

Integrated Evaluation of Road Transport Pollution Impact on the Urban Air

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Abstract. With the number of vehicles increasing, the analysis of urban air pollution becomes expedient. This article deals with the integrated evaluation of road transport realised pollutant impact on the urban air. During research, it was carried out complex measurements of the air quality involving passive diffusive sampling for nitrogen dioxide, active measurement for particle matters, lichen sampling for heavy metals and visual assessment of trees defoliation. Obtained results showed the statistically reliable ($p < 0.05$) strong correlation ($r = 0.83$) between the number of passing vehicles and the concentration of particulate matter and there is even stronger correlation ($r = 0.94$; $p < 0.05$) between the concentration of nitrogen dioxide and the number of passing vehicles. It was observed during the analysis, that in measuring sites in which was determined 30% more intense defoliation process, also determined a higher NO_2 ($>10\mu\text{g}/\text{m}^3$) and lead ($\sim 10\text{ mg}/\text{kg}$) concentrations. It can be argued that the source of mentioned pollutants is the same – motor transport, and their presence in the environment influences defoliation phenomenon.

Keywords: air pollution, nitrogen dioxide, particle matter, heavy metals, defoliation.

Conference topic: Environmental protection.

Introduction

The greatest impact on the urban air is made by the emissions of mobile sources of pollution. Transport, in particular, car transport is currently the largest source of atmospheric air pollution. Energy facilities such as thermal power plants, boiler rooms, industrial enterprises and transport have been the main sources of air pollution in Lithuania for many years. During the recent years, air pollution from the energy sector and industrial sources has decreased significantly, however the transport pollution has remained almost the same and become predominant.

The car emissions depend on such factors as a road carriageway coating, fuel type, technical condition and operating mode of a car engine. The main components of toxic compounds emitted by cars are the following: carbon monoxide (CO), nitrogen oxides (NO_x), sulphur compounds (SO_2), smoke, unburnt lead compounds, carbohydrates (CH_x) which lead to local, regional and global air pollution problems (Caussy *et al.* 2003). These pollutants have an influence on the environment: road vehicles contaminate the atmosphere, water and soil near the highway via atmospheric fallout. Meteorological conditions, for instance, wind, rainfall, profiles or traffic intensity are the factors affecting dispersion of contaminants (Viard *et al.* 2004).

Nitrogen oxides are among the main pollutants which are produced by vehicles and are emitted during the fuel combustion process in internal combustion engines, as well as during industrial and energy production processes. In 2012, 45% of nitrogen oxide emissions in the European Union were produced by transport, and 39% of it from road transport. Nitrogen dioxide also contributed to photochemical air pollution. Its amount and distribution is of significance directly for air quality and human health and indirectly as an ozone precursor (e.g. Schau *et al.* 2005; Vintar Mally, Ogrin 2015).

The main sources of heavy metals (Cu, Pb, Cr, Cd and Zn) in the air are traffic, domestic heating and longrange transport (Grigalavičienė *et al.* 2005). Motor transport is one of the most important sources of emissions of particulate heavy metals into the air. Road traffic involves numerous potential sources of metals, e.g. combustion products from fuel and oil; wearing products from tyres, brake linings, bearings and clutches; corrosion products of vehicle components and road construction materials; and resuspension of soil and road dust (Duong, Lee 2010). Across the European Environment Agency countries members (EEA-33), emissions of lead decreased by 92%, mercury by 73% and cadmium by 66% during the period from 1990 to 2014. Across the EEA-33 countries, emissions of lead from the road transport sector decreased by 98% during the period from 1990 to 2014. Nevertheless, the road transport sector still remains an important source of lead, contributing approximately 15% of total lead emissions in the EEA-33 region. The largest sources are industrial processes and product consumption, which together comprise 23% of emissions.

However, since 2004, little progress has been made in further reduction in emissions; 99% of the total reduction in emissions of lead from 1990 levels had been achieved by 2004 (EEA 2016). As environmental pollution by heavy metals creates hazards to human health and the environment, their concentrations in individual elements of the environment are monitored and emissions are inventoried (Faiz *et al.* 2009)

Air pollution (especially by sulphur and nitrogen oxides) and climate conditions (Laubhann *et al.* 2009) are considered to be the key factors influencing forest health (Percy, Ferretti 2004; de Vries *et al.* 2003; Tkacz *et al.* 2007; Malik *et al.* 2012). Recently, the adverse effects of high tropospheric ozone concentrations have become considerably important (see Ferretti *et al.* 2003; Percy, Ferretti 2004). Acid rain and ambient O₃ are among the key factors resulting in spatial and temporal changes of tree crown defoliation (Takemoto *et al.* 2001). Defoliation is a raw visual indicator of the relative amount of foliage on the tree crown compared to a reference standard tree, and is assessed visually by trained field teams (Ferretti *et al.* 1999; Pollastrini *et al.* 2016). Acidifying compounds in the environment can cause crown defoliation and foliage discoloration (Klap *et al.* 2000; Ozolinčius, Stakėnas 2001; Stravinskienė *et al.* 2013). Ozolinčius (Ozolinčius *et al.* 2007) points out that it is difficult to establish a clear relationship between a certain stress factor and the response variable (e.g. defoliation) because of a lot of concurrent environmental factors.

Trees are considered to be very sensitive indicators of the environmental status; they are a life form which is the most appropriate for assessment of environmental status. The condition of trees may provide information on the changes occurring in the environment. In order to be able to assess the changes in the environment determined by urbanisation and development of cities, it is necessary to carry out the monitoring of a part of green areas, i.e. trees in urban areas, and to analyse the indicators describing their state (Stravinskienė 2010). Defoliation of a tree crown shows not only the loss of mass of formed foliage or needles caused by unfavourable environmental factors (excluding autumn leaf fall and natural needle fall), but also the part of the potential foliage or needle mass which could have been formed under optimum conditions, however, have not been formed in the specific environment. Trees in urban environment are sensitive to weather changes, therefore the long-term monitoring of such trees helps to establish air quality. One of the monitoring aspects is tree defoliation process. During this process, leaves begin to sallow and fall. This depends on various factors and one of them is air pollution. The first signs of defoliation appear in July and August (Šiburskytė, Stravinskienė 2014).

The accumulation of metals in plants is determined by many factors, such as the availability of elements; the characteristics of the plants, such as species, age, state of health, type of reproduction, etc.; and other such as parameters as temperature, available moisture, substratum characteristics, etc. Contaminants deposit on lichens through normal and indirect (occult) precipitation (Conti, Cecchetti 2001).

The purpose of the paper is to measure the quantities of nitrogen dioxide, particulate matter and heavy metals in the streets with different traffic intensity as well as to assess their impact on the tree defoliation process.

Methods and material

Sampling area and strategy

Kuršėnai town in the district of Šiauliai has been chosen as the site of integrated measurements. The vehicles passing on the road section Šiauliai – Klaipėda have the greatest impact on air pollution in this town. Within this section, Vytautas street is mostly loaded and has no industrial objects, therefore this particular section reflects the impact of passenger cars and heavy vehicles on the quality of the ambient air. The main streets have industrial objects (Pramonės and Ventos streets) whose emissions into the atmosphere comprise approximately 25% of overall urban air pollution. Therefore the research on defoliation process, nitrogen dioxide, particulate matter and heavy metals was carried out close to the roads with different traffic intensity by assessing the pollution in each traffic zone (Fig. 1, Table 1).

The integrated scientific research was carried out during the period from July to September of 2015 (Table 2). At the beginning of research (on the 4th of July), the condition of all trees selected for research was recorded, the diffusive samplers were started to be exposed and the concentrations of particulate matter were measured. Later, the research was carried out once every two weeks. At the end of the 2-week period, the condition of the research trees was assessed. The exposed diffusive samplers were removed and transported to the laboratory for further analysis. The new diffusive samplers were placed; the concentrations of particulate matter were measured.

Lichens growing on the research trees were selected as the accumulating medium for research on heavy metals. Samples of lichens were taken in August, later they were prepared in laboratory and analysed by an atomic absorption spectrometer.

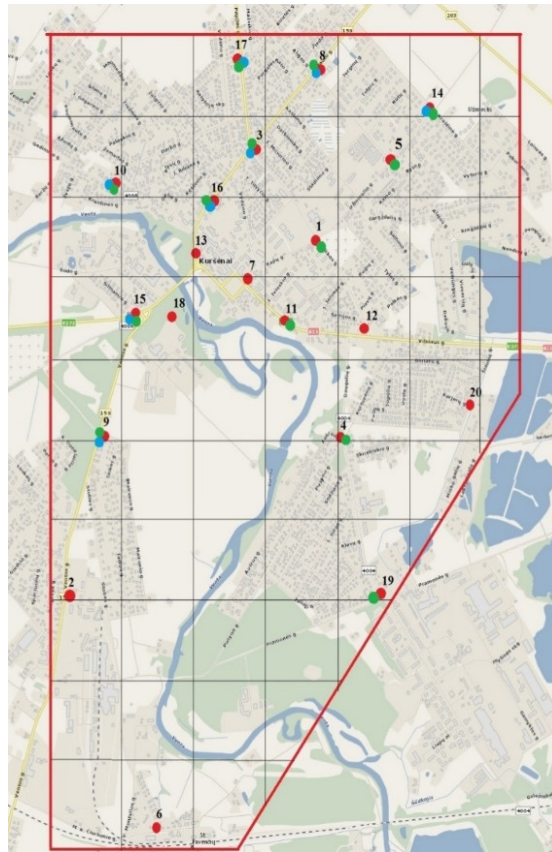


Fig. 1. Sampling sites

Table 1. Sampling site description

Sampling site No.	Description of traffic intensity	Pollutant	Tree species
1	Low traffic intensity	NO ₂ , Pb, Cd, Cu, Zn	Common ash (<i>Fraxinus excelsior</i>)
4		NO ₂ , Pb, Cd, Cu, Zn	Common ash (<i>Fraxinus excelsior</i>)
5		NO ₂ , Pb, Cd, Cu, Zn	Scots pine (<i>P. sylvestris L.</i>)
6		NO ₂	Norway Maple (<i>Acer platanoides</i>)
7		NO ₂	Small-leaved Lime (<i>Tilia cordata Mill.</i>)
10		NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn	Scots pine (<i>P. sylvestris L.</i>)
19		NO ₂	Norway spruce (<i>Picea abies</i>)
2		Average traffic intensity	NO ₂
11	NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn		Birch (<i>Betula sp.</i>)
12	NO ₂		Common ash (<i>Fraxinus excelsior</i>)
14	NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn		Norway spruce (<i>Picea abies</i>)
18	NO ₂		Scots pine (<i>P. sylvestris L.</i>)
20	NO ₂		Norway spruce (<i>Picea abies</i>)
3	Intensive traffic	NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn	Common ash (<i>Fraxinus excelsior</i>)
8		NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn	Common ash (<i>Fraxinus excelsior</i>)
9		NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn	Common ash (<i>Fraxinus excelsior</i>)
13		NO ₂	Norway spruce (<i>Picea abies</i>)
15		NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn	Small-leaved Lime (<i>Tilia cordata Mill.</i>)
16		NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn	Small-leaved Lime (<i>Tilia cordata Mill.</i>)
17		NO ₂ , PM ₁₀ , Pb, Cd, Cu, Zn	Small-leaved Lime (<i>Tilia cordata Mill.</i>)

Table 2. Sampling campaign

Research object	Sampling period	Sampling date
NO ₂	I	07 04–07 18
	II	07 25–08 08
	III	08 08–08 22
	IV	08 22–09 05
	V	09 05–09 19
KD ₁₀ , Defoliation	I	07 04
	II	07 17
	III	08 07
	IV	08 21
	V	09 04
Heavy metals	–	08 22

Sampling methodology

The passive sampling method was chosen for measurement of the *nitrogen dioxide* concentrations in the atmosphere. A diffusive sampler consist of a small diffusive tube with one end filled by a sorbent capable to collect pollutants from the ambient air without the additional active air intake (Vasiliauskienė *et al.* 2016).

Passive samplers were used for measuring the nitrogen dioxide concentration. The Palmes-type diffusion tube for nitrogen dioxide is a passive device requiring no power for its operation. It collects NO₂ by means of molecular diffusion, which takes place along the inert tube to an absorbent (in this case, triethanolamine (TEA)). The sampler consists of a polypropylene tube (21 mm internal diameter and 34 mm long) and stainless steel wire mesh. During preparation of the diffusive tubes, the stainless steel meshes were impregnated with a 20% solution of TEA in water. In the sampling site, the samplers were mounted vertically, and the lower plug was removed at the onset of sampling in order to allow NO₂ to be transported by molecular diffusion upwards the tube to the TEA where it is retained. The plug was replaced at the end of sampling, and the exposed sampler was transported to the laboratory for further analysis.

The collected pollutant was determined spectrophotometrically by applying the Saltzmann method. Ten minutes are required for full coloured development before optical absorbance of the coloured solution is measured spectrophotometrically. The principle of the chemical reaction within the tube is the following: NO₂ is chemiadsorbed in triethanolamine as a nitrite ion (NO₂⁻). During the analysis in the laboratory, the nitrite reacts with sulphanilamide and forms a diazonium compound which reacts with NEDA to form purple red azodye which is measured in an UV spectrophotometer at 542 nm. The concentration of the azodye is proportional to the amount of NO₂ chemiadsorbed over the sampling period. The mass of nitrite in the cartridge is obtained by reference to a linear calibration derived from the spectrophotometric analysis of standard solutions of sodium nitrite (Šerevičienė *et al.* 2014). Concentration is calculated from the quantity of nitrite in the sampler by means of Eq. 1:

$$C(\mu\text{g} \cdot \text{m}^{-3}) = \frac{m \cdot l}{A \cdot t \cdot D}, \quad (1)$$

where C represents the concentration of NO₂ ($\mu\text{g} \cdot \text{m}^{-3}$), m is the quantity of nitrite captured in the sampler (μg) and l , A , t and D denote the diffusion length (m), cross-sectional area (m^2), sampling time (s) and diffusion coefficient ($\text{m}^2 \cdot \text{s}^{-1}$), respectively.

The diffusion coefficient depends on certain environmental parameters such as temperature and pressure and could be calculated by means of Eq. 2:

$$D(\mu\text{g} \cdot \text{m}^{-3}) = D_{273\text{K}} \left(\frac{T + 273}{273} \right)^{1.81}, \quad (2)$$

where T is the temperature and $D_{273\text{K}}$ is the sampling rate at standard conditions ($0.1361 \text{ cm}^2 \cdot \text{s}^{-1}$ at $0 \text{ }^\circ\text{C}$ (273 K)).

In order to measure the concentrations of *particulate matter* in the air, the photometric method was used. Air enters the device through the inlet and is dispersed by light. The precise information on the number and size of the particles in the air is provided according to the absorption degree. The device shows the temperature and humidity of the inlet air. In order to measure the concentrations of particulate matter in the urban streets, the following instructions were followed: to measure the PM at the distance of 4 m from the carriageway, at the distance of 25 m from the centre of a crossroads; do not stand close to a wall; and air shall be sucked at the height of 1.5–1.7 m.

The measurements were carried out by using a device FLUKE 983 which measures the particles of 6 different sizes: 0.3; 0.5; 1; 2; 5 and 10 μm . With 1 litre of inlet air, the device shows the quantities of the particles of 0.3; 0.5; 1; 2; 5 and 10 μm separately. The obtained data (*PM number u.*) is recalculated into $\mu\text{g}/\text{m}^3$.

The measurements of the concentrations of particulate matter were carried out in 9 measurement sites; air intake lasted 30 min.

For estimation of the content of *heavy metals*, lichen samples of ~20 grams were taken from the trunks of analysis trees at the height of 3.5 metres from the side of road carriageway.

The flame atomic absorption spectrometry method was used for determination of heavy metals in lichens. This is one of the most popular methods allowing to establish concentrations of heavy metals in a solution. Air and acetylene gas mixture was used for formation of a flame. The detailed analysis of a sample is carried out as follows:

1) Collected lichens are dried at the temperature of 105°C. Dried lichens are crushed and 0.8 g of each sample are weighed with 1 analytical balance;

2) Preparation of samples for digestion: the weighed samples are placed into special container where 9 ml of concentrated nitric acid and 3 ml of concentrated hydrochloric acid are poured; all containers are placed into a drum which is inserted into a heating oven and the temperature as well as time (56 min) are selected according to the specification; after cooling of containers, the solution is poured into 50 ml flasks and thinned with deionised water up to the limit of 50 ml;

Defoliation of tree crown is determined by comparing a sample tree with a reference standard tree which has the whole (100%) foliage (Stravinskienė 2010). Trees are relatively healthy when defoliation of their crowns does not exceed 10 %. Defoliation of the whole tree crown and defoliation of the top third of it are established with an accuracy of 5 %. Either a tree of the same social class, the same type of branching, growing somewhere close to a sample tree or the respective picture of such tree (special atlases are used for this purpose), or the same tree with the ideal whole foliage is usually considered as a reference standard tree. Severe defoliation of a crown is the sign of a damage caused to a tree (Stravinskienė 2010). Before the research, it is necessary to assess not only physical, but also meteorological conditions. Research on 20 trees was carried out during the period from July to September on 20 measurements sites. When investigating the processes taking place in tree crowns, the trees of approximately the same age (preferably younger) were selected, since old trees may also be affected by other factors. At each observation, the object observed was photographed from two trajectories: against the light or to the direction of an angle of 45 degrees; with light from the side or in case of a high level of cloud depending on meteorological conditions (Dobbertin, Brang 2001).

Each research tree was assessed by determining the following: percentage of the foliage which has not been formed, percentage of fallow leaves and the part of fallen leaves. The degree of leaf loss was established in accordance with the classification of the United Nations Economic Commission for Europe (UNECE) and the European Union (EU) (Table 3).

Table 3. Defoliate classification according to UNECE and EU (Rullan-Silva *et al.* 2013)

Defoliation class	Leaf / needle loss	Defoliation degree
0	<10 %	none
1	11–25 %	slight (warning phase)
2	26–60 %	moderate
3	61–<100 %	severe
4	100%	dead

Statistic data processing and quality assurance

During the testing period two unopened control diffusive samplers intended for determining nitrogen dioxide were left in the storage location (in a cool dark place). In addition, during the testing period two unopened samplers were transported for the purposes of control of the process of samplers' transportation. After the sampling period, both the control samplers and those that have been exposed were forwarded for laboratory analysis. The Fluke 983 Particle Counter was calibrated at the beginning of research. To obtain the calibration curves for heavy metals analysis with "Buck Scientific" 210 VGP atomic absorption spectrophotometer, using flame atomic absorption spectroscopy, standard solutions were used.

Statistical data analysis was performed using such applications as Microsoft Excel and STATISTICA 8.0. The results including the mean values with the values of standard deviations were represented graphically.

Discussion and analysis

Air pollution in Kuršėnai town was assessed by means of the integrated research: the amount of nitrogen dioxide, particulate matter, and heavy metals was established and the content of heavy metals accumulating in the lichens growing on trees was measured. Moreover, the degree of defoliation of tree crowns was established and the qualitative research was carried out. The research on defoliation represents the visual evaluation of common pollution, which is later compared with the qualitative research.

Air pollution depends on meteorological conditions such as temperature, wind velocity and air humidity (Valuntaitė *et al.* 2009; Šerevičienė *et al.* 2014; Vasiliauskienė *et al.* 2016), therefore the data from the nearest weather station in Šiauliai was used during the research period (Table 4). The prevailing higher wind (up to 2 m/s) per day leads to faster dispersion of compounds in the ambient air. The higher air humidity (>60%) leads to faster chemical reactions resulting in the formation of secondary pollutants and part of them deposit on the objects in the environment (Viswanathan, Krishna Murti 1989).

Table 4. Average air temperature, humidity and average wind velocity during the research period

Sampling period	Temperature, °C	Relative humidity, %	Average wind velocity, m/s
I	16.4	50–70	2.5
II	15.9	65–90	2.1
III	17.9	25–65	1.5
IV	17.5	30–75	1.8
V	15.1	75–95	2.0

Having considered the location of the town and the natural conditions, it was decided that the background concentration of the town is reflected by the measurement site No. 1. There are traffic restrictions for heavy vehicles, two schools with the territory surrounded by trees at this point.

During the research periods, the NO₂ concentration in Kuršėnai town reached the level of concentrations of nitrogen dioxides, which poses risk to vegetation, i.e. 23 µg/m³. Exposure of plants to such concentrations may cause the following consequences: slower growing, acute injuries, brown spots as well as leaf browning and loss (Liu *et al.* 2015; Goyal, Chavhan 2015). The measured average NO₂ concentrations varied from 4.47 to 14.62 µg/m³ depending on different traffic intensities (Fig. 2). In many cases, the concentration was 2–4 times higher than the background concentration 3 µg/m³.

In the zone with high traffic intensity, when the average temperature during the research period was 15.9 °C, the concentrations of nitrogen dioxides did not exceed 10 µg/m³, however, an increase of temperature by 2 °C resulted in the increase of the chemical compound by 1.7 times (Fig. 2). The similar results have already been presented by the scientists Yuksel and Michalek (2015) who have found during their research that the increase of temperature by 1 °C leads to increase of air pollution by 2.2 times. Having calculated the correlation coefficient, it was established that it is statistically significant ($p < 0.05$) strong correlation ($r = 0.78$) between the temperature and the nitrogen dioxide concentration distribution in the air in the high traffic density zone (zone 3).

In general case, in July (period I–II), when the average air temperature was lower (15.4°C on the average) in comparison with the temperature in August (17.5 °C), the obtained results were influenced by the air humidity (~50–90% in July). Under the dry weather conditions (air humidity <50%), dispersion of nitrogen dioxide in the ambient air increases, otherwise, they react with water and enter the soil. The research carried out by Lovell-Smith and other scientists has showed that the dry air reduces the number of reactions, however increases the dispersion of chemical compounds in the ambient air: when the air humidity is higher than 60%, intensity of the reactions with H₂O increases by 1.5 times (Lovell-Smith *et al.* 2016).

During the research period, the highest concentration was established in the high traffic density zone (on the measurement site No. 16) where the level of nitrogen dioxide concentration posed risk to vegetation and reached 23.36 µg/m³. The nitrogen dioxide concentration in the air is influenced by many factors, however, one of the main factors is car transport (Kurtenbach *et al.* 2012; Carslaw, Rhys-Tyler 2013; Šerevičienė *et al.* 2014). The concentration of particulate matter in the ambient air is determined by many factors such as meteorological conditions, road surface and car transport (Kurtenbach *et al.* 2012). The concentration of particulate matter significantly increases close to motor highways (Zhang, Batterman 2013).

The streets of Kuršėnai were divided into groups according to traffic density:

- low traffic density, i.e. up to 999 vehicles per day;
- average traffic density, i.e. from 1,000 to 1,999 vehicles per day;
- high traffic density, i.e. from 2,000 to 7,913 vehicles per day.

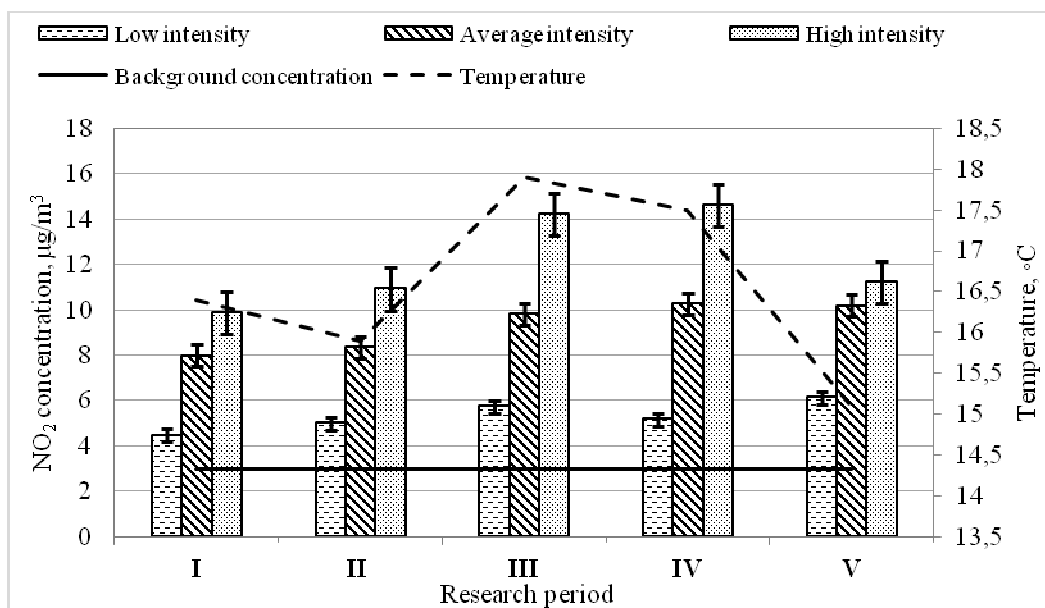


Fig. 2. Changes of NO₂ concentration during 5 sampling periods near different intensity traffic roads

The increase of the concentration of particulate matter is mostly influenced by heavy vehicles, which comprised 13% of all passing vehicles during the research period. The scientific research carried out by Snigdha Kundu and Asim Kumar Pal (2015) also has shown that the content of the particulate matter in the ambient air is increased by heavy vehicles by 30%.

With the decrease in car flow in each part of the town, the PM concentration decreased by 28% on the average (Fig. 3), while NO₂ decreased by 32% on the average. During the assessment of the results, it was found that the increase and decrease of the amount of nitrogen dioxide are mostly influenced by the number of motor vehicles. The changes in the PM concentration may also be influenced by the degree of road surface cleaning. However, under the conditions of south westerly higher wind and low air humidity, the increase of air pollution was recorded and the particles deposited on the surface of the plant become visually detectable. Plants are more sensitive than a human body, therefore increase of air pollution even by several micrograms has a negative impact on the ecosystem.

When analysing the content of particulate matter and nitrogen dioxide in the ambient air, the correlation coefficient was calculated and it showed that there is the statistically reliable ($p < 0.05$) strong correlation ($r = 0.83$) between the number of passing vehicles and the concentration of particulate matter and there is even stronger correlation ($r = 0.94$; $p < 0.05$) between the concentration of nitrogen dioxide and the number of passing vehicles (Fig. 3).

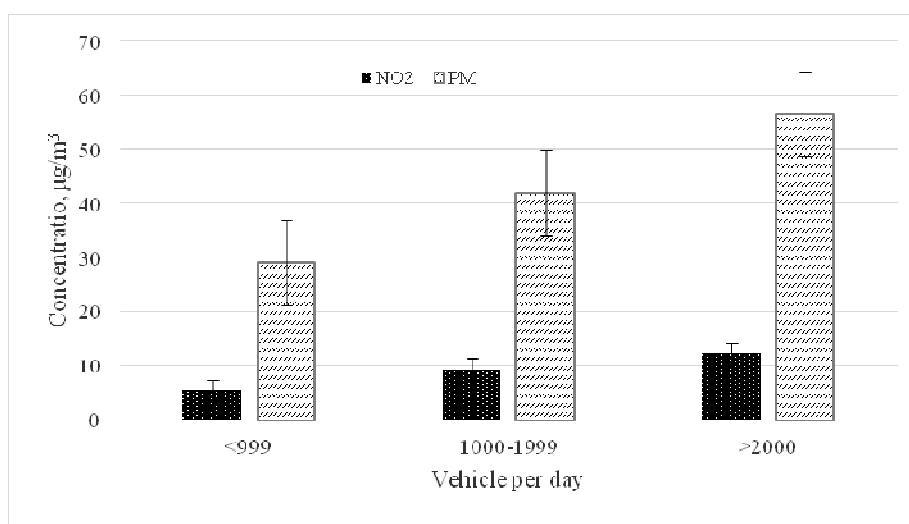


Fig. 3. Average concentrations of NO₂ and PM in different traffic intensities

Although heavy metals constitute an insignificant portion among transport pollutants, they play a potential role in assessing the quality of the roadside environment (Grigalavičienė *et al.* 2005). Heavy metals accumulate in the leaves, shoots and roots of trees depending on the path of uptake. Accumulation of such compounds in plants mostly depends on their species, growing conditions and development phase. It has already been researched that heavy metals enter plants in the following ascending order: $Cd > Pb > Zn > Cu$ and such entering depends on their agility and content in the soil. However, the best bioindicators showing the content of heavy metals are lichens which absorb heavy metals of various forms. Since the number of industrial objects is reduced in Kuršėnai town, vehicles have the highest impact on the occurrence of heavy metals in the environment.

As background concentration of the research area was adopted concentration of the heavy metals measured at the point No.1 (Pb – 4,21 mg/kg, Cd – 0,1 mg/kg, Zn – 37,5 mg/kg, Cu – 5,94 mg/kg).

Usually the lead concentration in plants varies from 0.05–3 mg/kg (Mažvila 2001), however the lead concentrations in the research lichen samples exceed (Fig. 4) the levels most frequently occurring in Lithuania (up to 3 mg/kg). The lead concentrations are mostly influenced by traffic and relief (Micu *et al.* 2016; Ferreira *et al.* 2014) since the vehicles running uphill use more fuel which results in higher emissions of lead compounds formed during the combustion process. The highest lead accumulation, i.e. 11.25 mg/kg, was found in the high traffic intensity zones (measurement site No. 16, Fig. 4).

Unusually high zinc concentrations were found in the high traffic density zones of Kuršėnai town. Concentrations varied from 29.38 to 182.71 mg/kg. The levels of this particular microelement exceed its normal concentrations on the sampling site No. 16 by 2 times (up to 100 mg/kg) (Fig. 4).

The cadmium concentration in lichens was not excessive (0.9 mg/kg). As it could be expected, the highest concentration of the cadmium (0.65 mg/kg) was found in the measurement site No. 16. The highest copper concentration found during the research was 23 mg/kg, when the most frequently occurring concentration of copper in Lithuania starts from 10–50 mg/kg.

The most sensitive to the environment conditions are deciduous trees according to prevailing meteorological and other conditions in Lithuania, they accounted for 60% in the overall context of researchers.

In the first measurement site growing common *ash* (*Fraxinus excelsior* L.) have lost a part of the leaves in July (about 25%), till the middle of August defoliation percentage amounted to 45%, such defoliation level is assigned to 2 class of defoliation. Although in this measurement site vegetation is exposed to only a small concentration of nitrogen dioxide ($\sim 3 \mu\text{g}/\text{m}^3$), but a small amount of nitrogen dioxide acting for a long time have an impact on the process. The concentration of lead was 4.21 mg/kg and is 1.4 times higher than in Lithuania usually determined value (3 mg/kg) and that is why tree leaves are also affected by lead (Mažvila 2001). Effect of plant defoliation reaches 10–30% under the operation of two compounds simultaneously (Sudachkova *et al.* 2015), it was observed in all monitoring sites (20 measuring points).

In road sections, in which was limited traffic of heavy vehicles, defoliation processes is weaker, but even up to $9 \mu\text{g}/\text{m}^3$ NO_2 concentration affects the tree foliage irrecoverably (Pavlov *et al.* 2009; Fabiánek *et al.* 2012; Marco *et al.* 2014). The most sensitive to changes of chemicals are young till 10-year-old trees. It is observed that leaves of young trees first begin to fall from the side of the carriageway. It was observed during the research of lichens taken from the investigated trees, that the concentration of lead is 2 times higher in the third measuring site (8.86 mg/kg) than in the first point, respectively defoliation process is more intense by 15%. During the evaluation of traffic zones it was noticed that defoliation of leaves is significantly higher and in some cases reaches up to 85%. Such a case was observed in sampling site no. 9, located between two intense traffic roads, in the park near the school.

Results of researches partly reflect the pollution of the environment, because pollutant concentration downhill is 1.5 times lower than its concentration uphill. Nitrogen dioxide concentration in 9 measuring site varies from 10.89 to $12.37 \mu\text{g}/\text{m}^3$, PM_{10} concentration reaches till $75 \mu\text{g}/\text{m}^3$, and lead concentration reaches till 10.99 mg/kg. This shows that the complex impact of chemical compounds is the most dangerous effect on plants. It was calculated the correlation coefficient between nitrogen dioxide and lead concentrations, established a very strong correlation ($r = 0.901$, $p < 0.05$) (Fig. 5).

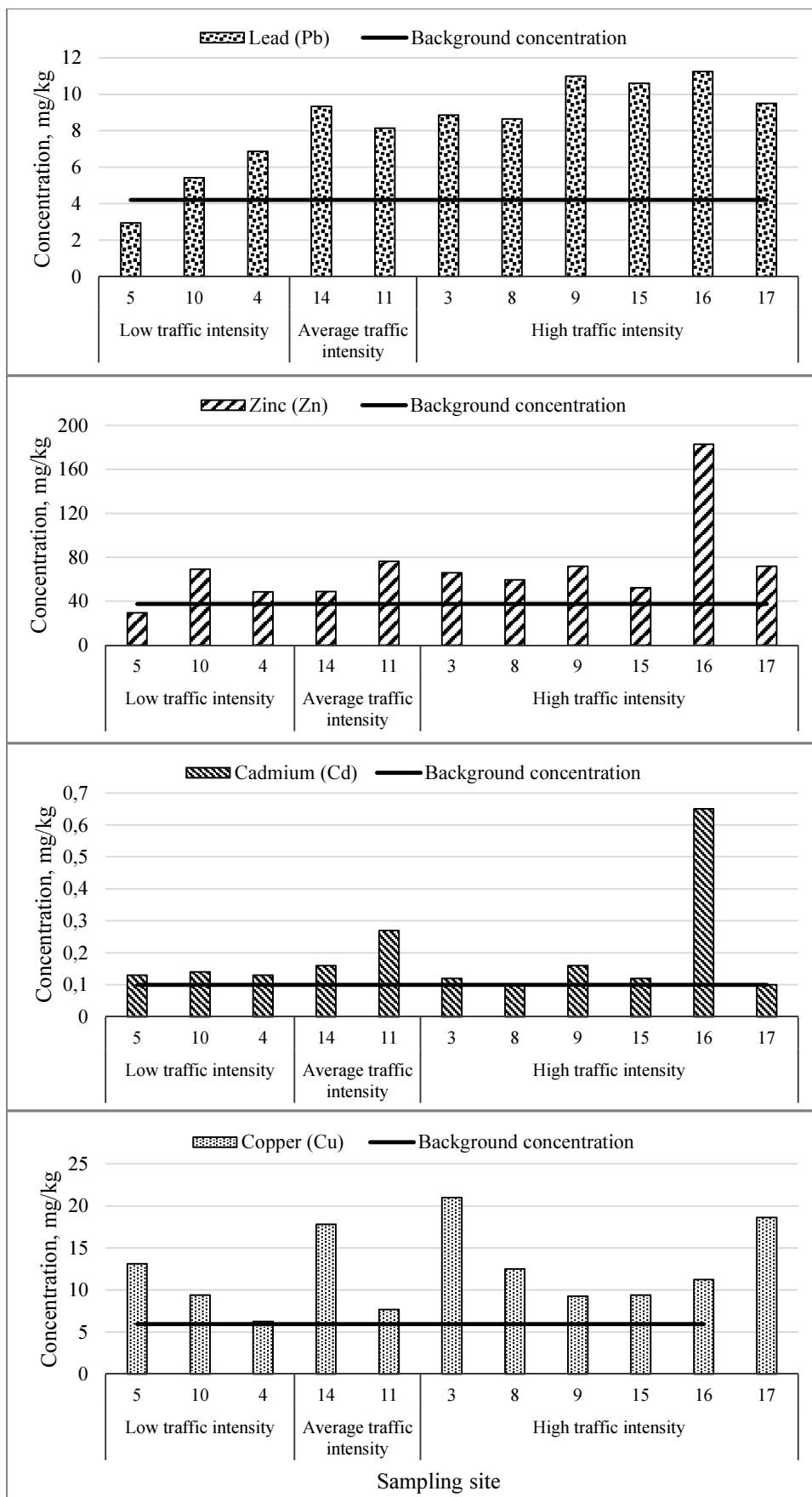


Fig. 4. Average concentrations of heavy metals in lichen samples in different traffic intensities

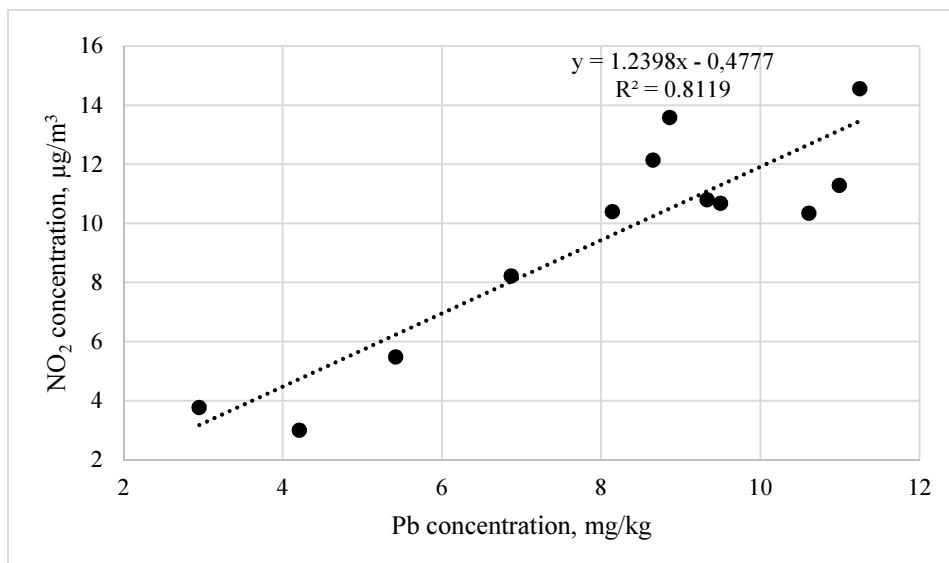


Fig 5. NO₂ concentration in urban air and Pb concentration measured in the same sampling sites

It was observed during the analysis, that in measuring sites in which was determined 30% more intense defoliation process, also determined a higher NO₂ (>10µg/m³) and lead (~10 mg/kg) concentrations. It can be argued that the source of mentioned pollutants is the same – motor transport, and their presence in the environment influences defoliation phenomenon.

Conclusions

Plants, i.e. bioindicators, are the most sensitive to ambient air pollution. The research has shown that one of the compounds which have the greatest impact on leaves of plants is nitrogen dioxide which by reacting with other elements forms the nitric acid. Heavy metals and particulate matter also contribute to the negative impact, i.e. they clog stomata and reduce the intensity of photosynthesis.

Having analysed the data obtained by means of NO₂ diffusive samplers exposed in Kuršėnai town, the statistically significant, strong correlations between the NO₂ content in the ambient air and the ambient temperature ($r = 0.78$, $p < 0.05$) as well as flows of motor vehicles ($r = 0.94$, $p < 0.05$) were established. In the high traffic density zone (>2,000 units/day) when the average diurnal temperature is higher than 16 °C, the nitrogen dioxide concentration increases by ~2 times (up to 23 µg/m³). With the decrease in car flow in each part of the town, the PM concentration decreased by 28% on the average, while NO₂ decreased by 32% on the average. During the assessment of the obtained results, it has been found that the increase and decrease of the amount of nitrogen dioxide are mostly influenced by the number of motor vehicles. Having measured the concentration of heavy metals (Pb, Cd, Zn, Cu) in the lichens growing in the research territory, it was established that the lead concentrations (2.95–11.25 mg/kg) exceeded the levels most frequently occurring in Lithuania (<3 mg/kg) on twelve measurements sites. The highest Pb, Cd and Zn concentrations have been established on the measurement site No. 16 which is close to the high density street.

During the assessment of the impact of the research pollutants on the intensity of defoliation, the statistically reliable strong correlation between defoliation and NO₂ concentration ($r = 0.87$, $p < 0.05$) and lead content in lichens ($r = 0.73$, $p < 0.05$) has been established. Nitrogen dioxide and lead have the greatest impact on the vegetation of Kuršėnai town near the transport roads. The calculations show very strong correlation between these compounds ($r = 0.901$, $p < 0.05$) which, in turn, proves that the source of these pollutants is the same, i.e. motor vehicle.

References

- Carslaw, D. C.; Rhys-Tyler, G. 2013. New insights from comprehensive on-road measurements of NO_x, NO₂ and NH₃ from vehicle emission remote sensing in London, UK., *Atmospheric Environmental* 81: 339–347. <https://doi.org/10.1016/j.atmosenv.2013.09.026>
- Caussy, D.; Gochfeld, M.; Gurzau, ER.; Nagu, C.; Ruedel, H. 2003. Lessons from case studies of metals: investigating exposure; bioavailability and risk, *Ecotoxicology and Environmental Safety* 56(1): 45–51. [https://doi.org/10.1016/S0147-6513\(03\)00049-6](https://doi.org/10.1016/S0147-6513(03)00049-6)

- Conti, M. E.; Cecchetti, G. 2001. Biological monitoring: lichens as bioindicators of air pollution assessment – review, *Environmental Pollution* 114: 471–492. [https://doi.org/10.1016/S0269-7491\(00\)00224-4](https://doi.org/10.1016/S0269-7491(00)00224-4)
- de Vries, W.; Vel, E.; Reinds, G. J.; Deelstra, H.; Klap, J. M.; Leeters, E. E. J. M.; Hendriks, C. M. A.; Kerkwooden, M.; Landmann, G.; Herkendell, J.; Haussmann, T.; Eisman, J. W. 2003. Intensive monitoring of forest ecosystems in Europe. 1. Objectives, set-up and evaluation strategy, *Forest Ecology and Management* 174: 77–95. [https://doi.org/10.1016/S0378-1127\(02\)00029-4](https://doi.org/10.1016/S0378-1127(02)00029-4)
- Duong, T. T. T.; Lee, B-K. 2010. Determining contamination level of heavy metals in road dust from busy traffic areas with different characteristics, *Journal of Environmental Management* 92(3): 554–562. 20 p.
- Dobbertin, M.; Brand, P. 2001. Crown defoliation improves tree mortality models, *Forest Ecology and Management* 141: 271–284. [https://doi.org/10.1016/S0378-1127\(00\)00335-2](https://doi.org/10.1016/S0378-1127(00)00335-2)
- European Environment Agency (EEA). 2016. *Heavy metal emissions. Indicator Assessment. Data and maps.*
- Fabiánek, P.; Hellebrandová, K.; Čepek, M. 2012. Monitoring of defoliation in forest stands of the Czech Republic and its comparison with results of defoliation monitoring in other European countries, *Journal of forests science* 58(5):193–202.
- Faiz, Y.; Tufail, M.; Tayyeb, M. T.; Chaudhry, M. M.; Naila-Siddique. 2009. Road dust pollution of Cd, Cu, Ni, Pb and and Zn along Islamabad Expressway, Pakistan, *Microchemica Journal* 92, 186–1*2.
- Ferreira, A. J. D.; Soares, D.; Ferreira, C. S. S.; Walsh, R. P. D. 2014. *Roads as sources of heavy metals in urban areas. The Covões Catchment experiment, Coimbra, Portugal.* EGU General Assembly 17: 2015–2025.
- Ferretti, M.; Brusasca, G.; Buffoni, A.; Bussotti, F.; Cozzi, A.; Petriccione, B.; Pompei, E.; Silibello, C. 2003. Ozone risk in the permanent plots of the Italian intensive monitoring of forest ecosystems – an introduction, *Annali dell'Istituto Sperimentale per la Selvicoltura* 30(Suppl. 1): 3–28.
- Ferretti, M.; Bussotti, F.; Cenni, E.; Cozzi, A. 1999. Implementation of quality assurance procedures in the Italian programs of forest condition monitoring, *Water Air Soil Pollution* 116: 371–376. <https://doi.org/10.1023/A:1005240000294>
- Goyal, S. K.; Chavhan, C. D. 2015. Assessment of uncertainty in ambient NO₂ concentration determination and its minimization through application of lab scale findings, *Journal of The Institution of Engineers (India)* 96(2): 131–137. <https://doi.org/10.1007/s40030-015-0118-y>
- Grigalavičienė, I.; Rutkoviėnė, V.; Marozas, V. 2005. The accumulation of heavy metals Pb, Cu and Cd at roadside forest soil, *Polish Journal of Environmental Studies* 14(1): 109–115.
- Yuksel, T.; Michalek, J. J. 2015. Effects of regional temperature on electric vehicle efficiency, range and emissions in the United States, *Environmental Science Technology* 49(6): 3974–3980. <https://doi.org/10.1021/es505621s>
- Klap, J. M.; Oude Voshaar, J. H.; De Vries, W.; Erisman, J. W. 2000. Effects of environmental stress on forest crown condition in Europe. Part IV: estimation of stress induced by meteorology and air pollutants, *Water, Air and Soil Pollution* 119: 387–420. <https://doi.org/10.1023/A:1005157208701>
- Kundu, S.; Pal, K. A. 2015. *Estimation of air quality in the opencast mine of Jharia coal field, India.* Department of Environmental Science & Engineering, 4: 156–164.
- Kurtenbach, R.; Kleffmann, J.; Niedojadlo, A.; Wiesen, P. 2012. Primary NO₂ emissions and their impact on air quality in traffic environments in Germany, *Environmental Science Europe* 2: 358–346.
- Laubhann, D.; Sterba, HReinds, G. J.; de Vries, W. 2009. The impacts of atmospheric deposition and climate on forest growth in European monitoring plots: an individual tree growth model, *Forest Ecology and Management* 258(8): 1751–1761. <https://doi.org/10.1016/j.foreco.2008.09.050>
- Liu, X.; Hou, F.; Li, G.; Sang, N. 2015. Effects of nitrogen dioxide and its acid mist on reactive oxygen species production and antioxidant enzyme activity in Arabidopsis plants, *Journal of environmental sciences* 34: 93–99. <https://doi.org/10.1016/j.jes.2015.03.011>
- Lovell-Smith, J. W.; Feistel, R.; Hatvey, A. H.; Hellmuth, O.; Bell, S. A.; Heinonen, M.; Cooper, J. R. 2016. Metrological challenges for measurements of key climatological observables, *Metrologia* 53(1): 40–59.
- Malik, I.; Danek, M.; Marchwińska-Wyrwał, E.; Danek, T.; Wistuba, M.; Krąpiec, M. 2012. Scots pine (*Pinus sylvestris* L.) growth suppression and adverse effects on human health due to air pollution in the upper silesian industrial district (USID), Southern Poland, *Water, Air and Soil Pollution* 223: 3345–3364. <https://doi.org/10.1007/s11270-012-1114-8>
- 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(11):171–180.
- Mažvila, J. 2001. *Sunkieji metalai Lietuvos dirvožemiuose ir augaluose: monografija.* Kaunas: Petro ofetas. 195 p.
- Micu, L. M.; Petanec, D. I.; Iosub-Ciur, M. D.; Andrian, S.; Popovici, R. A.; Porumb, A. 2016. The heavy metals content in leave of the forest fruits (*Hippophae rhamnoides* and *Rubus fruticosus*) from the tailings dumps mining, *Revista de Chimie (Bucharest)* 67(1): 64–68.
- Ozolinčius, R.; Stakėnas, V.; Serafinavičiūtė, B. 2007. Meteorological factors and air pollution in Lithuanian forests: possible effects on tree condition, *Environmental Pollution* 137(3): 587–595. <https://doi.org/10.1016/j.envpol.2005.01.044>
- Ozolinčius, R.; Stakėnas, V. 2001. Effect of air pollution and droughts on forest condition in Lithuania, *Biologija*, 2: 99–101.
- Pavlov, I. N.; Ageev, A. A.; Barabanova, O. A. 2009 Formation of annual rings of the main coniferous tree species in Siberia after defoliation of crown by *Dendrolimus superans sibiricus* Tschetv, *Khvoynye Boreal'noi Zony* 26(2): 161–172.
- Percy, E.; Ferretti, M. 2004. Air pollution and forest health: toward new monitoring concepts, *Environmental Pollution* 130: 113–126. <https://doi.org/10.1016/j.envpol.2003.10.034>

- Pollastrini, M.; Feducci, M.; Bonal, D.; Fotelli, M.; Gessler, A.; Grossiord, Ch.; Guyot, V.; Jactel, H.; Nguyen, D.; Radoglou, K.; Bussotti, F. 2016. Physiological significance of forest tree defoliation: results from a survey in a mixed forest in Tuscany (central Italy), *Forest Ecology and Management* 361: 170–178. <https://doi.org/10.1016/j.foreco.2015.11.018>
- Rullan-Silva, C. D.; Olthoff, A. E.; Delgado de la Mata; Pajares-Alonso, J. A. 2013. Remote monitoring of forest insect defoliation, *Forest Systems* 22(3): 377–391. <https://dx.doi.org/10.5424/fs/2013223-04417>
- Schau, D.; Weiss, A. K.; Kaiser, W. J.; Petritoli, A.; Richter, A.; Buchmann, B.; Burrows, J. P. 2005. A transboundary transport episode of nitrogen dioxide as observed from GOME and its impact in the Alpine region, *Atmospheric Chemistry and Physics* 5: 23–37. <https://doi.org/10.5194/acp-5-23-2005>
- Stravinskienė, V. 2010. Medžių būklės stebėseną ir vertinimą Kauno miesto aplinkoje, *Journal of Environmental Engineering and Landscape Management* 18(3): 217–225. <https://doi.org/10.3846/jeelm.2010.25>
- Stravinskienė, V.; Bartkevičius, E.; Plaušinytė, E. 2013. Dendrochronological research of Scots pine (*Pinus sylvestris* L.) radial growth in vicinity of industrial pollution, *Dendrochronologia* 31: 179–186. <https://doi.org/10.1016/j.dendro.2013.04.001>
- Sudachkova, N. E.; Milyutina, I. L.; Romanova, L. I.; Astrakhanseva, V. N. 2015. Effect of defoliation on the growth and metabolism of Scots pine, *Contemporary Problems of Ecology* 8(1): 21–27. <https://doi.org/10.1134/S199542551501014X>
- Šerevičienė, V.; Baltrėnas, P.; Baltrėnaitė, E.; Marčiulaitienė, E.; Paliulis, D. 2014. Investigation of NO₂ behaviour in the temperate continental climate road environment, *Water, Air and Soil Pollution* 225: 2173. <https://doi.org/10.1007/s11270-014-2173-9>
- Šiburskytė, S.; Stravinskienė, V. 2014. Pakruojo miesto ir dvaro parko želdinių būklės vertinimas, in *Žmogaus ir gamtos sauga. II dalis*, 39–42. <https://dx.doi.org/10.3846/mla.2016.94>
- Takemoto, B. K.; Bytnerowicz, A.; Fenn, M. E. 2001. Current and future effects of ozone and atmospheric nitrogen deposition on California's mixed conifer forests, *Forest Ecology and Management* 144: 159–173. [https://doi.org/10.1016/S0378-1127\(00\)00368-6](https://doi.org/10.1016/S0378-1127(00)00368-6)
- Tkacz, B.; Moody, B.; Castillo, J. V. 2007. Forest health status in North America, *The Scientific World Journal* 7(S2): 28–36. <https://doi.org/10.1100/tsw.2007.85>
- Valuntaitė, V.; Šerevičienė, V.; Girgždienė, R. 2009. Ozone concentration variations near high-voltage transmission lines, *Journal of environmental engineering and landscape management* 17(1): 28–35. <https://dx.doi.org/10.3846/1648-6897.2009.17>
- Vasiliauskienė, V.; Šerevičienė, V.; Zigmontienė, A. 2016. Spatial and temporal variation in ozone and nitrogen dioxide in the Seaside recreation area environment, *Polish Journal of Environmental Studies*: 25(2): 795–803. <https://doi.org/10.15244/pjoes/61283>
- Viard, B.; Pihan, F.; Promeyrat, S.; Pihan, J. C. 2004. Integrated assessment of heavy metal (Pb, Zn, Cd) highway pollution: bioaccumulation in soil, Gramineae and land snails, *Chemosphere* 55: 1349–1359. <https://doi.org/10.1016/j.chemosphere.2004.01.003>
- Vintar Mally, K.; Ogrin, M. 2015. Spatial variations in nitrogen dioxide concentrations in urban Ljubljana, Slovenia, *Moravian Geographical Reports* 23(3): 27–35. <https://doi.org/10.1515/mgr-2015-0015>
- Viswanathan, P. N.; Krishna Murti, C. R. 1989. Effects of temperature and humidity on ecotoxicology of chemicals, *Ecotoxicology and Climate* 140–154.
- Von Storch, H.; Costa-Cabral, M.; Hagner, C.; Feser, F.; acyna, J.; Pacyna, P. E.; Kolb, S. 2003. Four decades of gasoline lead emissions and control policies in Europe: a retrospective assessment, *Science of the Total Environment* 311(1–3): 151–176. [https://doi.org/10.1016/S0048-9697\(03\)00051-2](https://doi.org/10.1016/S0048-9697(03)00051-2)
- Zhang, K.; Batterman, S. 2013. Air pollution and health risks due to vehicle traffic, *Science of the Total Environment* 450–451: 307–316. <https://doi.org/10.1016/j.scitotenv.2013.01.074>