Air pollution deposition on a roadside vegetation barrier in a Mediterranean environment: Combined effect of evergreen shrub species and planting density

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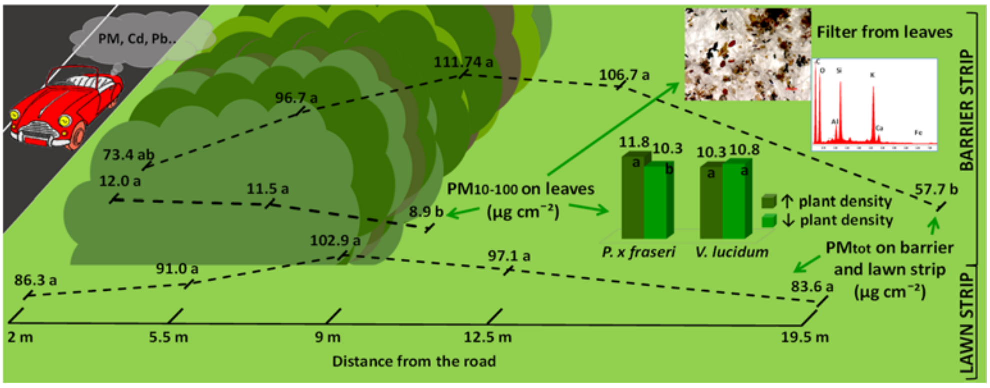
Highlights

- Higher LAI induced higher deposition, while planting density was not a determinant
- Vegetation barrier changed deposition dynamics in the experimental site
- Image analysis differentiated between on-leaf PM with different colorations
- On-leaf PM with different colorations had a different element composition

Graphical abstract



Abstract

Leaf deposition of PM_{10-100}, $PM_{2.5-10}$, $PM_{0.2-2.5}$ and of 21 elements was investigated in a roadside vegetation barrier formed by i) two evergreen shrub species (*Photinia x fraseri, Viburnum lucidum*), with ii) two planting densities (0.5,1.0 plant m^{-2}), at iii) three distances from the road (2.0, 5.5, 9.0 m), at iv) two heights from the ground (1.5, 3.0 m), and on v) three dates (Aug, Sep, Oct).

The presence of black and brown on-leaf PM_{10-100} and their element composition were detected by microscopy and image analysis. Pollutant deposition was also measured using passive samplers at five distances from the road (2.0, 5.5, 9.0, 12.5, 19.5 m) in the area of the barrier and in an adjacent lawn area.

V. lucidum had more $PM_{2.5-10}$ and $PM_{0.2-2.5}$ on leaves than *P. x fraseri,* while most elements were higher in *P. x fraseri.* Most pollutants decreased at increasing distances from the road and were higher at 1.5 m from the ground compared to 3.0 m.

Higher planting density in *P. x fraseri* enhanced the deposition of PM_{10-100} and $PM_{2.5-10}$, while in *V. lucidum,* the planting density did not affect the depositions.

Black PM_{10-100} decreased a long distance from the road and was entirely composed of carbon and oxygen, which was thus identified as black carbon from fuel combustion.

The vegetation barrier had a higher deposition of most PM fractions at 5.5-12.5 m, while in the lawn area, depositions did not change. At 19.5 m, the PM_{10-100} was 32% lower behind

the barrier than in the lawn area. In conclusion, the vegetation barrier changed the deposition dynamics of pollutants compared to the lawn area. These results strengthen the role of vegetation barriers and shrub species against air pollution and may offer interesting insights for the use of new road green infrastructures to improve air quality.

Keywords: green infrastructure, particulate matter, leaf deposition, element, microscopy, image analysis

1. Introduction

Urban air pollution is a threat for human health, causing nearly seven million premature deaths per year throughout the world (WHO, 2014). Air quality is particularly poor in urban areas because of the high level of emissions by anthropogenic activities (Sawidis et al., 2011). Its negative impact is exacerbated by the fact that 54% of the world's population live in these areas (United Nations, 2014). In Europe, it has been estimated that in 2015, 53% and 82% of urban populations were exposed to higher than WHO's recommendations for daily levels of coarse (PM_{10}) and fine ($PM_{2.5}$) particulate matter (EEA, 2017).

Particulate matter is the most abundant air pollutant, consisting in a mixture of heavy metals, elements, black carbon, soil and other substances (Thurston, 2008; Bell et al., 2011). Improvement of air quality through the reduction of air pollutants is now a widely accepted ecosystem service provided by urban vegetation (Simon et al., 2011; Sæbø et al., 2012; Janhäll, 2015). The dry deposition of solid particles (hereafter, deposition) on leaf surfaces is the basic mechanism of the beneficial action of plants on air quality (Bealey et al., 2007; Nowak et al., 2013).

Leaf deposition is influenced by several factors such as the characteristics of particles (concentration, diameter, composition, etc.), the vegetation (foliage density, porosity to air fluxes, plant height, leaf characteristics, etc.), and the site (proximity of vegetation to pollution source, meteorological conditions etc.) (Fowler et al., 2003; Freer-Smith et al., 2005; Litschke and Kuttler, 2008; Petroff et al. 2008). The deposition of particles with a diameter above one micrometer has been found to increase at higher air particle concentrations and at increasing particle diameters (Fowler et al., 2003).

Increased foliar density leads to higher leaf deposition although very high density can reduce the porosity of the canopy, thus limiting the air flux through the canopy and particle deposition (Tiwary et al., 2005; Baldauf, 2017). Plant height influences air quality in different ways depending on the characteristics of the planting site.

In open areas adjacent to roads, the improvement in air quality is more effective when the canopy is taller than the impacting dust plume from traffic which in proximity to the road can be around two meters (Etyemezian et al., 2003; Etyemezian et al., 2004). On the other hand, in street canyons, trees with a large and thick canopy can also increase air pollution concentrations at the pedestrian level because of a reduction in air pollutant dispersion (Buccolieri et al., 2009).

The proximity of vegetation to the pollution source is another important factor increasing leaf deposition (Pugh et al., 2012) which can limits the dispersion of pollutants to the surrounding environment. Leaf deposition is also influenced by meteorological conditions, for example rainfall can wash off leaf deposition (Nowak et al., 2006) to the ground, thus reducing the possibilities of re-suspension in the atmosphere. High wind speed increases not only deposition but also the re-suspension rate of deposited particles (Beckett et al., 2000; Fowler et al., 2003).

With regard to leaf traits, evergreen plants are able to intercept higher quantities of pollutants compared to deciduous species, especially during the winter season when the concentration of air pollutants is generally higher (Pikridas et al., 2013). Among deciduous species, plants with a longer in-leaf period are usually more effective. Species with smaller leaves and more complex shoot structures are more efficient at capturing PM (Freer-Smith et al., 2005). In addition, the presence of hairs and waxes has been associated with a higher particulate deposition (Sæbø et al., 2012).

Several species, mostly trees native to northern environments (Aničić, et al., 2011; Popek et al., 2017), have been investigated in terms of their capacity to remove air pollutants (Dzierzanowski et al., 2011; Sæbø et al., 2012). However there have been few studies on shrub species in southern Europe (Lorenzini et al., 2006; Mori et al., 2016).

Shrubs may represent a sound alternative to trees in reducing air pollution, especially in contexts where tree planting is not possible because of a lack of space or due to law enforcement (e.g. a ban on planting trees in proximity to roads) or because they are detrimental to air quality (Buccolieri et al., 2009; Jeanjean et al., 2017).

Finally, the capacity of plants to reduce air pollution is influenced by the kind of green infrastructure in which the vegetation is located (Abhijith et al., 2017). Roadside vegetation barriers (hereafter, vegetation barriers) are green infrastructures consisting in rows of shrubs and/or trees planted along roads (Abhijith et al., 2017; Baldauf, 2017). They screen people living in neighboring areas from the drift of air pollutants from linear traffic (Al-Dabbous and Kumar, 2014; Lin et al., 2016). The impact of vegetation barriers on air quality has been assessed by experimental (Hagler et al., 2012) and modeling approaches (Morakinyo and Lam, 2016) with contrasting results. While in the experimental study, no clear effects of a roadside vegetation barrier on air quality were found, in the modeling approach, beneficial influences of vegetation barriers were simulated.

Baldauf (2017) reviewed the characteristics of vegetation barriers that can increase the capacity of air pollution mitigation. The height from the ground of vegetation barriers should be higher than the initial dust plume derived from traffic; the recommended value for this parameter is 4-5 m (Baldauf, 2017). The thickness of the vegetation barrier should vary between 5 to 10 m and even more (Neft et al., 2016) depending on the foliar density. An appropriate combination of thickness and foliar density should enable the suspended particles to remain within the vegetation for a sufficient time to permit their removal from the air while limiting air blocking (Neft et al., 2016). The vegetation coverage is also crucial, given that pollutant fluxes can preferentially pass through vegetation gaps (e.g., spaces between plants) instead of through the vegetation barrier (Baldauf, 2017). In such conditions, the effect of the vegetation barriers may be negatively affected. Finally, at least a 50-m-length vegetation barrier is recommended in order to prevent pollutant fluxes from passing laterally to the vegetation instead of passing through it (Baldauf, 2017).

Besides this growing body of research, more investigations are needed to better understand and quantify the effects of roadside barriers on air pollution (Abhijith et al., 2017). There is a lack of experimental data regarding the interaction between vegetation barriers composed of evergreen shrubs, with different planting densities, and air pollutants in the Mediterranean environment. A direct comparison between areas with and without vegetation barriers thus may represents a novel approach.

In a previous study by our research group (Mori et al., 2015a), six evergreen shrub species were characterized for the leaf deposition of PM and 21 elements mainly from traffic in a peri-urban environment during a Mediterranean summer season.

Three out of six species (*Viburnum lucidum* L., *Photinia* × *fraseri* cv. Red Robin Dress and *Elaeagnus x ebbingei* L.) were found to be more suitable for air pollution deposition because of the higher growth (in terms of leaf area, leaf area index, plant biomass and more favorable leaf traits) compared with the other species tested.

The aims of the present work was to study leaf deposition of three PM fractions (PM_{10-100}, $PM_{2.5-10}$ and $PM_{0.2-2.5}$) and of 21 elements (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Ni, Pb, Sb, Se, Tl, V, Zn) in a vegetation barrier. In this study the barrier had similar characteristics to those recommended in the literature in terms of height, length, thickness and coverage (Baldauf, 2017). Several factors were studied: i) two different planting densities ii) two species, iii) three distances from the road, iv) two heights from the ground and v) three different sampling periods. *V. lucidum* L. and *Photinia* × *fraseri* Dress were chosen as test plants based on our previous results (Mori et al., 2015a).

The influence of the vegetation barrier in the experimental area was also investigated by direct comparison with an adjacent area characterized by lawn vegetation and by measuring the deposition in i) both areas, in ii) three different sampling periods, and at iii) five different distances from the road.

2. Materials and Methods

2.1 Experimental field

The experiment was conducted in Pescia, Tuscany, Italy (43°52'57.7992''N, 10°40'58.0692''E), (Fig. 1) in an experimental site, 35 km from the Tyrrhenian sea.

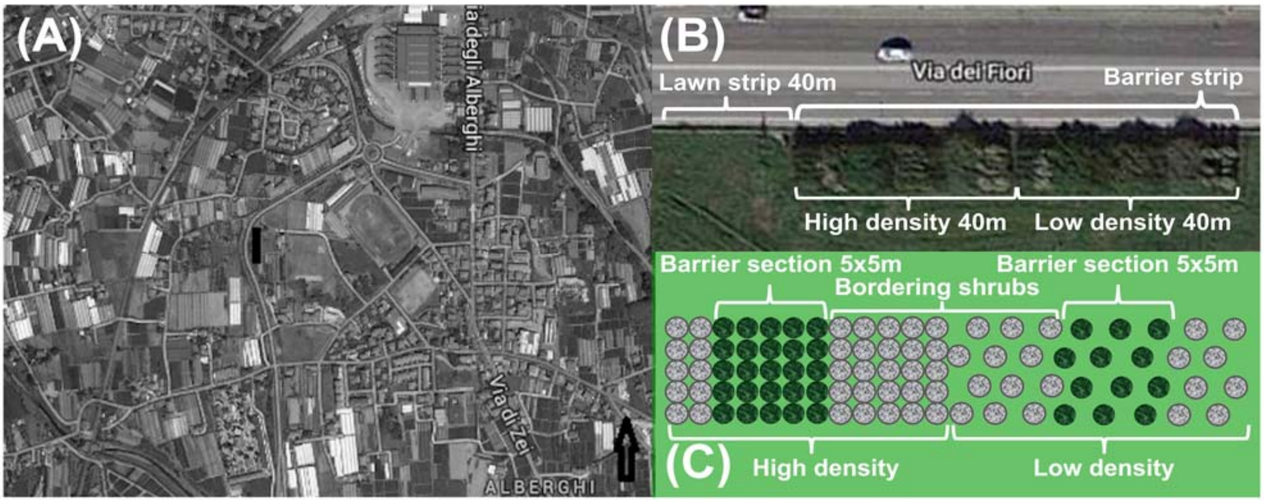


Fig. 1. Image of the area surrounding the experimental site taken from Mori et al. (2015a). The orientation of the vegetation barrier is indicated with a dark rectangle, while the black arrow indicates north (A). Aerial picture taken from Google Maps of the experimental area comprising the barrier strip and the lawn strip (B). A part of the vegetation barrier with two barrier sections (dark green circles) and the interposed shrub vegetation (light grey circles) with the two different planting densities (C).

Annual rainfall is 901 mm, and minimum and maximum temperatures are 9.17°C and 19.8°C, respectively (thirty-year average data). Meteorological conditions during the experiment (i.e. rainfall, wind speed, wind direction, relative humidity, and air temperature), were monitored from 1 July 2016 (one month before the start of the sampling campaign) to 3 October 2016 (end of sampling campaign) using an in-situ meteorological station iMetos® SD (Pessl Instruments, Weiz, Austria). The experimental field was located along a 4-lane road which is the main access for vehicles to the center of the town. Traffic was sampled manually similarly to Irga et al. (2015): vehicles passing in front of the barrier from 7:00 a.m. to 08:00 p.m. were counted manually on three different mid-week days.

The land use in the area was assessed as proposed by Irga et al. (2015), using satellite images taken from Google maps within 100 m, 250 m, and 500 m radii from the experimental site, which corresponded to 3.14, 19.6 and 78.6 ha, respectively. Images were then analyzed with WinDIAS software (Dynamax Inc., USA) differentiating between land surfaces occupied by buildings, streets, greenhouses, and green areas. Along the road, in proximity to the study area, only agricultural activities (greenhouses and cultivated fields) were present.

2.2 Experimental design

The vegetation barrier was parallel and downwind to the road with the predominant wind blowing from the coast striking both the road and the vegetation barrier orthogonally (Fig. 1). In addition, the absence of obstacles between the vegetation barrier and the road, together with a lack of road crossings or roundabouts near the vegetation barrier were other important characteristics to ensure the uniformity of the experimental site in terms of exposure to air pollution.

In March 2010, fifty 3-year-old *P. x fraseri* cv. Red robin and of *V. lucidum* plants (100 plants in total) were planted to form four different transects, of a vegetation barrier, as previously described by Mori et al. (2015a).

The vegetation barrier was planted 2 m away from the road. Each barrier transect was composed of 25 three-year-old plants per species, occupying an area of 5 by 5 m (with a total of 100 plants per 100 m^2). In winter 2015 (start of experiment), in one barrier transect per species (2 out of 4 barrier transects), the density of plantation was homogeneously reduced to 0.5 plant m^{-2} (Fig. 1). On the same date, plants were cut back to a height of 3.5 m and laterally pruned in order to separate the plant crowns from each other. No other pruning was carried out during the experiment in order to prevent any possible influence on foliar deposition (see later).

The height of plants was generally higher than the initial dust plume from traffic (2 m), (Etyemezian et al., 2003) and similar to those recommended by Baldaf (2017). The heights of all plants were measured again at the end of the experiment. The sides of each barrier transect that were parallel to the road (roadside and opposite-roadside) were free from any physical obstacles, while orthogonal roadsides verged with other barrier transects made up of shrubs of an equal height (3.5 m) compared with the studied species. The aim of this arrangement was to limit the intrusion of air fluxes laterally to the barrier transects and to favor air passing from the road within the barrier. The length of the total vegetation barrier (studied and non-studied barrier transects) was 80 meters in line with the recommendation reported by Baldauf (2017). In addition, due to the homogeneous and staggered distribution of plants within the different barrier transects, no free gaps were present.

A lawn area behind (9.0 to 19.5 m from the road) and adjacent (2.0 to 19.5 m from the road) to the vegetation barrier was also considered for measurements. The height of the grass in the lawn area was maintained below 20 cm tall throughout the experiment in order to not interfere with the passive samplers (see below). The area from 2.0 m to 19.5 m from the road in which the vegetation barrier was present was defined as the "barrier strip", while the area entirely characterized by lawn vegetation was defined as the "lawn strip".

The average leaf area index (LAI) of the various barrier transects was monitored using a ceptometer (AccuPAR LP-80, Decagon Devices, Inc., WA, USA). Data were recorded at 1 m height, positioning the linear optical sensor of the instrument parallel to the ground below each plant and between plants in two different orthogonal directions, following the ceptometer manual.

The total leaf area of the barrier transects was estimated by multiplying the mean LAI of each transect (2 species x 2 planting densities) by the corresponding projection of the canopy. Passive samplers (30) were placed in the barrier and in the lawn strip, at 2.0, 5.5, 9.0, 12.5 and 19.5 m from the road in order to collect PM_x and element deposition. The passive samplers were positioned: i) directly exposed to traffic (2.0 m from the road), ii) in the middle of the vegetation barrier (5.5 m from the road), iii) behind the barrier (9.0 m from the road), iv) as position (iii) plus the barrier height (9.0 + 3.5 = 12.5 m from the road), as position (iii) plus 3 times the barrier height (9.0 + 10.5 = 19.5 m from the road). The passive samplers were created using a wooden rod (1.5 m length) as a vertical support on which a rigid plastic plate (25 x 15 cm) was horizontally mounted. Two polytetrafluoroethylene membranes (PTFE) (i.e., one used to collect PM_x deposition and one used for the element deposition) were placed on the central part of the plastic plate. The stability of the PTFE membrane was insured by four PTFE screws and washers, which were positioned with millimetric precision thereby covering the same surface of membranes among samplers.

A fiberglass sheet (50 x 20 cm) was mounted on the top of the passive samplers by fixing the short sides of the sheet on the reciprocal sides of the plastic plates to protect the PTFE membranes from rainfall, while ensuring the passage of air flows in the orthogonal direction with respect to the longer side. Passive samplers were then positioned with the long side of the plastic plate parallel to the road. The distance (in height) between the fiberglass sheet and the PTFE membranes was 20 cm. Passive samplers were positioned in order to maintain PTFE membranes 1 meter from the ground, which was deemed a good compromise for collecting pollutant deposition at ground level while limiting contamination from the soil.

A preliminary test was performed to verify deposition homogeneity among samplers during a one-month period of exposure to traffic, 2.0 m from the road, in the same experimental location.

To determine the composition in elements, approximately 500 g of topsoil were collected from the 0 – 10 cm soil layer at five different distances from the road (2.0 m, 5.5 m, 9.0 m, 12.5 m, 19.5 m) in both the barrier and lawn strips (three replicates for a total of 30 samples). During sampling, any visible organic debris that could potentially affect the concentration of elements in the soil was avoided (Skrbìc et al., 2012). To determine the element concentration, topsoil samples were processed following the protocol reported in Mori et al., (2015a).

2.3 PM_x and elements deposition – sampling

Leaf samples of roughly 400 cm^2 each (Dzierzanowski et al., 2011; Saebo et al., 2012) were collected in two replicates as reported in Tab. 1.

Tab. 1. Type of collected samples, fixed factors per kind of collected sample, number of levels per fixed factor, number of samples per level of fixed factor, and description of the levels of each fixed factor.

Type of sample (variable factors)	Fixed factors	Levels (N°)	Samples per level (N°)	Description
Leaf deposition (PM_x and elements)	Species	2	72	*V. lucidum; P. x fraseri*
	Planting density	2	72	0.5 plants m-2; 1.0 plants m-2
	Distance from the road	3	48	2.0 m; 5.5 m; 9.0 m
	Height from the ground	2	72	1.5 m; 3.0 m

	Sampling dates	3	48	01 Ago 2016; 01 Sept 2016; 03 Oct 2016
Deposition on passive samplers (PM_x and elements)	Experimental strips	2	45	Barrier strip; lawn strip
	Distance from the road	5	18	2.0 m; 5.5 m; 9.0 m; 12.5 m; 19.5 m
	Sampling dates	3	30	01 Ago 2016; 01 Sept 2016; 03 Oct 2016
Percentage of filter surface occupied by PM_{10-100}	Species	2	216	*V. lucidum; P. x fraseri*
	Planting density	2	216	0.5 plants m-2; 1.0 plants m-2
	Distance from the road	3	144	2.0 m; 5.5 m; 9.0 m
	Height from the ground	2	216	1.5 m; 3.0 m
	Sampling dates	3	144	01 Ago 2016; 01 Sept 2016; 03 Oct 2016
	Color of particles	2	216	Black; brown

A total of 144 samples were collected for PM_x and another 144 for elements, based on previous findings reported by Mori et al., 2015a. The sampling period (01 August 2016 - 03 October 2016) was based on previous experience (Mori et al., 2015a) and in particular on the need to carry out measurements both during the dry summer season and the start of the rainy season (autumn). This enabled us to verify the influence of planting density, species, height of leaves etc. under different climatic conditions. Samples of the current season and healthy leaves were collected maintaining a distance of at least 1.5 m from the adjacent barrier transect in order to reduce contamination/side effects. However, to limit contamination of samples, samples were collected using disposable gloves and harvesting leaves from the petioles to prevent contact with the leaf blade. After sampling, leaves were placed in disposable paper bags before being processed in the laboratory.

PTFE membranes were sampled on the same days as leaves, with a total of 90 membranes for PM_x and another 90 membranes for elements collected throughout the entire experiment (Tab. 1). PTFE membranes were collected and replaced on the same day using plastic tweezers. Once removed, each PTFE membrane was placed into a 50 ml plastic tube. Each sampling was carried out at least 10 days after the last rainfall.

2.4 PM_x deposition - determination

The original protocol by Dzierzanowski et al. (2011) for PM_x leaf deposition analysis was modified as follows. Three different filters with different retention capacities were used (type 91 - retention 10 µm, type 42 – retention 2.5 µm and PTFE membrane - retention 0.2 µm), (Whatman, UK). Each filter was firstly dried at 60°C for 30 minutes in a KCW-100 drying chamber (PREMED, Poland) and then left for 60' at a constant relative air humidity (50 %) for weight stabilization. Filters were then pre-weighed on BCA 2005 scales (Orma s.r.l., Italy). Each leaf sample was washed for 60'' with 150 ml of deionized water under agitation. The washing solution was then filtered through a metal sieve (Haver and Boecker, Germany) in order to eliminate particles larger than 100 µm, and then sequentially filtered using the three filters in the increasing order of retention capacity (10 µm, 2.5 µm, 0.2 µm).

Filtration was carried out using an apparatus equipped with a 47 mm glass filter funnel (Scharlau, Scharlab, S.L., Barcelona, Spain) connected to a MV-50 vacuum pump (Comecta-Ivymen, Spain). Immediately before filtration, PTFE membranes were moistened with a few droplets of isopropyl alcohol to break the surface forces and speed up the process. After filtration, filters were dried, left to stabilize and then weighed again.

At the end of the entire filtration procedure, three fractions of PM_x were collected: (1) large: 100-10 µm (PM_{10-100}), (2) coarse: 10–2.5 µm ($PM_{2.5-10}$), and (3) fine: 2.5–0.2 µm ($PM_{0.2-2.5}$). After washing, the leaf area of each sample was measured using an RS 2 XA illumination and camera system (Haiser, Germany) and an A3 Lightbox (G.C.L., UK) with WinDIAS software (Dynamax Inc., USA). The protocol for PM_x deposition in the barrier and lawn strips was the same as the one used for leaf samples, except for the washing phase of PTFE membranes which was carried out directly in the 50 ml plastic tube used for collection using 50 ml of deionized water. The results of PM_x deposition on leaves and in the barrier and lawn strips were expressed per unit area of leaf and PTFE membranes, respectively. On the same day, one blank (consisting of 150 ml for leaf samples, and 50 ml for PTFE membrane samples) every 20 samples, was filtered with the same protocol as outlined above. The blank values were negligible.

2.5 Element deposition - determination

Leaf samples were washed using the same protocol as for PM_x determination, but only filtration with the metal sieve (Haver and Boecker, Germany) was carried out. The washing solution (150 ml) was collected in a beaker and reduced up to 10 ml at 70°C. The concentrated solution was then digested using a heating digester (DK20,Velp® Scientifica, Italy), at 170◦C per 120 min adding 12 ml of a mixture of HNO_3 (65%) and H_2O_2 (30%) (10:2) V/V. A water-cooling system was applied to reduce the loss of elements by evaporation and to prevent samples from drying. Finally, the volume was adjusted to 25 ml with deionized water.

The concentration of 21 elements (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Ni, Pb, Sb, Se, Tl, V, Zn) was determined by an Inductively Coupled Plasma – Optical Emission Spectrometer – (Optima 7000, Perkin Elmer, Massachusetts, U.S.). PTFE membranes were processed with the same protocol used for leaves, apart from the volume of deionized water used for washing (50 ml). The protocol used for the digestion phase, reported by Mori et al. (2015a; 2015b), was modified in terms of the temperature and duration. One blank (consisting of 10 ml of reduced deionized water) every 20 samples, was digested with the same aforementioned protocol, on the same day of analysis. Blank values were negligible.

The results of the element deposition were expressed per unit area of leaf surface or PTFE membrane surface. Total leaf deposition of PM_x and elements on vegetation barrier transects (two species for two different planting densities), were calculated by multiplying the mean leaf deposition per unit leaf surface of each transect by the corresponding total leaf area.

2.6 Microscope analysis

From each paper filter (type 91) used for the analysis of PM_{10-100} leaf deposition (see 2.4 section), three images were taken at 10x magnification, using an optical confocal microscope (Axioskop 40, Zeiss, Germany) equipped with an Axiocam camera (MR 5, Zeiss) and Axiovision Rel.4.6 software (Zeiss) with a total number of 432 images (Tab. 1). Images were then analyzed using an RS 2 XA illumination and camera system (Haiser, Germany) and A3 Lightbox (G.C.L., UK) with WinDIAS software (Dynamax Inc., USA) in order to differentiate between PM_{10-100} with different colorations (black or brown), and to calculate the relative percentage of paper filter surface occupied by different colored PM_{10-100}.

Nine PM_{10-100} paper filters, already used for optical microscope observations, representative of different sampling periods and distances from the road, were observed using a Quanta 200, Environmental Scan Electronic Microscope (ESEM), FEI Thermo Fisher Scientific (Oregon, USA), at 80–400x magnification. In addition, to investigate the element composition, X-ray microanalysis was carried out on two black and two brown particles per filter by an EDS-X-ray microanalysis system (EDAX Genesis, AMETEK, Mahwah, NJ, USA). Observations of the leaf surfaces of the two species were also carried out with ESEM in order to assess the presence of waxes and trichomes on the upper and lower leaf surface. Only current season and fully expanded leaves were considered for the above analysis.

2.7 Data analyses and statistics

Multi-way ANOVA was applied to i) data of leaf deposition; ii) barrier and lawn strip deposition and; iii) percentages of PM_{10-100} filter surfaces occupied by black and brown particles. The number of fixed factors (and thus of the number of ways of the ANOVA), per measured variable are reported in Tab. 1. Significant interactions between factors were subsequently investigated by one-way ANOVA. Mean values were separated by Tukey's

(HSD) post-hoc test ($P \leq 0.05$). The outliers were identified by a Box and Wisker plot, and removed from the dataset.

A correlation analysis between the leaf deposition and climatic parameters (air temperature, wind speed and precipitations) was performed using Pearson product–moment coefficients..

Principal component analysis (PCA) was performed in order to: i) group the studied air pollutants (PM_x and elements on leaves) explaining most of the experimental variability, and ii) obtain a graphical representation of the influence of the various factors (species, sampling date, planting density) in inducing variance within the model. The analysis was performed on the basis of a correlation matrix as suggested by (Catoni and Gratani, 2014). Each individual air pollutant was assigned to a different group on the basis of the principal component with the highest component weight in absolute value. All the statistical analyses were carried out using Statgraphics Centurion XV (Stat Point, Inc., Herndon, VA, USA), while the graphics were drawn with SigmaPlot 11.0, Systat Software Inc. (San Jose, California, USA).

3. Results

3.1 Experimental field

The meteorological data recorded throughout the entire experiment were representative of the typical Mediterranean climate, with high temperatures and almost no rainfall between July and August, followed by an increase in rainfall events and a decline in air temperature and in wind speed from September to October (Tab. 2A). The surrounding land was mainly used for agriculture and green areas in general (59.5%) (Tab. 2B), roads and parking ranked second with 29.8%, while buildings occupied the remaining 10.7%. A mean number of 10,844 vehicles per day passed on the road adjacent to the vegetation barrier between 7:00 a.m. and 08:00 p.m. Similarly to Hagler et al. (2012), and Irga et al. (2015), the fleet mix was not quantified for this study. Data regarding element composition in topsoil (data not shown) were similar to those from the same area reported in Mori et al. (2015a). In addition, no differences between the barrier and lawn strip and between distances from the road were found with the exception of Ca, which decreased significantly at increasing distances (data not shown).

Tab. 2 Meteorological data at the experimental area averaged in the different sampling periods (A). Land use partitioning (%) within 100, 250 and 500 m radii distant from roadside vegetation barrier (B).

(A) Period	Air temperature (°C)	Rainfall (mm)	Wind velocity (m s^{-1})	Wind direction	Air RH (%)
1 Jul – 1 Aug 16	24.25	1.6	0.88	WSW	57.62
2 Ago – 1 Sep 16	23.63	3.2	0.81	SW	55.24
2 Sep – 3 Oct 16	20.71	24.3	0.45	WSW	65.57
(B) Radii distance (m)	**Green areas and greenhouses (%)**		**Buildings (%)**	**Roads and parking (%)**	
100	68.3		4.8	26.9	
250	61.5		9.6	28.9	
500	48.7		17.6	33.7	

3.2 Plant growth

Planting density affected LAI in the barrier transects with *P. x fraseri* (*P*<0.001), with larger LAI developed at the higher planting density (8.07 ± 1.08 SD) compared with the lower planting density (4.82 ± 2.11 SD), but had no significant effect on *V. lucidum* (0.5 pt m^{-2} = 7.68 ± 1.73 SD; 1.0 pt m^{-2} = 8.72 ± 1.45). The height of plants increased on average by 8.5% from winter 2015 to the end of the experiment, without significant differences between species and planting densities (data not shown).

3.3 PM_x deposition per unit leaf area

In *P. x fraseri*, $PM_{2.5-10}$ and $PM_{0.2-2.5}$ leaf depositions were lower compared with *V. lucidum,* while no differences were detected for the other PM fractions (Fig. 2A). Leaf deposition of PM_{tot}, PM_{10-100} and $PM_{2.5-10}$ at 3.0 m height from the ground were 43.7%, 49.8% and 23.2%, lower than the deposition at 1.5 m (Fig. 2B). PM_{tot} and PM_{10-100} leaf deposition increased by 31.5% and 26.7%, respectively, from August to September and then decreased in October by 28.8% and 39.1% (Fig. 2C). On the other hand, $PM_{2.5-10}$ leaf deposition did not change significantly from August to September but increased by 17.9 % from September to October. $PM_{0.2-2.5}$ decreased, on average, by 40.5% from August to October. PM_{tot} and PM_{10-100} leaf deposition decreased by 25.0% and 26.3% respectively from 2.0 to 9.0 m from the road, while no differences for the smaller fractions were found (Fig. 2D). Higher planting density in *P. x fraseri* increased PM_{10-100} and $PM_{2.5-10}$ leaf deposition by 13.8% and 38.3%, respectively, compared with the lower planting density, while no other differences were found for the other fractions in either species (Fig. 2E, 2F).

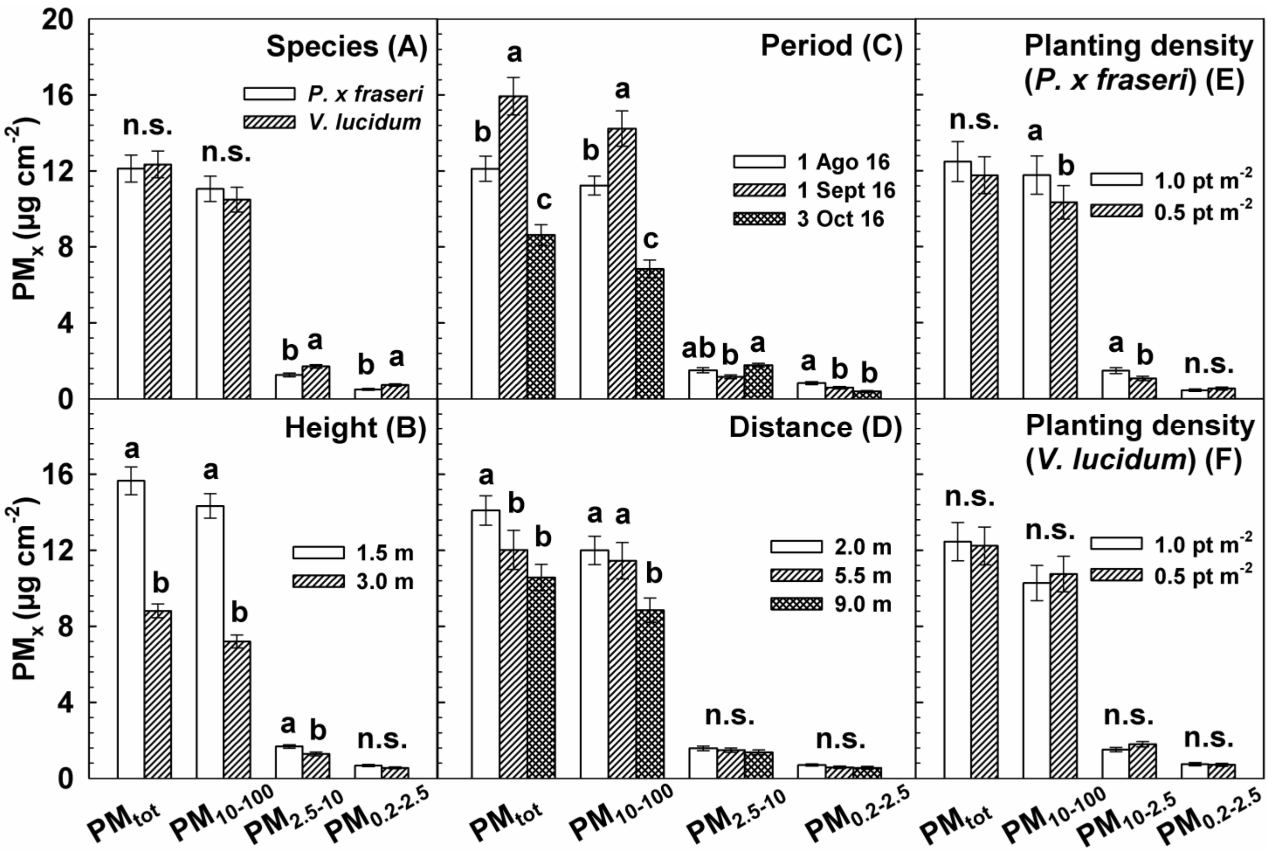


Fig. 2. Leaf deposition per unit leaf surface of different fractions of PM_x for: (1A) two different shrub species (*P. x fraseri* and *V. lucidum*) (n = 72), (1B) two different heights from the ground (1.5 m and 3.0 m) (n=72), (1C) three different sampling periods (01 Aug 2016, 1 Sep 2016 and 03 Oct 2016) (n = 48), (1D) three different distances from the road (2.0 m, 5.5 m and 9.0 m) (n = 48), (1E-F) two different planting densities (0.5 plants m^{-2} and 1.0 plants m^{-2}) in (E) *P. x fraseri* (n = 36) and in (F) *V. lucidum* (n = 36). Columns are means (μg cm^{-2}) ± SE. Different letters indicate significant differences for the same PM_x fraction at P = 0.05 using Tukey's (HSD). n.s. indicates no significant difference.

3.4 Element deposition per unit leaf area

Lead, Sb, Se and Tl were not detected in most leaf samples and were removed from the dataset (Tab. 3). The other elements were found in the following rank order: Ca > Fe > Al > Mg > K > Mn > Zn > Cu > Ba > V > As > Ni > Cr > Li > Mo > Co > Cd. *P. x fraseri* showed a higher leaf deposition in 13 out of 17 elements compared with *V. lucidum,* while no significant differences were found for Al, Cr, Cu and Zn (Tab. 3). Leaf deposition of all elements changed between sampling periods apart from Mn. Twelve out of 17 elements (Al, Ba, Co, Cr, Cu, Fe, K, Li, Mg, Ni, V, Zn) showed an increase from Aug to Sep, and thereafter a decrease in Oct (Tab. 3), whereas As, Cd, and Mo increased during the sampling periods (Tab. 3). Planting density influenced leaf deposition in 10 out of 17 elements, thereby increasing Al, Ba, Ca, Cr, Cu, Fe, K, Mg, and V (Tab. 3). The distance from the road had an effect on leaf deposition in 12 out of 17 elements, with generally higher values at 2.0 m from the road compared with values at 9.0 m (Tab. 3). The height from the ground affected leaf deposition in 14 out of 17 elements. In 12 cases (Al, Ba, Ca, Co, Cr, Cu, Fe, K, Mg, Ni, V, Zn), values were higher at 1.5 m compared with 3.0 m, while for As and Cd an opposite trend was observed (Tab. 3).

Tab. 3 Leaf deposition per unit leaf surface of different elements (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Ni, V, Zn) for: i) two different shrub species (*P. x fraseri* and *V. lucidum*) (n = 72), ii) three different sampling periods (01 Aug 16, 1 Sep 16 and 03 Oct 16) (n = 48), iii) two different planting densities (0.5 plants m^{-2} and 1.0 plants m^{-2}) (n = 72), iv) three different distances from the road (2.0 m, 5.5 m and 9.0 m) (n = 48), v) two different heights from the ground (1.5 m and 3.0 m) (n = 72). Values are means (μg cm^{-2}).

	Al	As	Ba	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Mo	Ni	V	Zn
Species (Sp)																	
P. x fraseri	0.20 a	0.0013 a	0.0049 a	0.67 a	0.00020 a	0.00029 a	0.0010 a	0.0082 a	0.25 a	0.16 a	0.00094 a	0.21 a	0.02 a	0.00033 a	0.0012 a	0.0017 a	0.020 a
V. lucidum	0.18 a	0.0011 b	0.0042 b	0.59 b	0.00018 b	0.00026 b	0.0009 a	0.0073 a	0.23 b	0.12 b	0.00080 b	0.16 b	0.03 b	0.00030 b	0.0011 b	0.0015 b	0.0208 a
Period (Pd)																	
01 Ago 16	0.21 b	0.0011 b	0.0041 b	0.74 a	0.00017 b	0.00028 b	0.0009 b	0.0058 b	0.25 b	0.13 b	0.00084 b	0.20 b	0.02 a	0.00029 b	0.0011 b	0.0018 b	0.0209 b
01 Sept 16	0.25 a	0.0012 b	0.0063 a	0.81 a	0.00019 a	0.00030 a	0.0012 a	0.0109 a	0.33 a	0.17 a	0.00095 a	0.23 a	0.03 a	0.00032 ab	0.0013 a	0.0022 a	0.0250 a
03 Oct 16	0.10 c	0.0013 a	0.0033 c	0.34 b	0.00020 a	0.00024 c	0.0007 c	0.0065 b	0.15 c	0.11 c	0.00082 b	0.12 c	0.02 a	0.00034 a	0.0010 b	0.0008 c	0.0164 c
Density (Dt)																	
1.0 pt m^{-2}	0.21 a	0.0012 a	0.0050 a	0.66 a	0.00018 a	0.00028 a	0.0010 a	0.0079 a	0.26 a	0.14 a	0.00088a	0.19 a	0.02 a	0.00031 a	0.0012 a	0.0017 a	0.0212 a
0.5 pt m^{-2}	0.18 b	0.0012 a	0.0042 b	0.60 b	0.00018 a	0.00027 a	0.0009 b	0.0076 b	0.22 b	0.14 b	0.00086 a	0.18 b	0.03 a	0.00031 a	0.0011 a	0.0015 b	0.0203 a
Distance (Dc)																	
2.0 m	0.20 a	0.0012 a	0.0048 a	0.77 a	0.00020 a	0.00029 a	0.0011 a	0.0092 a	0.27 a	0.13 a	0.00090 a	0.21 a	0.02 a	0.00034 a	0.0012 a	0.0017 a	0.0261 a
5.5 m	0.20 a	0.0012 a	0.0047 a	0.62 b	0.00018 a	0.00028 ab	0.0010 a	0.0079 b	0.26 a	0.14 a	0.00086 b	0.18 b	0.02 a	0.00031 ab	0.0011 ab	0.0016 a	0.0210 b
9.0 m	0.17 b	0.0012 a	0.0041 a	0.50 c	0.00019 a	0.00026 b	0.0008 b	0.0061 c	0.20 b	0.14 a	0.00085 b	0.17 b	0.02 a	0.00030 b	0.0011 b	0.0014 b	0.0151 c
Height (Ht)																	
1.5 m	0.25 a	0.0011 b	0.0056 a	0.80 a	0.00018 b	0.00030 a	0.0012 a	0.0098 a	0.33 a	0.16 a	0.00088 a	0.23 a	0.03 a	0.00031 a	0.0012 a	0.0021 a	0.0245 a
3.0 m	0.14 b	0.0013 a	0.0035 b	0.46 b	0.00020 a	0.00025 b	0.0007 b	0.0057 b	0.16 b	0.12 b	0.00086 a	0.14 b	0.02 a	0.00033 a	0.0010 b	0.0011 b	0.0170 b
Significance*																	
Sp	0.085	0.001	0.009	<0.001	0.002	<0.001	0.422	0.005	0.027	<0.001	<0.001	<0.001	0.046	<0.001	0.004	<0.001	0.922
Pd	<0.001	0.001	<0.001	<0.001	0.004	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.184	0.002	<0.001	<0.001	<0.001
Dt	<0.001	0.867	0.002	0.010	0.693	0.393	0.004	0.421	<0.001	0.178	0.157	0.007	0.061	0.919	0.159	<0.001	0.473
Dc	0.001	0.770	0.077	<0.001	0.230	0.001	<0.001	<0.001	<0.001	0.379	0.050	<0.001	0.837	0.020	0.001	<0.001	<0.001
Ht	<0.001	<0.001	<0.001	<0.001	0.012	<0.001	<0.001	<0.001	<0.001	<0.001	0.234	<0.001	0.075	0.031	<0.001	<0.001	<0.001
Sp x Dc	0.378	0.082	0.772	0.578	0.300	0.248	0.900	0.440	0.318	0.458	0.078	0.102	0.024	0.340	0.349	0.134	0.282

Sp x Dt	0.244	0.767	0.086	0.143	0.953	0.791	0.380	0.549	0.566	0.170	0.727	0.164	0.005	0.909	0.769	0.221	0.201

*P-Values are reported for all factors and for interactions. Different letters indicate significant differences within the same factor and element at P = 0.05 using Tukey's (HSD).

3.5 Total leaf deposition per barrier transect

The barrier transects composed of *P. x fraseri* had a lower total leaf deposition compared with *V. lucidum* in 12 out of 17 elements (Al, As, Cd, Co, Cr, Cu, Fe, Li, Mn, Mo, Ni, Zn), in PM_{tot}, $PM_{2.5-10}$ and in $PM_{0.2-2.5}$, while for the other air pollutants, no significant differences were found (Tab. A). A higher planting density increased total leaf deposition in all cases apart from Mn and $PM_{0.2-2.5}$ (Tab. A). Generally, *P. x fraseri* at 0.5 pt m^{-2} showed the lowest leaf deposition in most elements, in PM_{tot} and in PM_{10-100} (Tab. A), while planting density in *V. lucidum* did not show a clear effect on leaf deposition.

3.6 PM_x deposition in the barrier and lawn strip

PM_x deposition decreased from August to October (PM_{tot} = -65 %; PM_{10-100} = -56 %; $PM_{2.5-10}$ = -81 %; $PM_{0.2-2.5}$ in lawn strip = -76 %), except for $PM_{0.2-2.5}$ in the barrier strip which did not vary over time (Fig. 3A, B). The distance from the road significantly influenced deposition depending on the PM_x fraction and strip type (Fig. 3C, 2D). In the barrier strip, the deposition of PM_{tot}, PM_{10-100} and $PM_{2.5-10}$ showed a non-linear trend, increasing from 2.0 m to 12.5 m and decreasing thereafter with significantly lower values at a 19.5 m distance (Fig. 3C). The deposition in the lawn strip however, showed a linear trend without any significant reduction (Fig. 3D). The barrier strip at 19.5 m showed a 31% and 32% lower deposition for PM_{tot} and PM_{10-100} respectively, compared with the lawn strip at $P<0.05$, while no other differences between the same distance in the different strips were found.

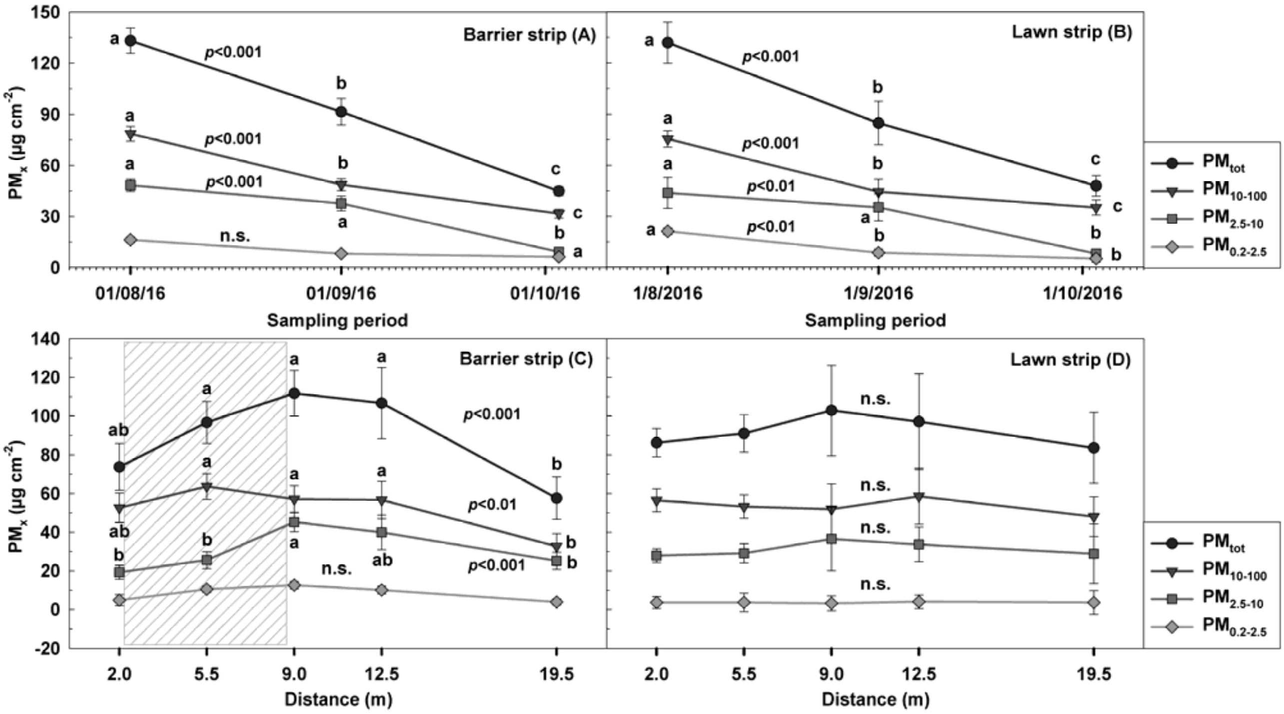


Fig. 3. Deposition in the barrier (A) and lawn (B) strip of different PM_x fractions per unit PTFE membrane area in three different sampling periods. Deposition in the barrier (C) and lawn (D) strip of different PM_x fractions at five different distances from the road. The rectangle in Fig. 3C indicates the lateral section of the vegetation barrier. Values are means (µg cm^{-2}) ± SE. Different letters indicate significant differences within the same PM_x fraction at *P*= 0.05 using Tukey's test (HSD). n.s. indicates no significant difference.

3.7 Element deposition in the barrier and lawn strip

Five out of 21 analyzed elements (Pb, Sb, Se, Tl and V) were not detected in the majority of samples and were therefore removed from the dataset. Similarly to the leaf deposition findings, elements showed different concentrations in the following rank order: Ca > Fe > Al > K > Zn > Mg > Mn > As > Ba > Ni > Cu > Li > Cr > Cd > Co > Mo (Tab. 4). Barrier and lawn strips differed in the deposition of 8 out of 16 elements (Ca, Al, K, Mg, Mn, Ni, Mo, Co), with higher values in the barrier strip compared with the lawn strip (Fig. 4).

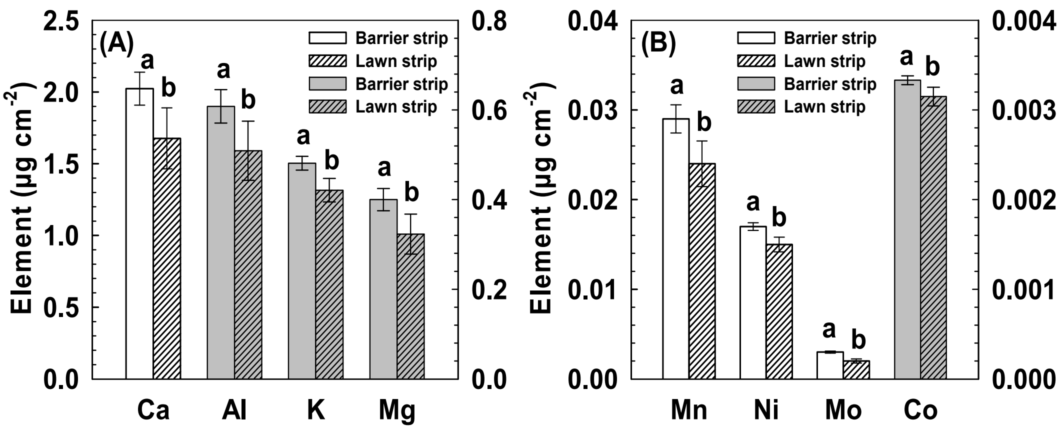


Fig. 4. Deposition per unit PTFE membrane area of Ca, Al, K, Mg (A) and Mn, Ni, Mo, Co (B) in the barrier strip and in the lawn strip. Ca, Mn, Ni, and Mo refer to left y axes; while Al, K, Mg, and Co refer to right y axes. Values are means (µg cm^{-2}) ± SE. Different letters indicate significant differences within the same element at *P*= 0.05 using Tukey's test (HSD).

Element deposition in the barrier and lawn strips changed similarly over time (Tab. 4). In 10 out of 17 elements (Al, Ba, Ca, Cd, Cr, Fe, K, Mg, Mn, Mo), there was a decrease from the August to September and an increase from September to October. Element deposition decreased significantly at increasing distances from the road with a higher decreasing rate in the barrier strip than in the lawn strip (Tab. 4).

Tab. 4. Deposition per unit PTFE membrane area of 16 elements (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mo, Ni, V, Zn) in the barrier strip (A) (n = 45) and in the lawn strip (B) (n = 45) in i) three different sampling periods (1 Aug 16, 1 Sep 16 and 3 Oct 16) (n = 15) and at ii) five different distances from the road (2.0 m, 5.5 m, 9.0 m, 12.5 m, 19.5 m) (n = 9). Values are means (µg cm^{-2}).

(A) Barrier strip

	Al	As	Ba	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Mo	Ni	Zn
Period (Pd)																
01 Aug 16	0.83 a	0.030 a	0.016 a	2.44 a	0.0043 a	0.003 b	0.009 a	0.005 a	1.07 a	0.53 a	0.009 a	0.52 a	0.034 a	0.003 a	0.017 b	0.24 a
01 Sep 16	0.34 c	0.031 a	0.013 a	1.32 b	0.0037 b	0.003 b	0.005 b	0.003 a	0.48 b	0.37 b	0.009 a	0.22 b	0.019 b	0.002 b	0.015 b	0.11 b
03 Oct 16	0.70 b	0.030 a	0.014 a	2.48 a	0.0041 a	0.004 a	0.009 a	0.007 a	1.14 a	0.55 a	0.009 a	0.49 a	0.035 a	0.003 a	0.020 a	0.18 a
Distance (Dc)																
2.0 m	0.81 a	0.034 a	0.017 ab	3.09 a	0.0042 a	0.003 a	0.010 a	0.006 ab	1.32 a	0.52 ab	0.009 a	0.51 ab	0.037 a	0.003 a	0.017 a	0.20 ab
5.5 m	0.86 a	0.031 ab	0.021 a	2.81 a	0.0038 a	0.003 a	0.009 a	0.013 a	1.30 a	0.56 a	0.009 a	0.54 a	0.039 a	0.003 a	0.017 a	0.23 a
9.0 m	0.59 b	0.028 b	0.013 b	1.79 b	0.0040 a	0.003 a	0.008 a	0.001 b	0.77 b	0.45 b	0.009 a	0.37 bc	0.025 b	0.003 a	0.018 a	0.15 bc
12.5 m	0.38 c	0.029 ab	0.011 b	1.20 c	0.0042 a	0.003 a	0.005 b	0.004 b	0.51 c	0.41 b	0.009 a	0.27 c	0.020 b	0.002 a	0.016 a	0.10 c
19.5 m	0.47 bc	0.030 ab	0.011 b	1.51 bc	0.0040 a	0.003 a	0.007 ab	0.002 b	0.59 bc	0.47 ab	0.009 a	0.36 bc	0.026 b	0.003 a	0.018 a	0.22 ab
Significance																
Pd	<0.001	0.769	0.151	<0.001	0.009	<0.001	<0.001	0.170	<0.001	<0.001	0.384	<0.001	<0.001	<0.001	<0.001	<0.001
Dc	<0.001	0.011	<0.001	<0.001	0.530	0.498	<0.001	<0.001	<0.001	<0.001	0.075	<0.001	<0.001	0.683	0.513	<0.001
Pd x Dc	<0.001	0.021	<0.001	0.019	0.381	0.079	0.185	0.151	0.001	0.019	0.477	0.066	<0.001	0.039	0.112	0.023

(B) Lawn strip

	Al	As	Ba	Ca	Cd	Co	Cr	Cu	Fe	K	Li	Mg	Mn	Mo	Ni	Zn
Period (Pd)																
01 Aug 16	0.587 a	0.032 a	0.015 a	1.78 a	0.0042 ab	0.003 b	0.007 a	0.005 a	0.79 a	0.42 ab	0.009 a	0.34 a	0.025 b	0.003 a	0.014 b	0.16 a
01 Sep 16	0.269 b	0.033 a	0.010 a	0.831 b	0.0031 b	0.002 c	0.004 b	0.001 a	0.37 b	0.33 b	0.009 a	0.14 b	0.014 c	0.002 b	0.011 c	0.06 b
03 Oct 16	0.726 a	0.028 b	0.018 a	2.558 a	0.0042 a	0.004 a	0.007 a	0.005 a	1.22 a	0.51 a	0.009 a	0.52 a	0.036 a	0.002 b	0.019 a	0.14 ab
Distance (Dc)																
2.0 m	0.727 a	0.033 a	0.013 a	2.454 a	0.0036 ab	0.003 a	0.005 b	0.005 a	1.22 a	0.51 a	0.009 ab	0.46 a	0.033 a	0.002 a	0.017 a	0.12 ab
5.5 m	0.773 a	0.033 a	0.021 a	2.412 a	0.0051 a	0.003 a	0.009 a	0.004 a	1.14 ab	0.52 a	0.009 ab	0.48 a	0.031 ab	0.002 a	0.017 a	0.21 a
9.0 m	0.554 ab	0.030 ab	0.019 a	1.701 ab	0.0035 ab	0.003 a	0.007 ab	0.005 a	0.81 abc	0.39 ab	0.009 a	0.33 ab	0.026 ab	0.002 a	0.015 ab	0.09 ab
12.5 m	0.351 ab	0.028 b	0.011 a	1.343 ab	0.0043 ab	0.003 a	0.005 b	0.003 a	0.52 bc	0.39 ab	0.009 ab	0.28 ab	0.019 ab	0.003 a	0.013 ab	0.11 ab
19.5 m	0.233 b	0.031 ab	0.008 a	0.696 b	0.0027 b	0.002 b	0.004 b	0.001 a	0.28 c	0.30 b	0.008 b	0.12 b	0.016 b	0.002 a	0.010 b	0.07 b
Significance																
Pd	0.003	0.001	0.075	0.001	0.035	<0.001	0.006	0.312	0.001	0.001	0.369	0.001	<0.001	0.001	<0.001	0.023
Dc	0.009	0.004	0.050	0.005	0.021	<0.001	0.004	0.816	0.004	0.001	0.048	0.012	0.021	0.039	0.013	0.040
Pd x Dc	0.234	0.001	0.039	0.265	0.11	<0.001	0.006	0.270	0.225	0.060	0.017	0.258	0.131	0.008	0.012	0.075

**P-Values* are reported for all factors and for interactions. Different letters indicate significant differences within the same factor and element at $P = 0.05$ using Tukey's test (HSD).

3.8 Microscope analysis

Percentage values of filter surface occupied by PM_{10-100} (data not shown) showed very similar trends in terms of species, planting density, sampling period, and distance from the road to those observed for the leaf deposition of PM_{10-100} (Fig. 2). The area occupied by black PM_{10-100} (5.41 %) was significantly lower than the area of brown PM_{10-100} (9.91 %), (Fig. 5). The filters from leaves at 2.0 m from the road were generally characterized by a more marked black coloration (Fig. 6A), while filters from 9.0 m were more brown (Fig. 6B). Black PM_{10-100} showed a constant decrease from the road (2.0 m) to the end of the barrier (9.0 m), while brown PM_{10-100} had higher values in the middle of the barrier (5.5 m) compared with the other distances (Fig. 5). X-ray microanalysis revealed that black PM_{10-100} was mainly composed of C and O with traces of Si (Fig. 6C). In the brown particles, a higher presence of Si, Al, Ca, Fe, Mg and K in addition to C and O was found (Fig. 6D). From the observations of leaf morphology at ESEM, no presence of hairs or trichomes on the upper leaf surface of both species was found (Fig. 7A), while in *V. lucidum,* there were agglomerations of waxes, with a diameter generally lower than 10 µM (Fig. 7E). The lower leaf surface of *P. x fraseri* was smooth (Fig. 7B), while in *V. lucidum,* the presence of trichomes was observed (Fig. 7D).

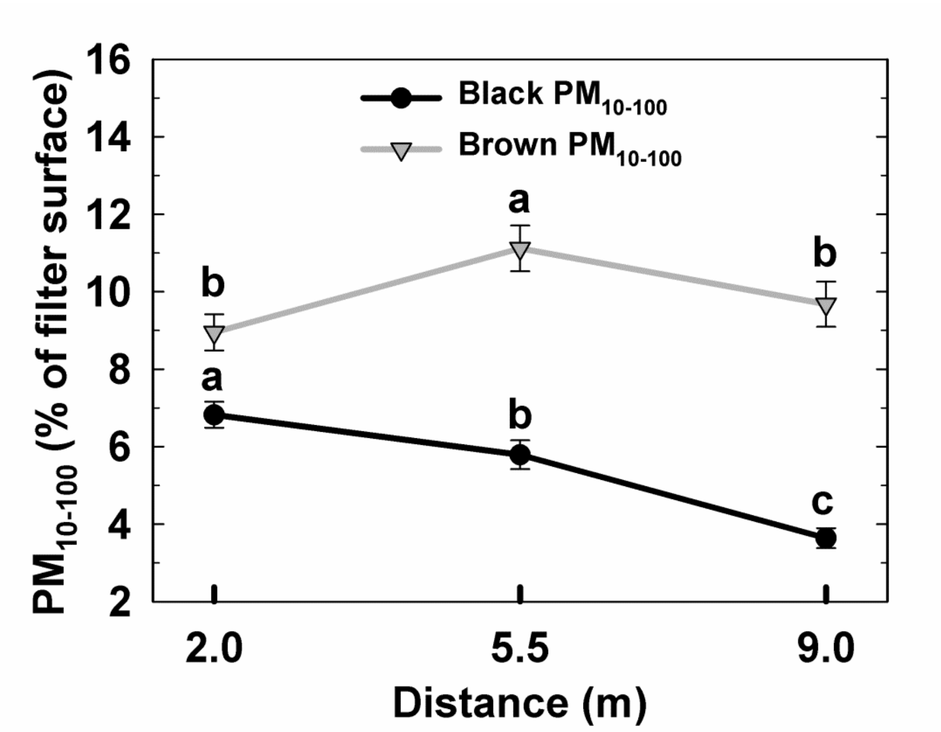


Fig. 5. Percentage of PM_{10-100} paper filter surface occupied by black and brown particles derived from leaves washed at different distances from the road. Values are means (percentage ± SE). Different letters indicate significant differences within the same PM_{10-100} color at P= 0.05 using Tukey's test (HSD).

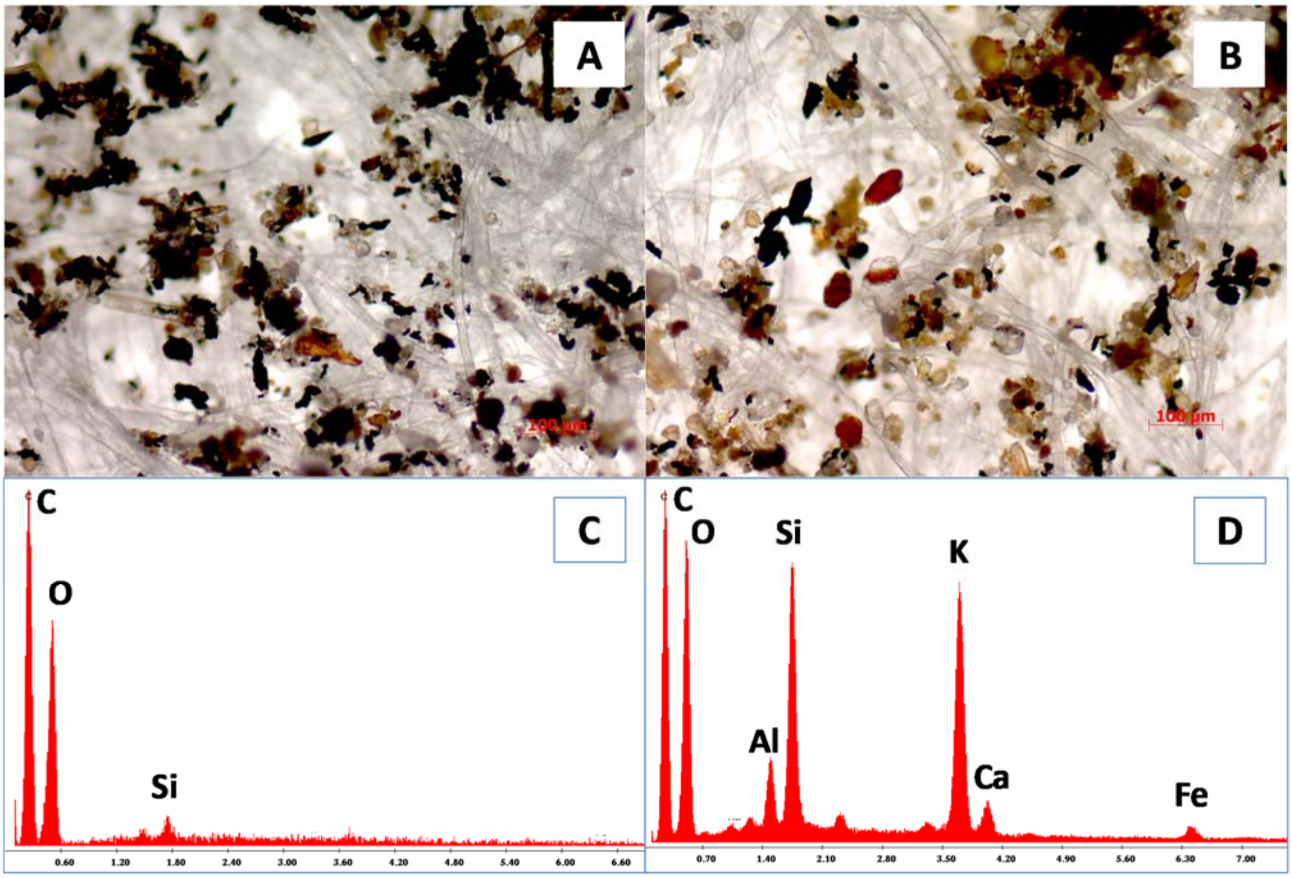


Fig. 6. Images at 10x magnification of PM_{10-100} paper filters (type 91) from (A) leaves at 2.0 m from the road, and from (B) leaves at 9.0 m from the road, carried out with a confocal microscope. Qualitative element composition in (C) black and (D) brown PM_{10-100} on type 91 filters carried out using X-ray microanalysis.

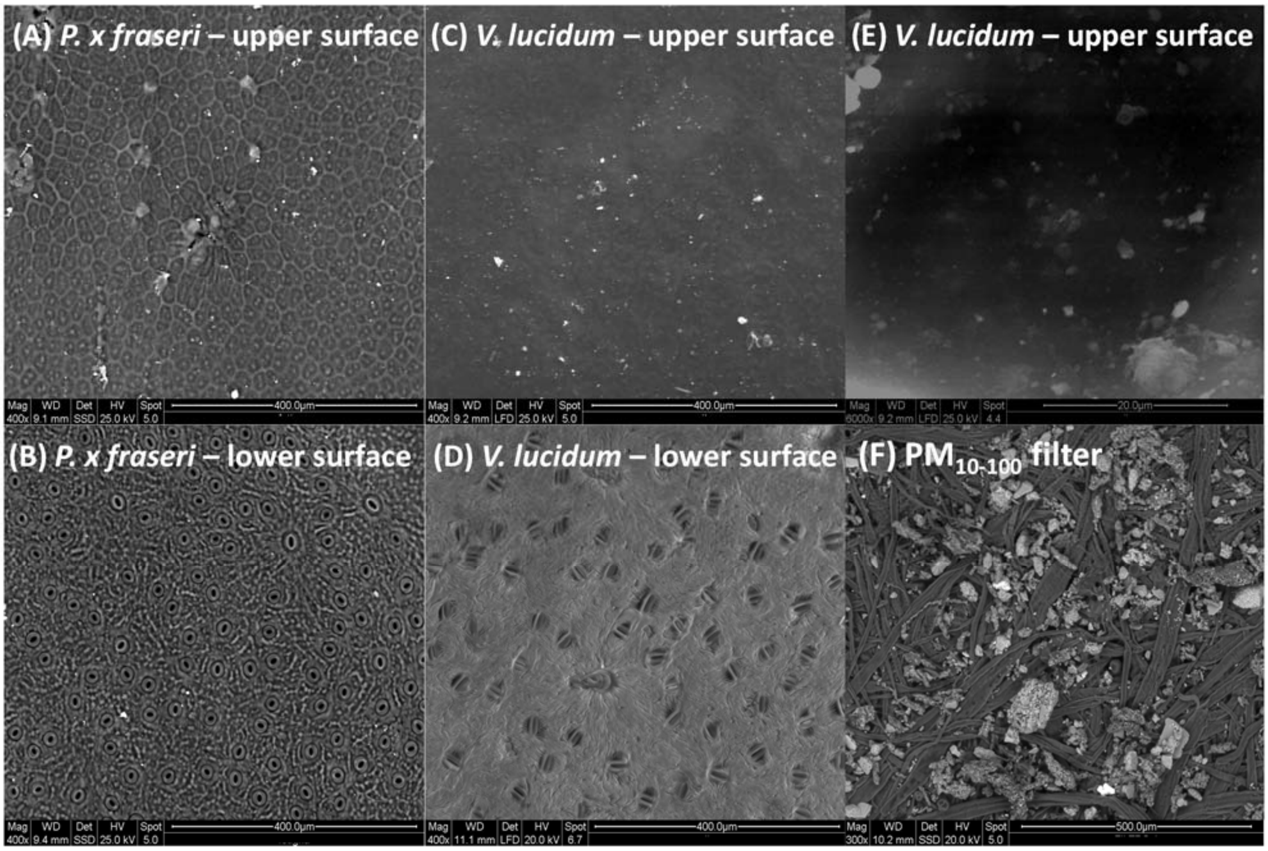


Fig. 7. Images taken with an environmental scanning electron microscope (ESEM) at 400x (*P. x fraseri* and *V. lucidum*, A-D), 300x (only upper surface of *V. lucidum*, E) and 6000x (only PM_{10-100} filter, F) magnification.

<u>3.9 Relationships between air pollutants and climate parameters</u>

Three different groups of air pollutants were identified through correlation analysis (Tab. B). The first group included air pollutants (PM_{10-100}, Al, Ba, Ca, Co, Cr, Cu, Fe, K, Mg, Ni, V and Zn) that showed high Pearson's coefficient values with each other (r = 0.6 – 0.9) and poor correlations with the other pollutants (r ≤ 0.3-0.4). Arsenic, Cd, and Mo formed a second group with an intermediate degree of correlation with each other (r = 0.4-0.5). A third group consisted of $PM_{2.5-10}$ and $PM_{0.2-2.5}$ which were weakly correlated with each other at r = 0.27 and showed even less correlation with the other variables.

Meteorological parameters affected leaf deposition in different ways depending on the pollutants considered (Tab. B). Increasing air temperatures and increasing wind velocity tended to increase leaf deposition in almost all air pollutants of the previously identified "first group", but induced a decrease in As and $PM_{2.5-10}$. Higher precipitations tended to decrease almost all the air pollutants in the "first group" and $PM_{0.2-2.5}$ but induced an increase in As and $PM_{2.5-10}$.

The first three PCA components explained 84.3% of the total variance (Fig. 8). The air pollutants were differently associated with the first three PCA components obtaining three different groups (Tab. 5). These groups corresponded well to the three different groups determined by the correlation analysis (Tab. B).

The PCA scores (indicated with circles in Fig. 8), showed how the two species in the third sampling period (*P. x fraseri* = black full circles; *V. lucidum* = black empty circles) were differentiated from each other and from the other samples. On the other hand, the samples from the first and second sampling period (Aug, Sep 16) were not well differentiated from each other.

Tab. 5. Component loadings (i.e. coordinates) of different air pollutants in the first three components of principal component analysis. The higher component weight in absolute values of every air pollutant is reported in bold, indicating membership in the respective component/group.

	Comp. 1- Group 1	Comp. 2 – Group. 2	Comp. 3 – Group. 3
PM_{10-100}	**0.27**	-0.15	0.08
$PM_{2.5-10}$	-0.13	0.01	**0.67**
$PM_{0.2-2.5}$	0.03	-0.33	**0.48**
Al	**0.27**	-0.16	-0.03
As	-0.01	**0.50**	-0.05
Ba	**0.26**	0.00	0.13
Ca	**0.28**	-0.16	0.04
Cd	0.10	**0.46**	0.15
K	0.25	0.02	**-0.36**
Co	**0.28**	0.06	0.07
Cr	**0.29**	-0.05	0.16
Cu	**0.23**	0.06	0.10

Fe	**0.30**	-0.08	0.03
Li	**0.26**	0.25	-0.09
Mg	**0.29**	-0.09	-0.09
Mo	0.11	**0.45**	0.25
Ni	**0.26**	0.20	0.11
V	**0.29**	-0.14	-0.05

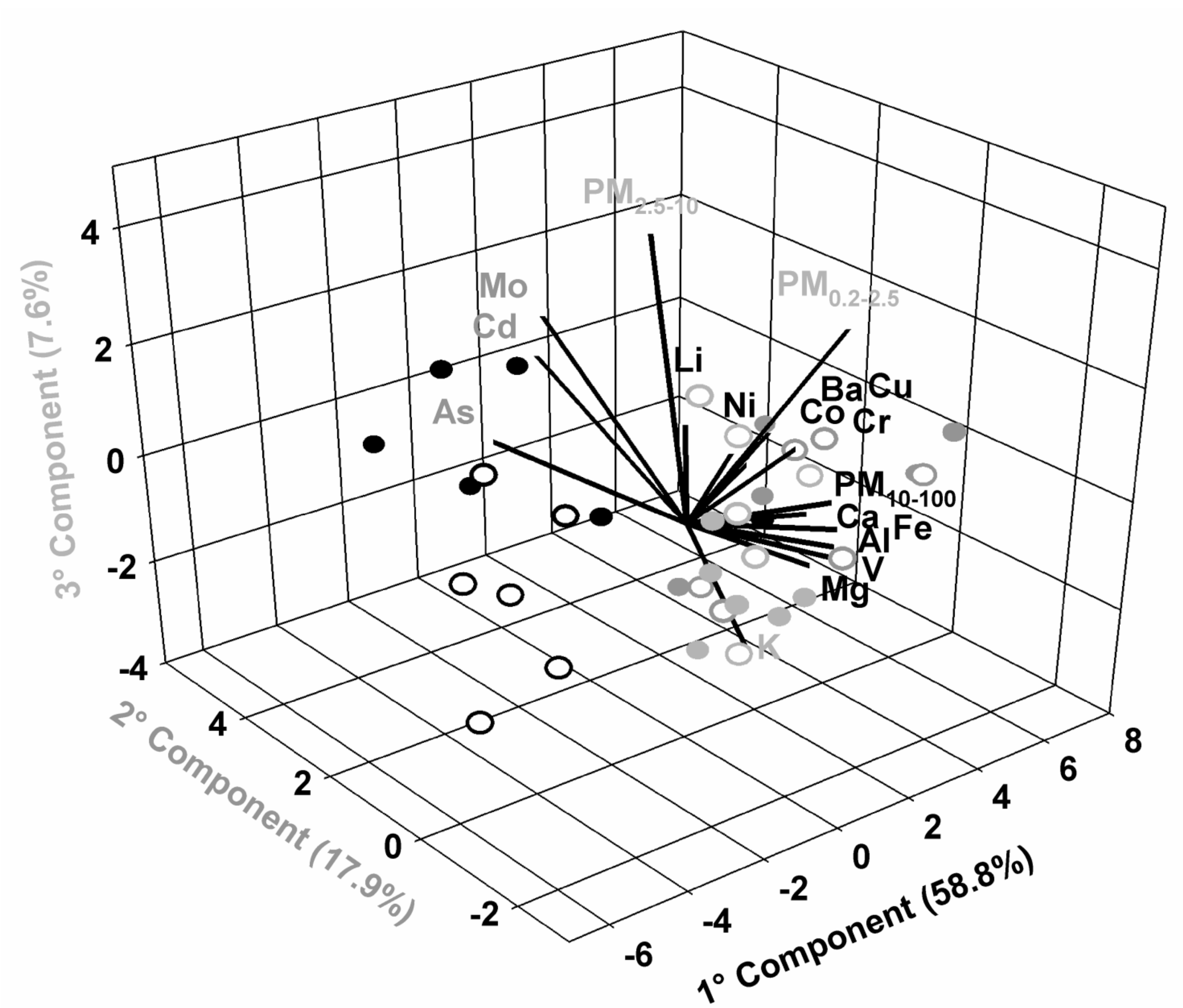


Fig. 8. 3D-Biplot reporting PCA results carried out on data of PM_x and element leaf deposition from the entire experiment. The first three components (axes) with the relative explained variance (%) are reported. Eigenvectors (line-plot) represent the loadings (coordinates) of different variables (PM_x and elements) on the first three components. Variables in black, dark grey, and light grey are mainly associated with component 1, 2

and 3, respectively. The scatter-plot reports the scores (coordinates) of different samples in relation to the first three components. The full-colored circles represent *P. x fraseri* in the first (light grey), second (dark grey) and third (black) sampling periods. Empty-rimmed circles represent *V. lucidum* in the first (light grey), second (dark grey), and third (black) sampling periods.

4. Discussion

4.1 Experimental site

The area surrounding the experimental site showed peri-urban characteristics with a decreasing order in terms of the percentage of occupied land surface between agricultural activities and green areas, buildings and roads and parking, at increasing distances (Tab. 2B).

The number of vehicles (10,844) passing in front of the experimental field from 7:00 a.m. to 08:00 p.m. was in line with the daily mean of 12,000 previously reported by Mori et al., (2015a) but was lower compared to other works (Hagler et al., 2012; Brantley et al., 2014; Mori et al., 2015b; Valotto et al., 2015). Despite this, in a previous work (Mori et al., 2015a) we demonstrated that the traffic load was sufficient to produce a detectable deposition of pollutants in the surrounding area.

4.2 PM_x and element deposition per unit leaf area

PM_x deposition on leaves showed similar trends to those observed in previous works carried out with the same methodology but on different species and in different environments (Dzierzanowski et al., 2011; Mori et al., 2015b). Dzierzanowski et al. (2011) reported a mean value of nearly 18 μm cm^{-2} total PM deposed on leaves of eight broad-leaved plant species. Our group (Mori et al., 2015b) found a mean deposition of nearly 16 μm cm^{-2} total PM on needles of two coniferous species.

The lower $PM_{2.5-10}$ and $PM_{0.2-2.5}$ leaf deposition of *P. x fraseri* (Fig. 2A) compared to *V. lucidum* were likely due to the presence of waxes and trichomes, on the upper and lower leaf surfaces respectively, which were observed only in *V. lucidum* (Fig. 7). Waxes and trichomes increase the capacity of leaves to capture particles (Saebo et al., 2012; Popek et al., 2013), especially those lower than 2.5 μm (Tiwary et al., 2005). However, both trichomes and waxes, during the washing-off phase, could have been partially removed from the leaf surface leading to an increase in leaf deposition. Epicuticular waxes can have a considerable weight per unit leaf area (Dzierzanowski et al., 2011). In fact, the size of waxes and trichomes (roughly 10 μm) would explain why the two species only differed in terms of $PM_{2.5\text{-}10}$ and $PM_{0.2\text{-}2.5}$ and not $PM_{10\text{-}100}$ (Fig. 2A).

Conversely, element deposition per unit leaf area was higher in *P. x fraseri* relative to *V. lucidum* (Tab. 3), which indicates that the efficiency of leaves in trapping fine PM differs from elements. This different behavior was also highlighted by correlation: $PM_{2.5\text{-}10}$ and $PM_{0.2\text{-}2.5}$ were generally either not or were weakly correlated with elements, while strong and numerous correlations between $PM_{10\text{-}100}$ and elements were found (Tab. B).

These results were generally in line with previous works (Li et al., 2003; Araujo and Nel, 2009; Mori et al., 2015a), which reported a higher content of metals in the coarser fractions compared to the finer ones, but in contrast with findings reported by Duzgoren-Aydin et al. (2006) and Zannoni et al., (2016). This variability could be related to different environmental factors, i.e., source of emissions, site characteristics, climatic parameters, etc. which have been reported to influence deposition (Adhikari et al 2006; Shah and Shaheen, 2008).

The variation in leaf deposition during the different sampling periods confirms their temporary permanence on leaf surface (Nowak et al., 2006). $PM_{10\text{-}100}$ and most elements followed a similar trend to each other (Fig. 2C, Tab. 3), which highlighted their close relationship confirmed by correlation analysis (Tab. B) and PCA (Fig. 8, Tab. 5).

As reported in the Introduction, climatic parameters have a strong influence on leaf deposition (Beckett et al., 2000; Fowler et al., 2003; Litschke and Kuttler, 2008; Cavanagh et al., 2009). Our findings (Fig. 2C, Tab. 3) regarding the removal action of precipitations and the impact of higher wind speeds, on $PM_{10\text{-}100}$ and the deposition of elements on the leaves, were found to be in line with previous studies (Beckett et al., 2000; Nowak et al., 2006). Rain washes off depositions onto the ground, while higher wind speeds tend to increase the deposition velocity, a parameter that is used to describe the phenomena of dry deposition (Fowler et al., 2003; Freer-Smith et al., 2005). In our study, temperature had a contrasting effect on leaf deposition, as previously observed by other authors (Fowler et al., 2003; Litschke and Kuttler, 2008; Cavanagh et al., 2009). $PM_{2.5-10}$, $PM_{0.2-2.5}$, As, Cd and Mo showed a different trend over time compared with the remaining air pollutants (Fig. 2C, Tab. 3). Smaller particles showed a lower tendency to deposit compared to coarser particles (Fowler et al., 2003). In addition, the deposition of smaller particles is more driven by interception and impaction, while above a 10 μm diameter, gravitational settling prevails (Fowler et al., 2003). A different response to climatic factors may also have contributed to the different trend observed. Przybysz et al. (2014) reported how the finer fraction of PM_x, compared to coarser ones, is less sensitive to the action of rain because of its higher adhesion to leaves. In addition, other works have reported how different air pollutants derived from the same sampling area may show different behaviors depending on the seasonal trend (Adhikari et al 2006; Shah and Shaheen, 2008) and different emission sources (Monaci et al., 2000).

In the present work, LAI had a greater effect on leaf deposition than planting density. In *V. lucidum*, LAI was similar in terms of high and low planting densities, and no differences in PM_x leaf deposition were found (Fig. 2E, 1F). Conversely, in *P. x fraseri*, the lower LAI observed in the lower planting density induced a significantly lower leaf deposition in PM_{10-100} and $PM_{2.5-10}$.

The role of LAI in roadside vegetation barriers in reducing air pollutants was discussed by Hagler et al. (2012). In that case, the unclear effect of vegetation on air pollution was attributed to the low LAI (values around 3) possibly associated with a low rate of interception and impaction of particles on leaf surfaces. In the case of elements, a clearer effect of planting density on leaf deposition was observed with higher values at a higher planting density independently from the species (Tab. 3). Conversely, the effect of LAI on element deposition in the two species was less marked compared to PM_x deposition.

It was clear from this and from a previous experiment (Mori et al., 2015a) that the total exposed leaf area affected deposition to a greater extent than leaf morpho-anatomical traits (Tab. A). In fact, barrier transects with a higher total leaf area, had the highest deposition per unit leaf area irrespectively of the species and the planting density (Tab. A).

The decrease in leaf deposition of PM_{tot}, PM_{10-100} and most elements at increasing distances from the road (Fig. 2D; Tab. 3) confirmed that vehicular traffic was the main source of pollution in the area. We had previously reached similar conclusions (Mori et al., 2015a), when various multivariate statistical methods were used to identify the possible source of leaf deposition of elements in the same experimental area. The fact that only the coarser fractions of PM_x decreased significantly at increasing distances may be linked to the larger diameters associated with a higher tendency to deposition (Fowler et al., 2003) compared with $PM_{2.5-10}$, and $PM_{0.2-2.5}$, which instead had a higher dispersion capacity.

In a previous work (Mori et al., 2015b), we found a significant reduction in large PM and 14 elements on needles of *Picea sitchensis* between a distance of 5 and 25 m from the road. In contrast, Peachey et al. (2009) did not find significant differences in soluble concentrations of eight metals in leaves of four tree species at 0, 2, 4, 6 and 12 m from the road. The higher leaf deposition of PM_x and most elements at 1.5 m from the ground compared with a 3.0 m height (Fig. 2; Tab. 3) confirmed the contribution of traffic to the presence of air pollutants. In fact, as reported by Etyemezian et al. (2004), when vegetation is within a distance of 20 m from the road verge, the impact zone on plants of the dust plume from traffic is concentrated in the first two meters from the ground.

4.3 PM_x and elements in the experimental site

The deposition of PM_x in the experimental site during the different sampling periods partially differed from the element deposition (Tab. 4; Fig. 3). From August to September both PM_x and elements generally decreased, likely due to lower traffic emissions during the summer holiday. From September to October on the other hand, a more heterogeneous response was observed with increasing elements and decreasing PM_x deposition. The latter were in agreement with the PM_x leaf deposition (Fig. 2). A similar variability was observed in previous works and was related to different responses to environmental factors (Adhikari et al 2006; Nowak et al., 2006; Shah and Shaheen, 2008) as also observed in the present work (Fig. 8 and Tab. B). In fact, although elements are commonly associated with PM_x (Valotto et al., 2015), they have been found to show a different trend since they are also directly released from tires, brakes or other parts of vehicles (Adhikari et al 2006; Nowak et al., 2006; Shah and Shaheen, 2008; Zannoni et al., 2016). Considering the strong influence of particle diameter on deposition dynamics (Fowler et al., 2003), it is possible to understand how the deposition dynamics of elements can change depending on their association with PM.

The deposition in the experimental area (passive samplers) showed different dynamics compared with the leaf deposition. This may be the consequence of several factors: i) leaves were exposed to traffic emissions throughout the entire experiment, while PTFE membranes were renewed at each sampling; ii) the action of climatic factors, especially rain, on PTFE membranes was partially limited by the structure of the passive samplers; iii) the surface of PTFE membranes and leaves have very different physical characteristics, height from the ground and angle of exposure.

The presence of the vegetation barrier induced a modification in the deposition of air pollutants in the experimental site (Fig. 3C, Fig. 3D, Fig. 4, Tab. 4). In general, vegetation barriers and high vegetation modify the direction and reduce the velocity of the impacting air fluxes (Tiwary et al., 2005). This thus promotes the deposition of particles greater than

0.1 µm through interception, impaction and gravitational settling (Fowler et al., 2003; Freer-Smith et al., 2005). The velocity reduction of the impacting air fluxes can occur within the barrier and in the first meters downwind of the vegetation because of partial air stagnation (Tiwary et al., 2005). In the stagnation area, there may be a higher air pollution concentration and a higher gravitational deposition may occur, especially for coarser particles (Fowler et al., 2003). In line with this, Hagler et al. (2012) reported that vegetation "may reduce air flow leading to increased pollutant concentrations due to the more stagnant conditions within and behind the trees". This explains why the PM_{10-100} and $PM_{2.5-10}$ deposition tended to be higher in the middle and just behind the barrier (5.5 m and 9.0 from the road) compared to the other sampling distances. Conversely, in the lawn strip, the absence of obstacles led to an even drift of PM_x from the road.

The more marked decrease of elements at increasing distances from the road and the higher total deposition of 8 out of 21 elements observed in the barrier strip compared with the lawn strip (Tab. 4; Fig. 4), could be directly related to the capacity of the vegetation to limit the drift of air pollutants from the road to the area close to the barrier. In fact, deposition increases at higher air concentrations of air pollutants (Fowler et al., 2003). Previous works have used both field measurements and models to investigate roadside vegetation barriers with comparable dimensions to those of the vegetation barrier of the present work (Hagler et al., 2012; Brantley et al., 2014; Morakinyo and Lam, 2016) with contrasting results on the effects on air pollution concentration.

Considering the diversity between air pollution concentrations and air pollutant deposition, the percentage reduction of the concentration of air pollution behind the roadside vegetation barriers (Tiwary et al., 2008; Chen et al., 2015; Lin et al., 2016) was in agreement with findings in the present work between the barrier and lawn strips (Fig. 3). The lack of significant differences in the deposition of PM_x and elements between the lawn and the barrier strip in front of the road (2 m) suggests the uniformity of exposure of the entire experimental site to the traffic-deriving pollutant fluxes.

4.4 Microscope analysis

The similarities between the results of PM_{10-100} leaf deposition (Fig. 2) and the percentage of filter paper surface occupied by of PM_{10-100} (data not shown) confirm the reliability of both methods for investigating leaf deposition. The use of image analysis differentiated between on-leaf PM_{10-100} with different colorations, thus highlighting the different deposition at different distances from the road between black and brown particles (Fig. 5). To our knowledge, no previous studies have used image analysis with the same aim. The association between image analysis and X-ray microanalysis (Fig. 6) enabled us to determine the element composition of the black and brown particles separately. Although X-ray microanalysis has already been used to detect the element composition of particles (Sgrigna et al., 2015), selectively analyzing PM on the basis of their color may represent a novelty. The different presence at different distances from the road (Fig. 5) and the different element composition between black and brown PM_{10-100}, suggest a different origin. In particular black particles, which showed a progressive decrease from the road and an almost complete composition in C and O, are probably black carbon which is a typical component of PM derived from traffic (Ferrero et al., 2014). Brown particles on the other hand may be due to the contribution of soil, considering both the lack of a clear reduction far from the road and the presence of several typical elements from soil such as Si, Al, K etc., (Zhao et al., 2006).

Conclusions

Different species showed a different deposition per unit leaf area with higher $PM_{2.5-10}$ and $PM_{0.2-2.5}$ in *V. lucidum* and a higher presence of elements in *P. x fraseri*, indicating a different capacity to remove different types of air pollutants. A higher LAI induced a higher deposition per unit leaf area, while a higher plant density was not a determining factor.

The barrier strip at 19.5 m from the road showed a 31 % and 32 % lower deposition for PM_{tot} and PM_{10-100} respectively, compared with the lawn strip. In addition deposition in the barrier strip at different distances showed a bell-shaped trend, while in the lawn strip no significant difference was found. The barrier strip induced a higher element deposition compared to the lawn strip with a more marked decreasing trend at increasing distances from the road.

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