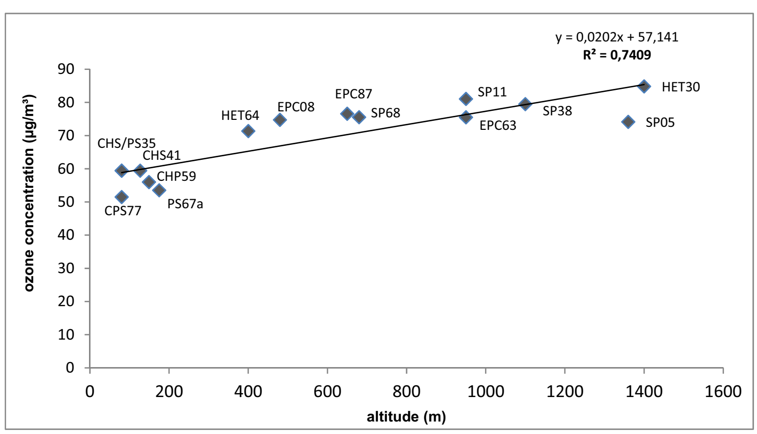


Fig. 8 Trend in average ozone concentrations (µg m^3) from 2013 to 2015 by plot and by increasing altitude gradient

Table 4 Annual average (± standard deviation) of temperature (mean, maximum, minimum), relative humidity (mean, minimum, maximum), annual total of hours when relative humidity is over 90%, number of days and hours when temperature superior at 30 °C, average ozone concentration (April-September), and minimum and maximum ozone concentrations, on all plots from 2013 to 2015

	2013	2014	2015
Cumulative rainfall (mm)	12,449.6 ± 296.1	11,505 ± 320.8	10,747.2 ± 278.1
Mean temperature (°C)	8.9 ± 1.8	10.1 ± 1.8	10.2 ± 1.5
Mean maximum temperature (°C)	13.5 ± 2.1	15.1 ± 2.1	15.2 ± 2.0
Mean minimum temperature (°C)	0.4 ± 0.1	0.4 ± 0.1	0.3 ± 0.0
Number of days (when temperature > 30 °C)	53.0 ± 4.9	21.0 ± 2.3	146.0 ± 8.6
Number of hours where temperature > 30 °C)	229 ± 26.2	93 ± 11.1	667 ± 50.1
Mean relative humidity (%)	83.0 ± 4.4	82.7 ± 3.8	80.1 ± 4.5
Average minimum relative humidity (%)	62.0 ± 6.4	60.2 ± 5.9	57.1 ± 6.3
Average maximum relative humidity (%)	96.2 ± 2.3	96.9 ± 2.2	95.7 ± 3.1
Number of hours where relative humidity > 90% (h)	47,957.2 ± 923	48,837.2 ± 964.2	44,711.11 ± 1022.7
Average ozone concentration (μg m^3)	67.1 ± 11.2	67.1 ± 10.9	74.3 ± 11.8
Minimum ozone concentration (μg m^3)	49.7	47.3	57.4
Maximum ozone concentration (μg m^3)	82.1	78.7	93.8

coastal areas (Nice-Toulon-Marseille), where ozone precursors are produced.

To better focus on parameters (soil water availability, solar radiation in combination to RENECOFOR network's dendrometry measurements) that influence defoliation and biological response of ozone pollution.

In a climate change context, pine tree forests can also be used as indicators of ozone contamination and markers of the evolution of the situation.

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References

Bytnerowicz, A., Fenn, M., McNulty, S., Yuan, F., Pourmokhtarian, A., Driscoll, D., & Meixner, T. (2013). Interactive effects of air pollution and climate change on forest ecosystems in the United States: current understanding and future scenarios. *Developments in Environmental Science, 13*, 333–369.

Bytnerowicz, A., Godzik, B., Grodzinska, K., Fraczek, W., Musselman, R., Manning, W., Badea, O., Popescu, F., & Fleischer, P. (2004). Ambient ozone in forests of the Central and Eastern European mountains. *Environmental Pollution, 130*, 5–16.

CAMS (Copernicus Atmosphere Monitoring Service), 2016. Interim Annual Assessment Report for 2015 European air quality in 2015.

Cristofori, A., Bacaro, G., Confalonieri, Cristofolini, F., Frati, L., Geri, F., Gottardini, E., Tonidandel, G., Zottele, F., & Ferretti, M. (2014). Estimating ozone risks using forest monitoring results for science, policy and society. *Annals of Forest Science, 72*(7), 887–896.

Contran, N., & Paoletti, E. (2007). Visible foliar injury and physiological responses to ozone in Italian provenances of Fraxinus excelsior and F. ornus. *The Scientific World Journal, 7*, 90–97.

Cooper, O. R., Sweeney, C., Gao, R. S., Tarasick, D., & Leblanc, T. (2012). Long-term ozone 656 trends at rural ozone monitoring sites across the United States, 1990–2010. *Journal of Geophysical Research, 657*, 117–D22307.

Dalstein-Richier, L., Mangin, A., Carréga, P., Gueguen, C., Vas, N., Sanchez, O., Théodore, B., & Bérolo, W. (2005). État des forêts d'altitude en relation avec la pollution de l'air par l'ozone dans la région niçoise. *Pollution Atmosphérique, 188*, 503–519.

Dalstein, L., & Vas, N. (2005). Ozone concentrations and ozone-induced symptoms on coastal and alpine Mediterranean pines in southern France. *Water, Air, and Soil Pollution, 160*, 181–195.

Dalstein, L., Vas, N., Ulrich, E., & Cecchini, S. (2004). Première approche de la dégradation de la forêt française en relation avec l'ozone. *Environnement et Technique, 236*, 42–44.

Dalstein, L., Vollenweider, P., Vas, N., & Günthardt-Goerg, M. S. (2002). L'ozone et les conifères du sud-est de la France. *Forêt Méditerranéenne, XXIII*(2), 105–116.

De Marco, A., Proietti, C., Cionni, I., Fischer, R., Screpanti, A., & Vitale, M. (2014). Future impacts of nitrogen deposition and climate change scenarios on forest crown defoliation. *Environmental Pollution, 194*, 171–180.

EEA. (2017). Report No 13/2017: air quality in Europe.

EEA. (2016). Report No 28/2016: air quality in Europe.

Fares, S., Vargas, R., Detto, M., Goldstein, A. H., Karlik, J., Paoletti, E., & Vitale, M. (2013). Tropospheric ozone reduces carbon assimilation in trees: estimates from analysis of continuous flux measurements. *Global Change Biology, 19*, 2427–2443.

Feng, Z., Paoletti, E., Bytnerowicz, A., & Harmens, H. (2015). Ozone and plants. *Environmental Pollution, 202*, 215–216.

Ferretti, M., Sanders, T., Michel, A. (2014). ICP-forest executive report 2014: the impact of nitrogen depositions and ozone on the sustainability of European forests.

Ferretti, M., & Fischer, R. (2013). *Methods for terrestrial investigations in Europe with an overview of North America and Asia*. London: Forest Monitoring.

Gottardini, E., Cristofolini, F., & Ferretti, M. (2017). Foliar symptoms on *Viburnum lantana* reflect annual changes in summer ozone concentration in Trentino (northern Italy). *Ecological Indicators, 78*, 26–30.

Gottardini, E., Calatayud, V., Ferretti, M., Haeni, M., Schaub, M. (2016). Spatial and temporal distribution of ozone symptoms across Europe from 2002 to 2014. Forest condition in Europe: 2016 technical report of ICP forests (editor(s) Michel, A.; Seidling, W.). Eberswalde: ICP Forests: 73-82. ISBN: 9783902762658 handle: http://hdl.handle.net/10449/33085.

Harmens, H., Mills, G., Hayes, F., Sharps, K., Frontasyeva, M., and the participants of the ICP Vegetation (2016). Air pollution and vegetation: ICP Vegetation annual report 2015/2016.

IPCC, 2013. Climate change 2013: the physical science basis.

Innes, J.L., Skelly, J.M., Schaub, M. (2001). Ozone and broadleaved species: a guide to the identification of ozone-induced foliar Injury. Birmensdorf, Eidgenössische Forschungsanstalt WSL, Haupt, Bern, Stuttgart,Wien, 136.

Kampa, M., & Castanas, E. (2008). Human health effects of air pollution. *Environmental Pollution, 151*, 362–367.

Karnosky, D. F., Werner, H., Holopainen, T., Percy, K., Oksanen, T., Oksanen, E., Heerdt, C., Fabian, P., Nagy, J., Heilman, W., Cox, R., Nelson, N., & Matyssek, R. (2007). Free-air exposure systems to scale up ozone research to mature trees. *Plant Biology, 9*, 181–190.

Kulkarni, P. S., Bortoli, D., Domingues, A., & Silva, A. M. (2015). Surface ozone variability and trend over urban and suburban sites in Portugal. *Aerosol Air Quality Research*, 1–15.

Lefohn, A., Malley, C., Simon, H., & Wang, T. (2017). Responses of human health and vegetation exposure metrics to changes in ozone concentration distributions in the European Union, United States, and China. *Atmospheric Environment, 152*, 123–145.

Lehmann, C. M. B., Bowersox, V. C., & Larson, S. M. (2005). Spatial and temporal trends of precipitation chemistry in the United States, 1985-2002. *Environmental Pollution, 135*, 347–361.

Matyssek, R., & Innes, J. L. (1999). Ozone—a risk factor for trees and forests in Europe? *Water, Air and Soil Pollution, 116*, 199–226.

Novak, K., Schaub, M., Fuhrer, J., Skelly, J. M., Hug, C., Landolt, W., Bleuler, P., & Krauchi, N. (2005). Seasonal trends in reduced leaf gas exchange and ozone-induced foliar injury in three ozone sensitive woody plant species. *Environmental Pollution, 136*, 33–45.

Michel, A., Seidling, W., Lorenz, M., Becher, G. (2014). Forest condition in Europe: 2013 technical report of ICP forests. Report under the UNECE Convention on Long-Range Transboundary Air Pollution. *Thünen Working Paper* 19.

Miller, P. R. (1973). Oxydant-induced community change in a mixed conifer forest. *American Chemical Society, 9*, 101–117.

Paoletti, E., Contran, N., Bernasconi, P., Günthardt-Goerg, M. S., & Vollenweider, P. (2009). Structural and physiological responses to ozone in manna ash (Fraxinus ornus L.) leaves in seedlings and mature trees under controlled and ambient conditions. *Science of the Total Environment, 407*, 1631–1643.

Paoletti, E. (2006). Impact of ozone on Mediterranean forest: a review. *Environmental Pollution, 144*, 463–474.

Parmeter, J. R., Bega, R. V., & Neff, T. (1962). A chlorotic decline of ponderosa pine in southern California. *Plant Disease Reporter, 46*, 269–273.

Ponette, Q., Ulrich, I., Collet, GO, Kot, J., Bruno, F., Lanier, M. (1996). Rapport RENECOFOR. Sous-reseau météorologique forestier Bilan de la phase test (1994/1995) et perspectives.

Ruoho-Airola, T., Anttila, P., & Salmi, T. (2004). Airbone sulfur and nitrogen in Finland trends and exposure in relation to air transport sector. *Journal Environmental Monitoring, 6*, 1–11.

Sandermann, H. (1996). Ozone and plant health. *Phytopathology, 34*, 347–366.

Sen, P. K. (1968). Estimates of the regression coefficient based on Kendall's tau. *American Statistical Association, 63*, 1379–1389.

Screpanti, A., & De Marco, A. (2009). Corrosion on cultural heritage buildings in Italy: a role for ozone? *Environmental Pollution, 157*, 1513–1520.

Schaub, M., Calatayud, V., Ferretti, M., Brunialti, G., Lövblad, G., Krause, G., Sanz, M.J. (2010). Monitoring of ozone injury, manual part X. 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, Hamburg.

Schaub, M., Emberson, L., Buker, P., & Krauchi, N. (2007). Preliminary results of modeled ozone uptake for *Fagus sylvatica* L. trees at selected EU/UN-ECE intensive monitoring plots. *Environmental Pollution, 145*, 636–643.

Sicard, P., Serra, R., & Rossello, P. (2016). Spatio temporal trends in ground-level ozone concentrations and metrics in France over the time period 1999–2012. *Environmental Research, 149*, 122–144.

Sicard, P., & Dalstein-Richier, L. (2015). Health and vitality assessment of two common pine species in the context of climate change in southern Europe. *Environmental Research, 137*, 235–245.

Sicard, P., Vas, N., Catalayud, V., Garcia-Breijo, F. J., Reig-Arminana, J., Sanz, M. J., & Dasltein-Richier, L. (2010). Dommages forestiers et pollution à l'ozone dans les réserves naturelles : le cas de l'arolle dans le sud-est de la France. *Forêt méditerranéenne XXXI, 3*, 273–286.

Sicard, P., Dalstein-Richier, L., & Vas, N. (2011). Annual and seasonal trends of ambient ozone concentration and its impacts on forest vegetation in Mercantour National Park (South-eastern France) over the 2000-2008 period. *Environmental Pollution, 159*, 351–362.

Skelly, J. M., Chappelka, A. H., Laurence, J. A., & Fredericksen, T. S. (1997). Ozone and its know and potential effects on forests in Eastern United States. *Ecological Studies, 127*, 69–93.

Stevenson, D. S., Young, P. J., Naik, V., Lamarque, J. F., Shindell, D. T., Voulgaraki, A., Skeie, R. B., Dalsoren, S. B., et al. (2013). Tropospheric ozone changes, radiative forcing and attribution to emissions in the Atmospheric Chemistry and Climate Model Inter-comparison Project (ACCMIP). *Atmospheric Chemical Physics, 13*, 3063–3085.

Tagliaferro, F., Ferrara, A. M., Spaziani, F., & Viotto, E. (2005). Sintomi fogliari di tipo ozone like su vegetazione spontaneae ornamentale in Piemonte. *Linea Ecologica XXVII*, 47–53.

The Royal Society report, (2008). Ground-level ozone in the 21st century: future trends, impacts and policy implications. Science Policy.

Tiwariet, S., Grote, R., Churkina, G., & Butler, T. (2016). Ozone damage, detoxification and the role of isoprenoids—new impetus for integrated models. *Functional Plant Biology, 43*, 324–326.

Ulrich, E., Dalstein, L., Günthardt-Goerg, M.S., Vollenweider, P., Cecchini, S., Vas, N., Sjöberg, K., Skarman, T., Karlsson, G.P. (2006). Effets de l'ozone sur la végétation, concentrations d'ozone (2000-2002) et symptômes d'ozone sur la végétation forestière (2001-2003). Office National des Forêts, Direction Technique, Département Recherche, ISBN 2-84207-300-2**,** 126.

UNECE ICP Forest Programme Co-ordingating Centre (Ed.). (2016). *Manual on methods and criteria for harmonized sampling assessment monitoring and analysis of the effects of air pollution on forests*. Eberswalde: Thünen Institute of Forest Ecosystems.

Vestreng, V., Ntziachristos, L., Semb, A., Reis, S., Isaksen, I. S. A., & Tarrason, L. (2009). Evolution of NOx emissions in Europe with focus on road transport control measures. *Atmospheric Chemical Physics, 9*, 1503–1520.

Vingarzan, R. (2004). A review of surface ozone background levels and trends. *Atmospheric Environment, 38*, 3431–3442.

Vollenweider, P., & Günthardt-Goerg, M. S. (2006). Diagnosis of abiotic and biotic stress factors using the visible symptoms in foliage. *Environmental Pollution, 137*, 455–465.