**DOI: 10.1515/frp-2015-0039**
**Available online: www.lesne-prace-badawcze.pl**

**Leśne Prace Badawcze** / *Forest Research Papers*
December 2015, Vol. 76 (4): 401–409

REVIEW ARTICLE

**e-ISSN 2082-8926**

# The effect of selected gaseous air pollutants on woody plants

**Michał Baciak, Kazimierz Warmiński, Agnieszka Bęś***

University of Warmia and Mazury in Olsztyn, Department of Environmental Toxicology, ul. Prawocheńskiego 17, 10-720 Olsztyn, Poland

* Tel. + 48 89 5233336, e-mail: agnieszka.bes@uwm.edu.pl

**Abstract.** The article discusses gaseous air pollutants that have the greatest impact on forest ecosystems. This group of pollutants ncludes sulfur dioxide ($SO_2$), nitric oxides (NO and $NO_2$) and ozone ($O_3$).

In the 20th century, the major contributor to forest degradation was sulfur dioxide, a gaseous substance with direct and powerful phytotoxic and acidifying effects. Since then, sulfur dioxide emissions have been significantly reduced in Europe and North America, but they continue to grow in East Asia along with China's economic boom. Nitric oxides affect woody plants directly by entering through the stomata and indirectly through soil acidification and environmental eutrophication. Ozone, in turn, is found in photochemical smog and is produced by conversion of its precursors (nitric oxides, organic compounds and carbon monoxide). It is a strong oxidizing agent which disrupts various physiological processes, mostly photosynthesis and water use in plants, but is also the air pollutant that exerts the most toxic effect on forest ecosystems.

**Keywords:** forest degradation, air pollution, ozone, sulfur dioxide, nitric oxides

## 1. Introduction

Forest ecosystems, which cover about one-third of the world's land area (Matyssek et al. 2012), present one of the main parts of biosphere. They affect the composition and the quality of atmosphere and also shape climate conditions both on regional (local) and global scales. The presence of forest ecosystems, which serve as the connection between atmosphere and biosphere, influences cycling of large amounts of matter as well as energy flow in nature (Noe et al. 2011). Forests could be found in almost all climate zones. They are very diverse both in their species composition and in their vertical and horizontal structure (Misson et al. 2007). Owing to the large area covered by forests, they are very sensitive to pollutants present in atmosphere, especially gases (including sulphur dioxide ($SO_2$), nitric oxides ($NO_x$) and tropospheric ozone ($O_3$)). Such pollutants affect forest ecosystems disrupting growth of trees (Wamelink et al. 2009; Liu et al. 2011), changing species composition or increasing susceptibility of forests to biotic factors such as insect outbreaks (Paoletti et al. 2010). Acid gases ($SO_2$ and $NO_x$) indirectly affect forests by acidifying soils, which changes soil fluids and influences accessibility of nutrients (Bytnerowicz et al. 2007; Wu et al. 2011). Currently, around the world, more and more attention is paid to air quality parameters. The research is being conducted in order to limit negative effects of gaseous air pollutants on forest ecosystems. Cross-border movement of air pollutants to large distances presents another significant problem. Owing to such reasons, Japanese forests are influenced by pollutants originating from Eastern Asia (Gbondo-Tugbawa, Driscoll 2002; Chiwa 2010). In Europe during 1970s and 1980s, increased emission of sulphur dioxide, nitric oxides and dust resulted in catastrophic destruction of forests in central and northern parts of the continent together with the so-called 'black triangle' covering the bordering areas of Poland, Czechoslovakia and GDR. Large number of environmental projects were implemented in order to improve air quality in that region as a result of which the situation in the area significantly improved. New forest areas emerged as a consequence of decreased emissions of toxic air polluting compounds (Blažková 1996; Szaro et al. 2002; Bytnerowicz et al. 2004; Maňkovská et

Submitted: 20.01.2015, reviewed: 01.02.2015, accepted after revision: 19.05.2015.

al. 2004; Muzika et al. 2004; Puig et al. 2008; Solberg et al. 2009; Neirynck et al. 2011).

## 2. Sulphur dioxide ($SO_2$)

For several decades, emission of sulphur dioxide and deposition of acids has been presenting a significant problem to forest areas around the world. The influence of $SO_2$ is the most famous example of direct phytotoxic impact on plants (Warmiński et al. 2005; Bytnerowicz et al. 2007). Similar to other gases, $SO_2$ belongs to the group of abiotic stress factors that cause decline in biomass growth (Noe et al. 2011). In forest ecosystems, sulphur dioxide can be deposited as dry or wet matter directly affecting assimilation system and indirectly through acidification of soil. Dry deposition depends on plant species, their leaf structure and wax chemical composition on their surface as well as atmospheric conditions. Coniferous forests more effectively intercept dry deposition in comparison to other plant cover (Derome et al. 2004). While wet deposition results from dissolving of sulphur dioxide in water. That leads to indirect influence of $SO_2$ through acidification of soils, which is revealed through decreased pH, increased concentration of available forms of aluminium and heavy metals as well as damage to mycorrhizal roots (Gbondo-Tugbawa, Driscoll 2002). Direct influence of sulphur dioxide occurs by penetration of gas particles through stomata to leaf interior. The speed of penetration through stomata depends on environmental conditions such as solar irradiance, humidity and temperature. After penetrating leaf cells, sulphur dioxide is oxidised to sulphites, which results in decreased leaf pH and disturbance of oxidation–reduction balancing in plant tissue. That causes loss of chlorophyll, disruption of photosynthesis process at the enzymatic level, of electron transportation and as a result in the decreased assimilation of $CO_2$ (Jim, Chen 2008; Sha et al. 2010). Biochemical response to elevated $SO_2$ levels also includes changes in secondary metabolites levels including phenolic compounds (Giertych, Karolewski 1993). Symptoms of $SO_2$ injury in broadleaf trees include interveinal necrosis indicated by changes in colour from white to black as well as multiple chlorosis (light green spots) on mature leaves. While on needles of conifers sulphur dioxide injury could be seen as light to dark brown necrosis usually having a very clear borderline with the healthy part of a needle (Karolewski 1992; Flagler 1998; Warmiński et al. 2005). In the end, leaves get damaged and fall off prematurely (Białobok 1989; Legge, Krupa 2004).

In the areas with high density of industrial factories, total sulphur content in the needles of Scots pine could reach from 0.6% to 1.0%, while in the areas with no air pollution, sulphur content in pine needles varies between 0.03% and 0.12% (Grodzińska 1977). In the areas affected by highly urbanised and industrialised places, high concentration of sulphur dioxide causes damages to forest vegetation. Multiple research studies of different authors revealed general reduction in tree growth observed through the decrease in stem diameter and height as well as morphological damage of needles and leaves (Gadzikowski 1980; Kawecka 1981; Karolewski 1992; Miś 1995; Dincer et al. 2003; Malik et al. 2011). At the same time, layers of a forest cause variations in environmental parameters, such as wind speed, light availability or temperature. That hampers mixture of air layers above and below tree crowns. As a consequence, different concentrations of toxic gas substances could be observed in different forest layers (Noe et al. 2011).

## 3. Nitric oxides ($NO_x$)

Toxicity of nitric oxides is most likely related to, as it is shown by Woźny (2004), inefficiency of nitrate ($NO_3^-$) reductive pathway. Disruption of cell metabolism occurs as an effect of lower pH of cytoplasm and disruptions in ion transportation. Nitric acid III (nitrous acid) and nitric acid V, which are formed from absorbed nitric oxide as the result of chemical and biochemical reactions (Hu, Sun 2010), can damage biological membranes and chloroplasts as well as cause chlorophyll degradation. Indirect influence of nitric oxides includes soil and water acidification by those compounds, which as a result of plant nutrient uptake infiltrate plant interior causing various types of damage. Favourable sunlight conditions cause wider opening of leaf stomata, which increases intensity of nitric oxides penetration into leaf interior. This process intensifies in the conditions of high humidity (Bobbink, Lamers 2004). It would be important to remember that nitrogen, as a biologically important chemical element, can have different effects on forest ecosystems. From one side, its presence can increase photosynthetic efficiency and, therefore, boost forest biomass growth. From the other side, high nitrogen deposition reduces respiration rate, accelerates nitrogen saturation in areas which already have high amounts of nitrogen in soil as well as decreases deposition of nitrogen on small roots. As a result of the above processes, leaching of nutrients and decline of net production of forest ecosystems can occur (Litton et al. 2007; Liu et al. 2011; Wei et al. 2012). It is estimated that dry deposition presents a significant part of the total nitrogen deposition (Wu et al. 2011). Deposition loads of nitrogen compounds in forests are usually higher than that in other types of ecosystems. It is related to higher aerodynamic coarseness of forest areas and their ability to intercept small particles. Increased nitrogen deposition in forests results in acidification and eutrophication, causing changes in biodiversity of forest ecosystems (Simpson et al. 2006). High nitrogen deposition can also lead to increased susceptibility to late spring and early autumn frosts (Matyssek et al. 2012). The presence of nitric oxides and soil nitrogen compounds is linked to the process of carbon sequestration, which is especially important in case of forest ecosystems. The research conducted in European and North American boreal and temperate forests allowed to state a key positive role of nitrogen deposition in carbon sequestration, which is expressed in increased net primary production (Wamelink et al. 2009; Wei et al. 2012). Nitrogen deposition in forests is also affected by a tree canopy structure. Throughfall, or the process of transporting deposition through tree crowns, as well as nitrogen stemflow, or the flow of those compounds from leaves, branches and further down the trunk to soil, could increase the speed of deposition of nitrogen compounds in soil. However, that could take place in areas with higher dry and wet nitrogen deposition. Both these processes depend on leaf shape and texture, crown density and shape, type of precipitation, its intensity and length (Pelster et al. 2009).

Symptoms of leaf damage include irregular brown to cream stains on a leaf blade between veins which eventually change into white necrosis. High amounts of nitric oxides result in appearance of rusty-reddish necrosis on needles or needle discoloration (Flagler 1998; Karolewski 1998; Greszta et al. 2002).

## 4. Ozone ($O_3$)

Tropospheric ozone, because of its strong oxidation capabilities, can have a serious negative effects on forest ecosystems. Currently, compared to other air pollutants, ozone presents the biggest threat to plant biomass production, also including that in forests (Schaub et al. 2005; Zapletal et al. 2011). High ozone concentration (above 40 ppb) can cause decreased ability of forests to absorb carbon dioxide (Karlsson et al. 2006). Ozone belongs to the group of secondary air pollutants. It is formed as a result of photochemical reactions involving initial substances such as nitric oxides and volatile organic compounds (VOC) under favourable atmospheric conditions (high temperature and insolation). The majority of European countries as well as the United States considerably limited the emissions of ozone precursors. However, it did not completely eliminate potential dangers of ozone. High industrialisation of India and China contributes to constant supply of precursors that can travel long distances across the borders and cause ozone formation in different places of the globe as well as harm forest ecosystems (Cape 2008).

Numerous research projects are conducted around the world to study the phytotoxicity of ozone. It would be important to underline that in Poland, research on influence of gaseous air pollutants, which includes effect of ozone on tree plants, was started relatively early, that is, in 1970s (a.o. Białobok, Karolewski 1978; Karolewski, Białobok 1979). At that time, they demonstrated a link between ozone exposure and appearance of damage on Scots pine (*Pinus sylvestris* L.) and European larch (*Larix decidua* L.) assimilation organs.

Increased concentrations of ozone in European countries as well as the United States have different effects on forest ecosystems. At the physiological level, trees damaged by ozone had a decline in chlorophyll content as well as more rapid ageing of leaves and needles. Other physiological effects include increased respiration rate, lower photosynthesis rate, disruptions of carbon assimilation and transportation of water and nutrients between roots and branches, which cause overall decline of tree biomass growth (Tjoelker et al. 1995; Karnosky et al. 2007; Augustaitis, Bytnerowicz 2008; Malik et al. 2011; Zapletal et al. 2011). Trees growing in the areas with high ozone pollution showed lower increment of tree trunk compared to trees growing in less polluted places (Weinstein et al. 2005).

The reaction of ozone with cell structures produces reactive oxygen species ($H_2O_2$, $O_2^{\bullet-}$, $HO^{\bullet}$, $^1O_2^{\bullet}$), which cause denaturation of membrane lipids and their peroxidation. This process could be described by the oxidation state (Hunová et al. 2010). Morphological changes are revealed through appearing chlorotic stains on the surface of leaves and needles, which quickly change into necrosis. With time, such stains cover the whole area of the leaf, which indicates its complete dying (Ashmore 2004; Hayes et al. 2007). Deformation of Scots pine (*P. sylvestris* L.) needles along with their abnormal growth under the influence of ozone was observed by Białobok et al. (1980).

Besides infiltration of ozone through stomatal pores in case of tree plants, it could also be absorbed by lenticels, which are located within bark of well-developed branches. In such cases, ozone damage occurs similarly to that in leaf cells (Wittmann et al. 2007).

Phytotoxicity of ozone is significantly affected by meteorological conditions. They include high air humidity, strong sunlight as well as soil moisture. During the day and whilst having optimal soil moisture content, stomata apertures are open, which results in increased gas intake, also including ozone, into leaves (Karlsson et al. 2007). Besides that, ozone hinders closing functions of stomatal pores, which in turn results in over intensive losses of water during transpiration (Wagg et al. 2012). Implemented experiments allowed to conclude that increased watering intensified ozone infiltration process through stomatal apertures, which induced visual damage in plants (Karlsson et al. 2007).

Intensity of phytotoxic responses of tree plants depends on the amount of ozone absorbed directly through stomatal apertures; however, this parameter is not suitable for regular monitoring measurements. To avoid the above problem, various exposure indices are used. The most known and commonly used index is AOT40. It describes ozone accumulated over a threshold 40 ppb (80 $\mu g \cdot m^{-3}$). It is recognised that above this level, ozone is toxic for plants, whilst its toxicity grows proportionally to exposure time. This is true for daylight time, when the most intensive gas exchange occurs through stomatal pores. Therefore, in order to measure AOT40, differences between measured ozone concentration (mean hourly) and threshold value are summed. Calculations are done during the peak of plant growing season, taking into account daylight measurements and only concentrations above 40 ppb. Day length and growing season depend on geographical location, whilst day length additionally depends on season. So whilst doing ozone measurement, they should be clearly defined. During AOT40 calculations for the purposes of forest protection, growing season is assumed to be equal to 6 months (from April 1 to September 30), and for the purposes of protection of cultivated plants, the growing season used is from May 1 to July 31. The daylight time is the period when insolation is above 50 $W \cdot m^{-2}$ or more simply the time from 8 in the morning to 8 in the evening of Central European Time. For legislative purposes, the second definition is used (Gerosa et al. 2007; Dyrektywa 2008/50/WE).

AOT40 value is easily estimated based on the continuous ozone measurements done in air quality monitoring stations using automatic ozone monitors (Sicard et al. 2011). However, more and more often AOT40 deficiencies are pointed out in forecasting negative effects of ozone on plants (Fares et al. 2010). It is also possible to estimate AOT40 with the help of models based on ozone concentration measurements, which uses weekly measurements (1–2 weeks) obtained from passive samplers. Such method is less expensive than automatic measurements (Gerosa et al. 2007). That is also why in many countries AOT40, in spite of its weaknesses, is used in legislature. Table 1 presents AOT40 values received during the studies of Gerosa et al. (2008), Marzuoli et al. (2008), Gerosa et al. (2009) and Calatayud et al. (2011) using various broadleaf tree species in open-top chambers (OTC). During those studies, plant responses to elevated ozone levels in different humidity conditions were described. The table presents AOT40 levels at which first symptoms of leaf damage caused by ozone were observed. Mentioned studies indicated that trees growing in humid conditions responded quicker to elevated ozone concentrations than trees growing in dry conditions. Pyrenean oak *(Quercus pyrenaica)* appeared to be the most susceptible to ozone damage.

**Table 1.** Selected tree species and AOT40 values in OTC research

| Tree species | Year | AOT 40 [ppb · h] | Source |
|---|---|---|---|
| *Quercus faginea* Portuguese oak | 2006 | 28 223 | Calatayud V. et al. (2011) |
| | 2007 | 26 181 | |
| *Quercus pyrenaica* Pyrenean oak | 2006 | 2883 | |
| | 2007 | 3720 | |
| *Quercus robur* English oak | 2006 | 26 181 | |
| | 2007 | 23 367 | |
| *Populus nigra* Black poplar | 2004 | 10 227 (a)<br>11 719 (b) | Marzuoli et al. (2008) |
| | 2005 | 9541 (a)<br>13 184 (b) | |
| *Fagus sylvatica* European beech | 2004 | 23 340 (a)<br>27 552 (b) | Gerosa G. et al. (2008) |
| | 2005 | 18 793 (a)<br>24 446 (b) | |
| *Fraxinus excelsior* European ash | 2004 | 23 340 (a)<br>27 552 (a) | Gerosa G. et al. (2009) |
| | 2005 | 14 829 (a)<br>22 703 (b) | |

Explanations: a – wet conditions, b – dry conditions

## 5. Biological indicators of environmental damage

Plant organisms, which are sensitive to the presence of damaging compounds in atmosphere, are called bio-indicators (Greszta et al. 2002). Majority of biological monitoring methods are based on such organisms, which indicate the presence of pollutants in the natural environment. Bio-indicator shows the presence of pollutants in atmosphere by the appearance of typical symptoms, which are not characteristic in case of other changes occurring in the environment. The organism could react with a specific response in case of one or several pollutants. Plants, which are used for indication of pollution, should be wide spread for a given environment, well studied from a biological and ecological point of view and also easily collected and marked (Greszta et al. 2002; Wesołowski, Radecka 2003). Classifications of organisms as bio-indicators could vary. Whilst describing changes occurring in the environment and the degree of their change, bio-indicators are divided into qualitative, quantitative and mixed. Taking into account sensitivity of plants to pollution, they could be used as indicator species, monitors or test plants (Falińska 1997; Alloway, Ayres 1999; Greszta et al. 2002; Wesołowski, Radecka 2003; Merkert et al. 2012).

Amongst vascular plants, broadleaf trees can also play the role of biological indicators, including European beech (*Fagus sylvatica),* pedunculate oak (*Quercus robur),* black poplar (*Populus nigra),* European ash (*Fraxinus excelsior),* Norway maple *(Acer platanoides),* and sycamore maple (*Acer pseudoplatanus* L.*)*, as well as coniferous: Scots pine

(*P. sylvestris),* Norway spruce (*Picea abies),* European larch (*L. decidua)* and Weymouth pine (*Pinus strobus*).

The German Federal Environmental Specimen Bank (ESB) stores the following tree species: Norway spruce (*P. abies*), Scots pine (*P. sylvestris),* European beech (*F. sylvatica)* and black poplar (*P. nigra* Italica). In Poland, there is no official environmental specimen bank; however, for many years, environmental samples are collected for the purposes of environmental monitoring. In our country, many polluted areas, such as Górny Śląsk or Legnicko-Głogowski Copper Region, were evaluated based on the indicator species. As a standard species recommended by the ESB for the research of such land ecosystems in Europe, Scots pine could be used. Pine is a main forest species in Poland. It also implements most of the requirements stated by the ESB. More rarely, Norway spruce is used as bio-indicator because of the fact that borderline of its distribution is located in Poland and also spruce is very sensitive to pollution (Jaszczak 2003; Merkert et al. 2012).

## 6. Summary

In addition to natural biotic and abiotic factors, contemporary forests are also affected by anthropogenic factors, which include besides others air pollution by phytotoxic gases and dust. Gaseous air pollution in the first place affects tree plants by decreasing their biomass growth and, what is more significant, by reducing their resistance to biotic and abiotic stress factors. The reports of the European Environment Agency (EEA 2015) present up-to-date data on atmospheric air pollution levels by phytotoxic gases. Amongst the most important phytotoxic gases found in the atmospheric air are sulphur dioxide ($SO_2$), nitric oxides (NO and $NO_2$) as well as ozone ($O_3$). Limit values are set for those gaseous pollutants for rural areas in consideration with vegetation protection (Table 2).

Gaseous air pollution presents a significant problem at a global scale. The presence of phytotoxic gases in the air worsens growth conditions for forest ecosystems. Many research studies and also field work pay special attention to the response of plants to specific air pollutants. From one side, familiarity with symptoms and knowledge of damage mechanisms help to implement activities that are targeted at abatement of damaging impact of toxic substances on plants. From the other side, specific responses of plant species to pollution allow the use and implementation of monitoring activities. Monitoring research, which is also called bio-indication, enables conducting relatively inexpensive and quick environmental analysis. Owing to that, protection of environment becomes more effective and preventive measures could be undertaken more promptly.

Sulphur and nitric oxides are primary air pollutants, which are emitted mainly from the anthropogenic sources. The sources of ozone in atmosphere vary, as ozone is not directly emitted from any sources, but it is formed as a result of photosynthetic reactions of its precursors (nitrogen oxides, organic compounds and carbon dioxide).

Sulphur dioxide and nitric oxides are the gases that enter the plants through their stomatal apertures, and directly affecting physiological processes in leaves, mainly $CO_2$ assimilation and photosynthesis. Besides that, they are acid gases, which after dissolving in water from atmospheric precipitation engage in acidification of the environment.

In the 20th century, sulphur dioxide played the biggest role in forest degradation, whilst currently ozone took its place. Ozone is not an acid gas, whilst it has very strong oxidation potential. Similar to other gases, ozone enters plants through their stomatal pores. It quickly damages cells lying in close proximity to stomatal openings and later affects tissues responsible for photosynthesis. Besides its direct influence on the process of photosynthesis, ozone also causes disruptions to plant's vascular system because of damages caused to stomatal pores, which result in the decrease of biomass growth and resistance of trees. It is currently acknowledged that amongst air pollutants, ozone presents the most significant danger for forest ecosystems. Owing to such reasons in the EU countries, ozone concentration limits in the interest of plant protection are being gradually decreased. If before 2010 in Poland, the value acceptable for ozone level expressed by the AOT40 index was 24,000 $\mu g \cdot m^{-3} \cdot h$, after January 1, 2010, it was decreased to 18,000 $\mu g \cdot m^{-3} \cdot h$, whilst the long-term AOT40 goal, which should be achieved by 2020, is equal to only 6,000 $\mu g \cdot m^{-3} \cdot h$ (Table 2).

## Conflict of interests

Authors declare the absence of potential conflicts.

## Acknowledgements and financial support

The research was implemented within the framework of the statute project of the Environmental Toxicology Department 1019.0834 – Bio-indicators in evaluation of environmental pollution.

## References

Alloway B.J., Ayres D.C. 1999. Chemiczne podstawy zanieczyszczenia środowiska. PWN, Warszawa. ISBN 83-01-12947-6.

Ashmore M.R. 2004. Wpływ utleniaczy na poziomie organizmu i zbiorowiska roślinnego, in: Zanieczyszczenia powietrza a życie

**Table 2.** Limit values of gaseous pollutants in ambient air for vegetation protection in Poland

| Substance | Parameter | Value | Unit of measure | Period over which measurements are averaged | Date for meeting the target valuein ambient air |
|---|---|---|---|---|---|
| $SO_2$ | limit value | 20 | $\mu g \cdot m^{-3}$ | calendar year and winter season (from 1 X to 31 III) | 2003 r. |
| $NO_x$ [a)] | limit value | 30 | $\mu g \cdot m^{-3}$ | calendar year | 2003 r. |
| $O_3$ | target value (as AOT40) [b)] | 18 000[c)] | $\mu g \cdot m^{-3} \cdot h$ | from 1V to 31 VII | 2010 r. |
| | long-term objective (as AOT40) [b)] | 6 000 | $\mu g \cdot m^{-3} \cdot h$ | from 1V to 31 VII | 2020 r. |

Source: the above limit values were set based on the Regulation of the Minister of Environment of 24 August 2012 on the concentrations of selected substances in ambient air (Journal of Laws, 2012, item 1031). The regulation implements the provisions of Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe (OJ L 152/1 of 11 June 2008).
Key for the table:
a) – Total nitrogen dioxide and nitrogen oxide expressed in units of mass concentration of nitrogen dioxide;
b) – Expressed as AOT40, which is the sum of the differences greater than 80 $\mu g\ m^{-3}$ between mean hourly concentrations expressed in $\mu g \cdot m^{-3}$ and the value of 80 $\mu g \cdot m^{-3}$, calculated daily every hour between 8:00 and 20:00 Central European Time (CET); if continuous data are not available in a series of measurements, the value of AOT40 is multiplied by the quotient of the number of possible measurement dates and the number of measurements performed in that period.
c) – Until 31 December 2009, the AOT40 limit was set at 24,000 $\mu g \cdot m^{-3} \cdot h$ (based on the now repealed Regulation of the Minister of Environment of 6 June 2002 on concentration limits of selected substances in ambient air, alert thresholds for selected substances in ambient air and margins of tolerance for selected substances, Journal of Laws 2002, No. 87, item 796).

roślin (eds. J.N.B. Bell, M. Treshow). Wydawnictwo Naukowo -Techniczne. Warszawa. 101–132. ISBN 83-204-2947-1.

Augustaitis A., Bytnerowicz A. 2008. Contribution of ambient ozone to Scots pine defoliation and reduced growth in the Central European forests: A Lithuanian case study. *Environmental Pollution* 155: 436–445. DOI:10.1016/j.envpol.2008.01.042.

Białobok S. 1998. Wpływ kwaśnych opadów atmosferycznych na drzewa i lasy, in: Życie drzew w skażonym środowisku (ed. S. Białobok). Wyd. Polska Akademia Nauk, Instytut Dendrologii, 169–193. ISBN: 83-01-07352-7.

Białobok S., Karolewski P. 1978. Ocena stopnia odporności drzew matecznych sosny zwyczajnej i ich potomstwa na działanie $SO_2$, $O_3$ oraz mieszaniny tych gazów. *Arboretum Kórnickie* 23: 299–310.

Białobok S., Karolewski P., Oleksyn J.1980. Sensitivity of Scots pine needles from mother trees and their progenies to the action of $SO_2$, $O_3$, a mixture of the gases, $NO_2$ and HF. Arboretum Kórnickie 25: 289–303.

Blažková M. 1996. Black Triangle - The Most Polluted Part of Central Europe, in: Regional Approaches to Water Pollution in the Environment (ed. P.E. Rijtema, V. Eliáš). Springer Netherlands, 227–249. DOl: 10.1007/978-94-009-0345-6.

Bobbink R., Lamers L.M.P. 2004. Skutki wzrostu depozycji azotu, in: Zanieczyszczenia powietrza a życie roślin. (eds. J.N.B. Bell, M. Treshow). Wydawnictwo Naukowo-Techniczne, Warszawa, 345–383. ISBN 83-204-2947-1.

Bytnerowicz A., Godzik B.,. Grodzińska K, Frączek 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. DOI:10.1016/j.envpol.2003.10.032.

Bytnerowicz A., Omasa K., Paoletti E. 2007. Integrated effects of air pollution and climate change on forests: A northern hemisphere perspective. *Environmental Pollution* 147: 438–445. DOI:10.1016/j.envpol.2006.08.028.

Calatayud V., Cerveró J., Calvo E., García-Breijo F.-J., Reig-Armiñana J., Sanz M.J. 2011. Responses of evergreen and deciduous *Quercus* species to enhanced ozone levels. *Environmental Pollution* 159: 55–63. DOI:10.1016/j.envpol.2010.09.024.

Cape J.N. 2008. Interactions of forests with secondary air pollutants: Some challenges for future research. *Environmental Pollution* 155: 391–397. DOI:10.1016/j.envpol.2008.01.038.

Chiwa M. 2010. Characteristics of atmospheric nitrogen and sulfur containing compounds in an inland suburban-forested site in northern Kyushu, western Japan. *Atmospheric Research* 96: 531–543. DOI: 10.1007/s11270-012-1294-2

Derome J., Nieminen T., Saarsalmi A. 2004. Sulphur dioxide adsorption in Scots pine canopies exposed to high ammonia emissions near the Cu-Ni smelter in south-western Finland. *Environmental Pollution* 129: 79–88. DOI:10.1016/j.envpol.2003.09.021.

Dincer F., Muezzinoglu A., Elber T. 2003. $SO_2$ levels at forested mountains Izmir, Turkey and their possible source. *Water, Air, and Soil Pollution* 147: 331–341. DOI: 10.1023/A:1024581531855.

Dyrektywa Parlamentu Europejskiego i Rady 2008/50/WE z dnia 21 maja 2008 r. w sprawie jakości powietrza i czystszego powietrza dla Europy (Dz. Urz. UE, 11.6.2008, L 152/1).

EEA 2015. Środowisko Europy 2015 – Stan i prognozy: Synteza. Europejska Agencja Środowiska, Kopenhaga. DOI: 10.2800/95535.

EEA 2015. Projections in hindsight. An assessment of past emission projections reported by Member States under EU air pollution and GHG legislation. Technical report No 4. DOI: 10.2800/894788.

Falińska K. 1997. Ekologia roślin: podstawy teoretyczne, populacje, zbiorowisko. PWN, Warszawa. ISBN: 83-01-12065-7.

Fares S., McKay M., Holzinger R., Goldstein A.H., 2010. Ozone fluxes a *Pinus ponderosa* ecosystem are dominated by non-stomatal processes: Evidence from long-term continuous measurements. *Agricultural and Forest Meteorology* 150: 420–431. DOI:10.1016/j.agrformet.2010.01.007.

Flagler R.B. 1998. Recognition of Air Pollution Injury to Vegetation: A Pictorial Atlas, Second Edition, Air & Waste Management Association. ISBN 0-923204-14-8.

Gadzikowski R. 1980. Oddziaływanie Zakładów Azotowych w Puławach na środowisko leśne w latach 1967–1978. *Sylwan* 5: 17–29. ISSN: 0039-7660.

Gbondo-Tugbawa S.S., Driscoll Ch.T. 2002. Evaluation of the effects of future emission control of sulfur dioxide and nitrogen oxides in acid-base status of the northern forest ecosystem. *Atmospheric Environment* 36: 1631–1643. DOI:10.1016/S1352-2310(02)00082-1.

Gerosa G., Ferretti M., Bussotti F., Rocchini D. 2007. Estimates of ozone AOT40 from passive sampling in forest sites in South -Western Europe. *Environmental Pollution* 145(3): 629–635. DOI:10.1016/j.envpol.2006.02.030.

Gerosa G., Marzuoli, R., Desotgiu R., Bussotti F., Ballarin-Denti A., 2008. Visible leaf injury in young trees of *Fagus sylvatica* L. and *Quercus robur* L. in relation to ozone uptake and ozone exposure. An Open-Top Chambers experiment in South Alpine environmental conditions. *Environmental Pollution* 152: 274–284. DOI:10.1016/j.envpol.2007.06.045.

Gerosa G., Marzuoli R., Desotgiu R., Bussotti F., Ballarin-Denti A. 2009. Validation of the stomatal flux approach for the assessment of ozone visible injury in young forest trees. Results from the TOP (transboundary ozone pollution) experiment at Curno, Italy. *Environmental Pollution* 157: 1497–1505. DOI:10.1016/j.envpol.2008.09.042.

Giertych M.J., Karolewski P. 1993. Changes in phenolic compounds content in needles of Scots pine (*Pinus sylvestris* L.) seedlings following short term exposition to sulphur dioxide. *Arboretum Kórnickie* 38: 43–52.

Greszta J., Gruszka A., Kowalkowska M. 2002. Wpływ immisji na ekosystem. Wydawnictwo Naukowe Śląsk, Katowice. ISBN 83-7164-369-1.

Grodzińska K. 1977. Acidity of tree bark as a bioindicator of forest pollution in southern. *Poland. Water, Air and Soil Pollution* 8: 3–7.

Hayes F., Mills G., Harmens H., Norris D. 2007. Evidence of widespread ozone damage to vegetation in Europe (1990–2006). *Centre for Ecology and Hydrology*. http://icpvegetation.ceh.ac.uk/publications/documents/EvidenceReportFINALPRINTEDVERSIONlow-res.pdf [17.01.2015].

Hu Y., Sun G. 2010. Leaf nitrogen dioxide uptake coupling apoplastic chemistry, carbon/sulfur assimilation, and plant nitrogen status. *Plant Cell Reports* 29: 1069–1077. DOI 10.1007/s00299-010-0898-5.

Hunová I., Novotný R., Uhlírová H., Vráblík T., Horálek J., Lomský B., Srámek V. 2010. The impact of ambient ozone on mountain spruce forests in the Czech Republic as indicated by malondialdehyde. *Environmental Pollution* 158: 2393–2401. DOI:10.1016/j.envpol.2010.04.006.

Jaszczak R. 2003. Wpływ zanieczyszczeń z Legnicko-Głogowskiego Okręgu Miedziowego na stan koron sosny zwyczajnej (*Pinus sylvestris* L.) w Nadleśnictwach Góra Śląska i Włoszakowice. *Sylwan* 9: 10–26. ISSN: 0039-7660.

Jim C.Y., Chen W.Y. 2008. Assessing the ecosystem service of air pollutant removal by urban trees in Guangzhou (China). *Journal of Environmental Management* 88: 665–676. DOI:10.1016/j.jenvman.2007.03.035.

Karlsson P.E., Braun S., Broadmeadow M., Elvira S., Emberson L., Gimeno B.S., Le Thiec D., Novak K., Oksanen E., Schaub M., Uddling J., Wilkinson M. 2007. Risk assessments for forest trees: The performance of the ozone flux versus the AOT concepts. *Environmental Pollution* 146: 608–616. DOI:10.1016/j.envpol.2006.06.012.

Karlsson P.E., Hansson M., Höglund H.-O., Pleijel H. 2006. Ozone concentration gradients and wind conditions in Norway spruce (*Picea abies*) forests in Sweden. Atmospheric Environment, 40: 1610–1618. DOI: 10.1016/j.atmosenv.2005.11.009.

Karnosky D.F., Skelly J.M., Percy K.E., Chappelka A.H. 2007. Perspectives regarding 50 years of research on effects of tropospheric ozone air pollution on US forests. *Environmental Pollution* 147: 489–506. DOI:10.1016/j.envpol.2006.08.043.

Karolewski P. 1992. Ocena wrażliwości jedenastu gatunków z rodzaju *Pinus* na działanie $SO_2$, HF, $NO_2$ i $O_3$ w kontrolowanych warunkach. *Arboretum Kórnickie* 37: 75–81.

Karolewski P. S. 1998. Oddziaływanie tlenków azotu na rośliny drzewiaste, in: Życie drzew w skażonym środowisku (ed. S. Białobok). Polska Akademia Nauk, Instytut Dendrologii, 129–141. ISBN: 83-01-07352-7.

Karolewski P., Białobok S. 1979. Wpływ dwutlenku siarki, ozonu, mieszaniny tych gazów i fluorowodoru na uszkodzenie igieł modrzewia europejskiego. *Arboretum Kórnickie* 24: 297–305.

Kawecka A. 1981. Próby kształtowania fitocenoz w warunkach działania emisji związków azotu. *Sylwan* 4: 41–48. ISSN: 0039-7660.

Legge A. H., Krupa S. V. 2004. Wpływ dwutlenku siarki, in: Zanieczyszczenia powietrza a życie roślin (eds. J.N.B. Bell, M. Treshow). Wydawnictwo Naukowo-Techniczne, Warszawa, 151–179. ISBN 83-204-2947-1.

Litton CM, Raich J.W., Ryan M.G. 2007. Carbon allocation in forest ecosystems. *Global Change Biology* 13: 2089–109. DOI: 10.1111/j.1365-2486.2007.01420.x.

Liu X., Duan L., Mo J., Du E., Shen J., Lu X., Zhang Y., Zhou X., He Ch., Zhang F. 2011. Nitrogen deposition and its ecological impact in China: An overview. *Environmental Pollution* 159: 2251–2264. DOI:10.1016/j.envpol.2010.08.002

Malik I., Wistuba M., Danek M., Danek T., Krąpiec M. 2011. Wpływ emisji zanieczyszczeń atmosferycznych przez Zakłady Chemiczne w Tarnowskich Górach (północna część Wyżyny Śląskiej) na szerokość przyrostów rocznych sosny zwyczajnej (*Pinus sylvestris* L.). *Ochrona Środowiska i Zasobów Naturalnych* 47: 9–21. ISSN: 1230-7831-08-7.

Maňkovská B., Godzik B., Badea O., Shparyk Y., Moravčík P. 2004. Chemical and morphological characteristics of key tree species of the Carpathian Mountains. *Environmental Pollution* 130: 41–54. DOI:10.1016/j.envpol.2003.10.020.

Marzuoli, R., Gerosa, G., Desotgiu, R., Bussotti, F., Ballarin-Denti, A. 2008. Ozone fluxes and foliar injury development in the ozone-sensitive poplar clone Oxford: a dose-response analysis. *Tree Physiology* 29(1): 67–76. DOI: 10.1093/treephys/tpn012.

Matyssek R., Wieser G., Calfapietra C., de Vries W., Dizengremel P., Ernst D., Jolivet J., Mikkelsen T.N., Mohren G.M.J., Le Thiec D., Tuovinen J.-P., Weatherall A., Paoletti E. 2012. Forest under climate change and air pollution: Gaps in understanding and future directions for research. *Environmental Pollution* 160: 57–65. DOI:10.1016/j.envpol.2011.07.007.

Merkert B., Wünschmann S., Diatta J., Chudzińska E. 2012. Innowacyjna obserwacja środowiska – bioindykatory i biomonitory: definicje, strategie i zastosowania. *Ochrona Środowiska i Zasobów Naturalnych* 53: 115–152.

Miś R. 1995. Wpływ przemysłowych zanieczyszczeń powietrza na wzrost wysokości i jakość sosny zwyczajnej (*Pinus sylvestris* L.). *Sylwan* 139 (1): 87–97.

Misson L., Baldocchi D.D., Black T.A., Blanken P.D., Brunet Y., Curiel Yuste J., Dorsey J.R., Falk M., Granier A., Irvine M.R., Jarosz N., Lamaud E., Launiainen S., Law B.E., Longdoz B., Loustau D., McKay M., Paw U.K.T., Vesala T., Vickers D., Wilson K.B., Goldstein A.H. 2007. Partitioning forest carbon fluxes with overstory and understory eddy-covariance measurements: a synthesis based on FLUXNET data. *Agricultural and Forest Meteorology* 144: 14–31. DOI:10.1016/j.agrformet.2007.01.006.

Muzika R.M., Guyette R.P., Zielonka T., Liebhold A.M. 2004. The influence of $O_3$, $NO_2$ and $SO_2$ on growth of *Picea abies* and *Fagus sylvatica* in the Carpathian Mountains. *Environmental Pollution* 130: 65–71. DOI:10.1016/j.envpol.2003.10.021.

Neirynck J., Flechard C.R., Fowler D. 2011. Long-term (13 years) measurements of $SO_2$ fluxes over a forest and their control by surface chemistry. *Agricultural and Forest Meteorology* 151: 1768– 1780. DOI:10.1016/j.agrformet.2011.07.013.

Noe S.M., Kimmel V., Hüve K., Copolovici L., Portillo-Estrada M., Ülle Püttsepp Ü., Jõgiste K., Niinemets Ü., Hörtnagl L., Wohlfahrt G. 2011. Ecosystem-scale biosphere–atmosphere interactions of a hemiboreal mixed forest stand at Järvselja, Estonia. *Forest Ecology and Management* 262: 71–81. DOI:10.1016/j.foreco.2010.09.013.

Paoletti E., Schaub M., Matyssek R., Wieser G., Augustaitis A., Bastrup-Birk A.M., Bytnerowicz A., Günthardt-Goerg M.S., Müller-Starck G., Serengil Y. 2010. Advances of air pollution science: From forest decline to multiple-stress effects on forest ecosystem services. *Environmental Pollution* 158: 1986–1989. DOI:10.1016/j.envpol.2009.11.023.

Pelster D.E., Kolka R.K., Prepas E.E. 2009. Overstory vegetation influence nitrogen and dissolved organic carbon flux from the atmosphere to the forest floor: Boreal Plain, Canada. *Forest Ecology and Management* 259: 210–219. DOI:10.1016/j.foreco.2009.10.017.

Puig R., Àvila A., Soler A. 2008. Sulphur isotopes as tracers of the influence of a coal-fired power plant on a Scots pine forest in Catalonia (NE Spain). *Atmospheric Environment* 42: 733–745. DOI:10.1016/j.atmosenv.2007.09.059.

Schaub M., Skelly J.M., Zhang J.W., Ferdinand J.A., Savage J.E., Stevenson R.E., Davis D.D., Steiner K.C. 2005. Physiological and foliar symptom response in the crowns of *Prunus serotina*, *Fraxinus americana* and *Acer rubrum* canopy trees to ambient ozone under forest conditions. *Environmental Pollution* 133: 553–567. DOI:10.1016/j.envpol.2004.06.012.

Sha Ch., Wang T., Lu J. 2010. Relative Sensitivity of Wetland Plants to $SO_2$ Pollution. *Wetlands* 30: 1023–1030. DOI 10.1007/s13157-010-0095-x.

Sicard P., Dalstein-Richier L., Vas N. 2011. Annual and seasonal trends of ambient ozone concentration and its impact on forest vegetation in Mercantour National Park (South-eastern France) over the 2000–2008 period. *Environmental Pollution* 159(2): 351–362. DOI:10.1016/j.envpol.2010.10.027.

Simpson D., Butterbach-Bahl K., Fagerli H., Kesik M., Skiba U., Tang S. 2006. Deposition and emissions of reactive nitrogen over European forests: A modelling study. *Atmospheric Environment* 40: 5712–5726. DOI: 10.1016/j.atmosenv.2006.04.063.

Solberg S., Dobbertin M., Reinds G.J., Lange H., Andreassen K., Fernandez P.G., Hildingsson A., de Vries W. 2009. Analyses of the impact of changes in atmospheric deposition and climate on forest growth in European monitoring plots: A stand growth approach. *Forest Ecology and Management* 258: 1735–1750. DOI:10.1016/j.foreco.2008.09.057.

Szaro R., Bytnerowicz A., Oszlányi J. 2002. Effects of air pollution on forest health and biodiversity in forest of the Carpathia Mountains. NATO Science Series I, vol. 345. ISBN: 1 58603 258 5.

Tjoelker M.G., Volin J.C., Oleksyn J., Reich P.B. 1995. Interaction of ozone pollution and light on photosynthesis in a forest canopy experiment. *Plant, Cell, and Environment* 18: 895–905. DOI: 10.1111/j.1365-3040.1995.tb00598.x.

Wagg S., Mills G., Hayes F., Wilkinson S., Cooper D., Davies W.J. 2012. Reduced soil water availability did not protect two competing grassland species from the negative effects of increasing background ozone. *Environmental Pollution* 165: 91–99. DOI:10.1016/j.envpol.2012.02.010.

Wamelink G.W.W., Wieggers H.J.J., Reinds G.J., Kros J., Mol-Dijkstra J.P., van Oijen M., de Vries W. 2009. Modelling impacts of changes in carbon dioxide concentration, climate and nitrogen deposition on carbon sequestration by European forests and forest soils. *Forest Ecology and Management* 258: 1794–1805. DOI:10.1016/j.foreco.2009.05.018.

Warmiński K., Rogalski L., Bęś A. 2005. Oddziaływanie dwutlenku siarki i siarczanów(IV) na zanik chlorofilu w niektórych roślinach wskaźnikowych. *Zeszyty Problemowe Postępów Nauk Rolniczych* 505: 491–501.

Wei X., Blanco J.A., Jiang H., Kimmins J.P.H. 2012. Effects of nitrogen deposition on carbon sequestration in Chinese fir forest ecosystems. *Science of the Total Environment* 416: 351–361. DOI: 10.1016/j.scitotenv.2011.11.087.

Weinstein D.A., Laurence J.A., Retzlaff W.A., Kern J.S., Lee E.H., Hogsett W.E., Webera J. 2005. Predicting the effects of tropospheric ozone on regional productivity of ponderosa pine and white fir. *Forest Ecology and Management* 205: 73–89. DOI:10.1016/j.foreco.2004.10.007.

Wesołowski M., Radecka I. 2003. Znaczenie roślin w monitoringu zanieczyszczenia środowiska naturalnego pierwiastkami metalicznymi. *Ekologia i Technika* 11(4): 14–22.

Wittmann Ch., Rainer Matyssek R., Pfanz H., Humar M. 2007. Effects of ozone impact on the gas exchange and chlorophyll fluorescence of juvenile birch stems (*Betula pendula* Roth.). *Environmental Pollution* 150: 258–266. DOI: 10.1016/j.envpol.2007.01.013.

Woźny A. 2004. Wybrane gazowe czynniki stresowe ($SO_2$, $NO_x$, $O_3$), in: Komórki roślinne w warunkach stresu. (eds. A. Woźny, K. Przybył), Tom I, cz. 2. Wydawnictwo Naukowe UAM, 78–165. ISBN: 83-232-1443-3.

Wu Z., Wang X., Chen F., Turnipseed A.A., Guenther A.B., Niyogi D., Charusombat U., Xia B., Munger W.J., Alapaty K. 2011. Evaluating the calculated dry deposition velocities of reactive nitrogen oxides and ozone from two community models over a temperate deciduous forest. *Atmospheric Environment* 45: 2663–2674. DOI: 10.1029/2011JD016751.

Zapletal M., Cudlín P., Chroust P., Urban O., Pokorný R., Edwards-Jonášová M., Czerný R., Janouš D., Taufarová K., Večeřa Z., Mikuška P., Paoletti E. 2011. Ozone flux over a Norway spruce forest and correlation with net ecosystem production. *Environmental Pollution* 159: 1024–1034.

## Author's contribution

M.B. – collection of information; K.W. – interpretation of collected data, preparation of literature review; A.B. – collection of information, text layout, also including data tables.

Translated by: Olga Zyrina