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Plant-microorganisms interaction promotes removal of air pollutants in Milan (Italy) urban area

Andrea Franzetti[a,*], Isabella Gandolfi[a], Giuseppina Bestetti[a], Emilio Padoa Schioppa[a], Claudia Canedoli[a], Diego Brambilla[a], David Cappelletti[b], Bartolomeo Sebastiani[b], Ermanno Federici[b], Maddalena Papacchini[d], Roberto Ambrosini[c]

[a] Dept. of Earth and Environmental Sciences (DISAT), University of Milano-Bicocca, Milano, Italy
[b] Dipartimento di Chimica, Biologia e Biotecnologie, Università degli Studi di Perugia, Perugia, Italy
[c] Dept. of Environmental Science and Policy, Università degli Studi di Milano, Milano, Italy
[d] INAIL, Settore Ricerca, Certificazione e Verifica, Dipartimento di Innovazione Tecnologica (DIT), Laboratorio di Biotecnologie, Rome, Italy

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ABSTRACT

Plants and phyllosphere microorganisms may effectively contribute to reducing air pollution in cities through the adsorption and biodegradation of pollutants onto leaves. In this work, during all seasons, we sampled atmospheric particulate matter (PM_{10}) and leaves of southern magnolia *Magnolia grandiflora* and deodar cedar *Cedrus deodara*, two evergreen plant species widespread in the urban area of Milan where the study was carried out. We then quantified Polycyclic Aromatic Hydrocarbons (PAHs) both in PM_{10} and on leaves and used sequencing of 16S rRNA gene, shotgun metagenomics and qPCR analyses to investigate the microbial communities hosted by the sampled leaves. Taxonomic and functional profiles of epiphytic bacterial communities differed between host plant species and seasons and the microbial communities on leaves harboured genes involved in the degradation of hydrocarbons. Evidence collected in this work also suggested that the abundance of hydrocarbon-degrading microorganisms on evergreen leaves increased with the concentration of hydrocarbons when atmospheric pollutants were deposited at high concentration on leaves, and that the biodegradation on the phyllosphere can contribute to the removal of PAHs from the urban air.

1. Introduction

Air pollution in urban areas is a global concern due to its detrimental effects on human health and ecosystem functioning (Lelieveld et al., 2015). Currently, this issue is managed by both emission reduction and local mitigation strategies (Wei et al., 2017); among the latter ones, the role of vegetation in urban areas is gaining interest (Baró et al., 2014). Indeed, many studies indicated that the management of urban forests could be a cost-effective strategy to meet specific environmental standards or policy targets (Escobedo et al., 2011; Escobedo et al., 2010). Indeed, plants have been suggested to effectively contribute to the enhancement of ecosystem services (i.e. the direct and indirect contributions of ecosystems to human well-being) (TEEB, 2011) in urban areas, including air pollution reduction and greenhouse gas emission offsetting (Beckett et al., 1998; Dzierzanowski et al., 2011; Nowak and Crane, 2002; Nowak et al., 2006). The process that primarily contributes to the removal of inorganic and organic pollutants from the air is the adsorption of pollutants onto leaves (Yang et al., 2015; Sæbø et al., 2012). However, although this process contributes to the removal of pollutants by adsorption from the air, it does not lead to the mineralisation of the contaminants. The potential ability of the microbial communities of the aerial parts of plants to degrade pollutants has been taken into consideration only recently (Wei et al., 2017; Weyens et al., 2015; Espenshade et al., 2019).

The phyllosphere, comprising the aerial parts of plants and dominated by leaves, represents a suitable habitat for microbes, and it has been estimated that the global bacterial population present in it could comprise up to 10^{26} cells (Morris and Kinkel, 2015). These communities do not represent random assemblies of microorganisms, rather they undergo selection processes that result, at least partially, in predictable microbial communities represented by few dominant phyla and other less represented taxa (Vorholt, 2012). Bacterial community structures can also show both temporal and spatial dynamics and can vary among plant host species (Redford et al., 2010). To date, the identification of traits that are important for microbial survival on leaf environment, for their interaction with host plants, and for pathogenicity has been the

* Corresponding author at: Dept. of Earth and Environmental Sciences (DISAT), University of Milano-Bicocca, Milano, Piazza della Scienza 1, 20126 Milano, Italy.
E-mail address: andrea.franzetti@unimib.it (A. Franzetti).

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main focus of studies on phyllosphere microbial ecology (Rosier et al., 2016; Urooj and Muthappa, 2015; Vacher et al., 2016), while the processes selecting pollutant-degrading bacteria on leaf surface have been addressed in few works only (Gandolfi et al., 2017; Smets et al., 2016).

In this work, we used culture-independent techniques to test the main hypothesis that the abundance of hydrocarbon-degrading microorganisms on leaf surfaces increases with the concentration of hydrocarbons caught by leaves from the air in an urban environment, and that such microorganisms could thus contribute to the mineralization of these compounds. We were also interested in unveiling possible seasonality patterns in both the structure and the functions of the microbial communities of the phyllosphere. To this end, we sampled along a year atmospheric particulate matter (PM_{10}), leaves of southern magnolia *Magnolia grandiflora* (simply 'magnolia' hereafter) and needles of deodar cedar *Cedrus deodara* ('cedar' hereafter), two evergreen plant species widespread in urban areas of central Europe (McBride, 2017). Polycyclic Aromatic Hydrocarbons (PAHs), widespread harmful constituents of urban particulate matter, were quantified on leaves and in PM_{10}. 16S rRNA gene amplicon sequencing, shotgun sequencing and quantitative (qPCR) analyses of genes related to PAH biodegradation were also applied to the bacterial communities of leaves.

2. Materials and methods

2.1. Sampling

Leaves from magnolia and cedar trees were collected in the area of "Parco Nord", an urban park located in Milan (Italy) (45°32'34.9" N; 9°12'54.5" E), from four different trees for each of the two species. Both magnolia and cedar plants were located in a grass area, far from other trees (Fig. S1). Sampling was performed on eight days in 2016, two for each season, for a total of 64 samples (Table S1). Magnolia leaves and small cedar branches were cut with a pruner and tweezers cleaned with ethanol before the collection of each sample. Samples were placed in aseptic plastic bags and transferred to the lab within 3 h, where they were kept at −20 °C until further processing.

PM_{10} was also sampled in the Parco Nord area (45°32'16.1" N; 9°12'34.3" E), for 6–8 days per season in 2016 (Table S1). PM_{10} samples were collected on UV-sterilized quartz fibre filters (Whatman, Maidstone, England) by a high-volume sampler (ECHO HiVol, TCR TECORA, Milan, Italy) that worked for 24 h at a flux speed of 200 L min^{-1}. After sampling, filters were wrapped in aluminium tinfoil, brought to the lab within 3 h, and kept at −20 °C until further processing.

PM_{10} and benzo(a)pyrene concentrations in the air, daily average temperature, rainfall, daily average radiation and relative humidity during 2016 recorded by the automatic station of the Regional Environmental Protection Agency (Arpa Lombardia) nearest to the locations where we collected leaves and PM_{10} (45°28'42.7"N, 9°13'54.0"E) are reported in Fig. S2.

2.2. PAH quantification

Extraction of PAHs from PM_{10} samples was carried out by ultrasonication of a portion of 13 cm^2 of the quartz filters in 10 mL of dichloromethane. The process was repeated three times for 10 min, and the extracts were unified afterward. Plant leaves (approximately 2 g per sample) were submerged in a solution of 30 mL of dichloromethane and gently washed with two successive mechanical agitations of 1 min each. Leaf surface area was estimated as reported in Supplementary Materials.

Before the extraction, the samples were spiked with an internal standard solution of 13 deuterated PAHs for quantification. The extracted solutions were concentrated, dissolved in hexane and purified in a 3% w/w H_2O deactivated silica gel column (70–230 mesh ASTM, Merck) for the successive analysis.

Thirty-nine PAH congeners were quantified on the extracts using a High Fast GC–MS system (Agilent Technologies, 7890A). The system was equipped with a capillary column (Select-PAH Agilent J&W, CP 7461, 15 m, 150 µm, 0.1 µm), coupled with a MSD quadrupole detector (Agilent 5975C, VL MSD, Triple-Axis Detector).

2.3. Biomolecular analyses

2.3.1. DNA extraction

Four magnolia leaves or 500 cedar needles were put in sterile plastic bags with 50–75 mL of a leaf wash solution (TrisHCl 20 mM, EDTA 10 mM pH 8, Tween 20 0.1%) to extract total genomic DNA. Bags were sonicated for 10 min, and the supernatant was filtered on 0.45 µm pore-size nitrocellulose membranes. DNA was extracted with FastDNA™ SPIN Kit for Soil (MP Biomedicals, Solon, OH, USA) from half filter, which was cut into small pieces and put into Lysing Matrix E Tube; extraction was performed according to manufacturer's instructions.

2.3.2. Taxonomic characterization of bacterial communities

The V5-V6 hypervariable regions of 16S rRNA gene were sequenced by Illumina MiSeq (Illumina Inc., San Diego, CA, USA) using a 2 × 250 bp paired-end protocol as previously reported (Gandolfi et al., 2017). Further library preparation with the addition of standard Nextera indexes (Illumina, Inc., San Diego, CA, USA) and sequencing were carried out at Parco Tecnologico Padano (Lodi, Italy).

The obtained sequences were demultiplexed according to the indexes and the internal barcodes used. The Uparse pipeline (Edgar, 2013) was used for subsequent elaborations, as previously reported. Representative sequences were first classified using SINA with SILVA database (Pruesse et al., 2012), and sequences not classified as belonging to Bacteria domain (i.e. Archaea, chloroplasts and mitochondria) were discarded. The remaining OTUs were then classified again with RDP database. The abundance of each OTU was estimated by mapping the sequences of each sample against the OTU representative sequences.

2.3.3. Shotgun metagenomics sequencing and sequence processing

Shotgun metagenomics sequencing was performed on 16 samples of leaves: 8 samples of magnolia (4 in winter and 4 in summer), and 8 samples of cedar (4 in winter and 4 in summer), with Illumina HiSeq 2000 using a 2 × 100 bp paired-end protocol. Sequence reads were processed as reported in the methodological details (Supplementary Materials).

2.3.4. Quantification of genes coding for naphthalene dioxygenase

Quantitative PCR (qPCR) was used to estimate the abundance of the gene coding for the naphthalene 1,2 dioxygenase of Gram-negative bacteria (*nah*Ac). The target 269-bp fragment was obtained from *Pseudomonas fluorescens* by PCR amplification with the primer pair P1& 2 F and P1&2 R (Meynet et al., 2015). More details are reported in Supplementary Materials.

2.4. Statistical methods

The structure of bacterial communities was compared between plant species and among seasons by means of redundancy analysis (RDA) on Hellinger-transformed OTU abundance, defined on the basis of 16S rRNA gene amplicon sequencing data. Hellinger transformation was chosen because it decreases the importance of OTU abundance over occurrence and avoids the double-zero problem when comparing OTU composition between samples (De Cáceres et al., 2010; Legendre and Legendre, 2012). RDA was followed by post hoc pairwise comparisons whose significance was adjusted by the False Discovery Rate (FDR) method (Benjamini and Yekutieli, 2001). Variation partitioning was also used to quantify the variation of community structures according to plant species and season (De Cáceres et al., 2010; Legendre and

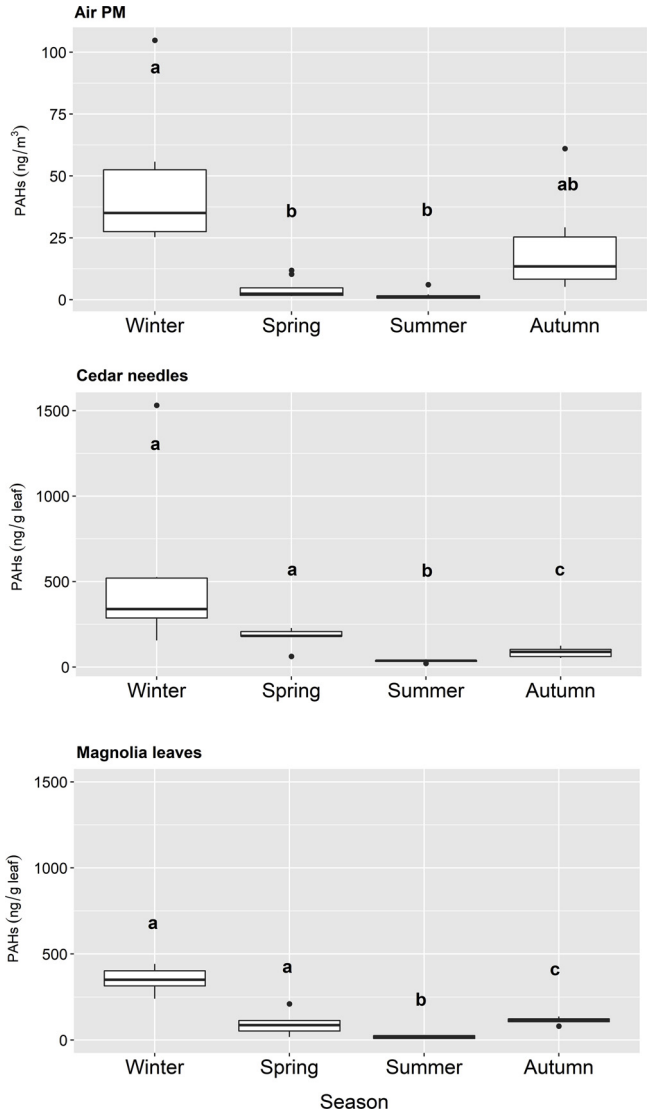

Fig. 1. Box-and-whisker plots reporting seasonal concentrations of PAHs in PM_{10} and in plant leaves. Data for PM_{10} are reported as mass/air volume concentrations (ng of PAHs / m^3 of air sampled) while in the case of plant leaves data are reported as mass/ mass units (ng of PAHs / g of leaf). The lower and upper hinges correspond to the first and third quartiles (the 25th and 75th percentiles). The upper and the lower whiskers extend from the hinge to the largest/smallest values no further than ± 1.5 x IQR (inter-quartile range) from the hinge. Data beyond the end of the whiskers are plotted individually. Different letters denote significant differences ($P < 0.05$) at Tukey post-hoc tests conducted separately for PM_{10} and for each plant species.

Legendre, 2012). The same analyses were performed on shotgun sequencing data to investigate the effects of plant host species and season on the coverage of the genes coding for the annotated enzymatic reactions. In this case, RDA was performed on Hellinger-transformed gene abundances in metagenomes. Comparisons of gene abundances were performed by generalized least squares models, in order to account for inhomogeneity of variance among groups, followed by Tukey post hoc comparisons. Analyses were performed in R 3.3.2 (R. Core Team, 2016) with the BiodiversityR, nlme, multcomp and multtest packages.

3. Results

3.1. PAH concentration on leaves and in PM_{10}

Trends of total PAH concentration (sum over 39 congeners) are shown in Fig. 1 for PM_{10} and leaf samples. The values for the individual 39 PAHs are reported in Table S2 of the Supplementary Materials. The seasonal trends of PAH concentrations showed significant differences between seasons in all cases ($F_{1,3} \geq 8.128$, $P < 0.001$), with maxima in winter and minima in summer in all cases, in agreement with well-established literature results (Perrone et al., 2010).

Interestingly, total PAH concentration in PM_{10} dropped to levels comparable to summer ones already during spring, while on leaves spring PAH concentrations either did not differ from winter ones, as observed on cedar needles, or they decreased to intermediate values between winter and summer ones, as observed on magnolia leaves. These patterns suggest that PAHs could persist on leaves for a longer time than in PM_{10}. Data about average seasonal relative abundance of each PAH (Table S2, Fig. S3) also suggest that the drop of total PAH concentrations in warm seasons was mainly due to the disappearance of lighter congeners, particularly naphthalene, while the relative increase of heavier mass (3, 4 and 5 rings) congeners plays a substantial role in the increase observed in autumn and winter. Moreover, the average relative abundance of naphthalene appeared to be higher on leaves than in PM_{10} in all seasons (Table S2). Naphthalene was also the most abundant compound in most samples of magnolia leaves in all seasons except for autumn, while on cedar needles and in PM_{10} it was generally the most abundant compound in winter only (Table S3).

PAH concentrations generally did not differ between the plants in the same seasons, with the exception of summer, when they were higher on cedar than on magnolia ($z = 3.899$, $P = 0.001$; $|z| \leq 2.204$, $P \geq 0.229$ in all the other cases).

3.2. Structure of bacterial communities on leaves

16S rRNA gene amplicon sequencing revealed that the structure of bacterial communities differed significantly both between plant species and among seasons (Table 1, Fig. 2a), with significant differences among all pairs of seasons at post-hoc tests ($F_{1,30} \geq 2.691$, $P_{FDR} \leq 0.001$). Variation partitioning analysis also showed that season and plant species *per se* explained similar amounts of variation in bacterial community structures (season: adjusted-$R^2 = 0.180$; plant species: adjusted-$R^2 = 0.186$), while their shared contribution was null (i.e. their effects on the structure of bacterial communities were independent to one another; Fig. 2b).

The observed differences in bacterial communities between plant species and among seasons seem mainly due to different abundances of the same taxa rather than to the presence of different orders. Indeed, the most abundant orders (Actinomycetales, Burkholderiales, Cytophagales, Rhizobiales, and Sphingomonadales) were shared by all samples, although with different abundances (Fig. S4a).

At genus level (Fig. S4b), the most abundant genera in all samples were *Hymenobacter, Sphingomonas, Methylobacterium* and *Massilia*. These genera have already been indicated as common populations of phyllospheric bacterial communities (Vorholt, 2012; Gandolfi et al., 2017; Bulgarelli et al., 2013; Rastogi et al., 2013). However, a few bacterial populations were more abundant in bacterial communities hosted by one of the two considered plant species. For example, the genus *Amnibacterium* (order Actinomycetales) was more abundant on magnolia leaves, reaching up to 7.6% in winter, while on cedar leaves it

Table 1
RDA of variation of Hellinger-transformed bacterial OTU abundances according to plant species and season.

Predictor	df	Variance	F	P
Host plant	1	0.053	18.304	0.001
Season	3	0.059	6.776	0.001
Residuals	59	0.171		

$F_{4,59} = 9.658$, $P = 0.001$, Adjusted-$R^2 = 0.355$.

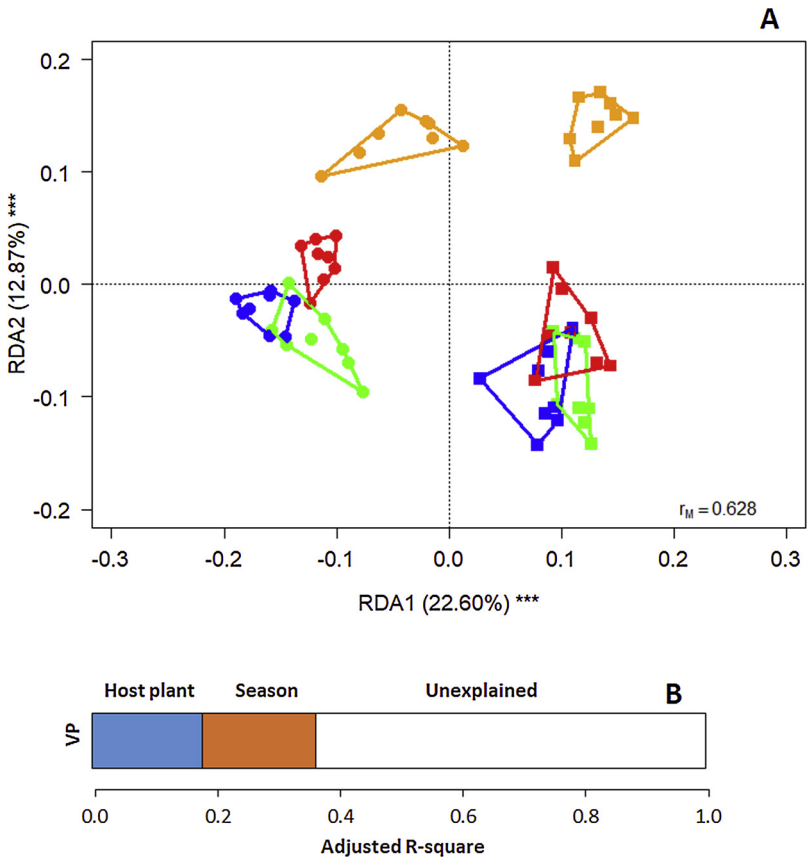

Fig. 2. a) Biplot from RDA of Hellinger-transformed bacterial OTU abundance on season and species. Each point represents one sample. Samples collected in different months are indicated by different colours (green = spring, red = summer, orange = autumn, blue = winter), while those collected from different species are indicated by different symbols (dots = cedar, squares = magnolia). Polygons include samples collected in the same season and from the same species. The percentage of variance explained by each axis and its significance (***: $P < 0.001$) are reported. r_M is the Mantel correlation coefficient between the Hellinger distance between samples and the Euclidean distance between the corresponding symbols in the graph. Values close to one indicate that the graph accurately represents the distance between samples. b) Results from the variation partitioning showing the amount of variance explained by the independent and combined effects of the predictors entered in the RDA. The amount of variance explained by the shared contribution of season and host plant was null.

never exceeded 0.9%. In contrast, populations belonging to families Rhizobiales and Acetobacteraceae, which were not classified at genus level, were more abundant on cedar needles in all seasons.

3.3. Phyllosphere metagenomes

3.3.1. Sequencing output and read processing

The number of sequence reads obtained from shotgun metagenomics sequencing ranged from 9,621,960 to 50,527,302 across the 16 samples with a total number of bases of 76,723,795,338. After the quality-trimming, the number of reads ranged from 6,556,044 to 19,560,963 (total bases 51,936,335,600). The co-assembly step produced 1,595,692 contigs, which included 1,517,106,361 bases. From this assembly, 2,224,815 coding sequences were predicted and 523,466 of them were successfully aligned against the Uniprot database. The final parsing using the Metacyc database led to the annotation of 4,663 enzymatic reactions, which belonged to 1806 metabolic pathways.

3.3.2. Effects of seasonality and tree species on epiphytic microbial community functions

RDA on the coverage of all the genes coding for the annotated enzymatic reactions showed significant differences between plant host species ($F_{1,15} = 23.424$, $P = 0.001$) and seasons ($F_{1,15} = 5.272$, $P = 0.031$) (Table 2; Fig. 3a). The net contribution of tree species accounted for almost all the variance explained by the model (Fig. 3b).

3.3.3. Abundance of catabolic genes for aromatic hydrocarbon degradation in the metagenomes

To gain more insights into the effect of PAH pollution on phyllospheric microbial communities, we retrieved 4 key enzymes corresponding to key reactions of the degradative pathway of aromatic hydrocarbons included in Metacyc database (Fig. 4).

All the considered catabolic genes were significantly more abundant on magnolia leaves than on cedar needles. The abundance of naphthalene 1,2-dioxygenase was also significantly affected by the interaction between tree species and season (Table 3). In particular, post-hoc tests showed that the abundance of this gene was significantly higher in magnolia than in cedar samples in winter and that, on magnolia leaves, it was higher in winter than in summer ($z \geq 3.475$, $P \leq 0.002$).

Table 2
RDA of variation of Hellinger-transformed gene abundances in metagenomes according to plant host species and season.

Predictor	df	Variance	F	P
Host plant	1	0.075	23.424	0.001
Season	1	0.017	5.272	0.031
Residuals	13	0.042		

$F_{2,13} = 14.348$, $P = 0.001$, Adjusted-$R^2 = 0.640$.

3.4. Quantification of nahAc gene on leaves

The abundance of the gene coding for the enzyme naphthalene 1,2-dioxygenase (*nah*Ac), which catalyses the first step of the aerobic biodegradation of naphthalene in Gram-negative bacteria, was quantified on cedar and magnolia leaves through qPCR. The log-transformed average number of *nah*Ac copies per cm^2 of leaf surface was similar on magnolia leaves and on cedar needles (magnolia: 3.752 ± 0.184 SE; cedar: 3.580 ± 0.095 SE, $t_{22.5} = 0.830$, $P = 0.415$). Moreover, on magnolia, naphthalene dioxygenase was significantly more abundant in winter and in spring than in summer and autumn ($F_{3,12} = 15.056$, $P < 0.001$, Tukey post-hoc tests: $|z| \geq 4.408$, $P < 0.001$ in all cases except for comparisons between winter and spring and summer and autumn where $|z| \leq 1.550$, $P \geq 0.391$; Fig. 5), while on cedar no significant difference among seasons was found ($F_{3,12} = 1.110$, $P = 0.383$; Fig. 5).

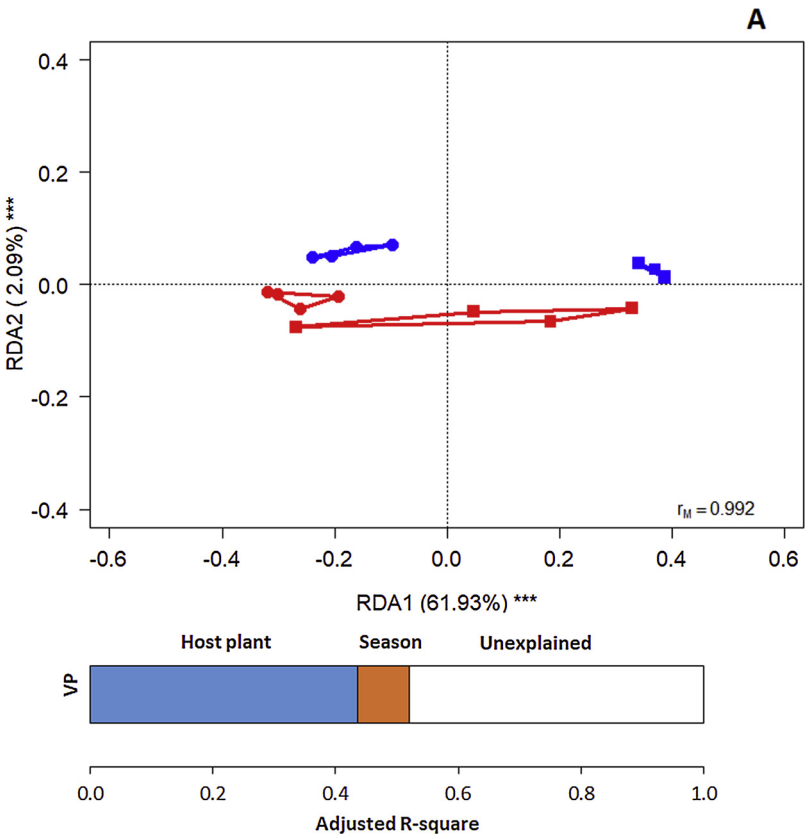

Fig. 3. a) Biplot from RDA of Hellinger-transformed gene abundance on season and species. Each point represents one sample. Samples collected in different months are indicated by different colours (red = summer, blue = winter), while those collected from different species are indicated by different symbols (dots = cedar, squares = magnolia). Polygons include samples collected in the same season and from the same species. The percentage of variance explained by each axis and its significance (***: P < 0.001) are reported. r_M is the Mantel correlation coefficient between the Hellinger distance between samples and the Euclidean distance between the corresponding symbols in the graph. Values close to one indicate that the graph accurately represents the distance between samples. b) Results from the variation partitioning showing the amount of variance explained by the independent and combined effects of the predictors entered in the RDA. The amount of variance explained by the shared contribution of season and host plant was null.

4. Discussion

The critical role of vegetation in removing PAHs from the atmosphere has been known for more than 20 years, when Simonich and Hites (Simonich and Hites, 1994) estimated that more than 40% of atmospheric PAHs were trapped by vegetation and delivered to soil, whereas more recent works reported lower values (Tian et al., 2008).

The temporal trends of PM_{10} and PAH atmospheric concentrations we observed were consistent with those reported in previous studies, which showed higher concentrations in cold seasons (autumn and winter), probably due to the use of heating systems, low winds and stable atmospheric conditions (see i.e. (Ferrero et al., 2014)), and lower concentrations in summer probably due to photodegradation reactions and atmospheric dispersion. The temporal trend of PAH concentration extracted from leaf samples in the present study was generally consistent with the air concentrations. This behaviour is in agreement with several previous reports of PAH deposition on plant leaves that showed that leaf concentrations were higher in urban/industrial areas compared to peri-urban or remote areas (Alfani et al., 2001; Tavera Busso et al., 2018). However, these results contrast with the findings by Tian and colleagues (Tian et al., 2008), who found no correlation between air and leaf concentrations of PAHs along the year.

The concentration of PAHs were not generally different between magnolia leaves and cedar needles in any season, except for summer. Considering that magnolia leaves have much higher content in wax than cedar ones (Güth et al., 1992; Maffei et al., 2004), this finding is in disagreement with previous reports indicating that PAH concentration on leaves increases with wax content (Prajapati and Tripathi, 2008). However, other evidences showed that PAH concentration in the cuticle is negatively correlated with the wax cuticle thickness (Wang et al., 2008), which is larger in magnolia than in cedar needles.

Naphthalene was the most abundant compound in most samples of magnolia leaves in all seasons except autumn, while on cedar needles and in PM_{10} it generally was the most abundant one only in winter (Table S2). Such abundance of naphthalene on leaves might be due to the high vapour pressure of the lighter PAHs, which facilitates both the direct absorption from the atmosphere through stomata and the exchange from the particulate phase to the wax-rich surface of the plant leaves. The stomatal conductance of a leaf, in particular, may determine the capturing efficiency of semi-volatile pollutants such as low-molecular-weight PAHs (De Nicola et al., 2017), while high-molecular-weight PAHs are usually deposited to the plant surface bound to particles in wet and dry deposition (Howsam et al., 2000).

Multivariate analyses of 16S rRNA gene amplicon data pointed out that the taxonomic composition of epiphytic bacterial communities was significantly influenced by both tree species and seasonality, which explained similar amounts of variation in bacterial community structures. This piece of evidence is in agreement with several studies that identified host plant as the shaping force of the structure of bacterial phyllosphere communities (Knief et al., 2010; Laforest-Lapointe et al., 2016; de Oliveira Costa et al., 2012; Ortega et al., 2016; Kim et al., 2012). RDA analyses of metagenomics data showed that both tree species and seasonality also affected the gene abundances, thus suggesting that epiphytic microbial communities on cedar and magnolia leaves had different functional profiles that also changed between winter and summer samples. However, in this case, variation partitioning analysis indicated that the contribution of the season in explaining data variability was limited, although significant, whereas variation in gene abundances was mainly due to host tree species. This fact implies that epiphytic bacterial communities simultaneously vary their functional and structural compositions between cedar and magnolia leaves and, at a lesser extent, along the seasons. In other words, the taxonomic variation of bacterial populations between tree species and seasons was paralleled by a functional variation that was greater between cedar and magnolia than between summer and winter. However, since the shotgun metagenomics targeted the DNA of all microorganisms, we cannot exclude that Archaea and Fungi could also significantly contribute to the observed diversity of the functional profiles.

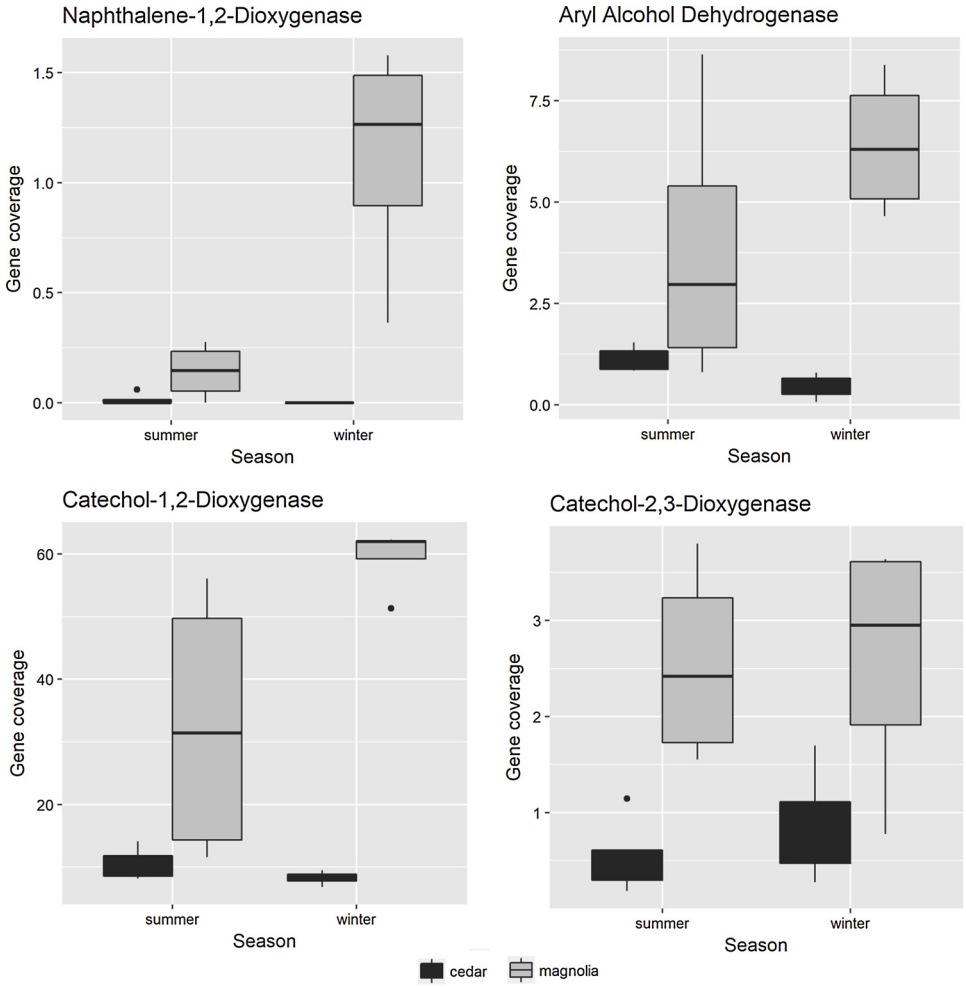

Fig. 4. Box-and-whisker plots reporting the coverage of the key genes involved in the biodegradation of PAHs. The lower and upper hinges correspond to the first and third quartiles (the 25th and 75th percentiles). The upper and the lower whiskers extend from the hinge to the largest/smallest values no further than ± 1.5 * IQR (inter-quartile range) from the hinge. Data beyond the end of the whiskers are plotted individually.

Table 3
Generalized Least Squares models of hydrocarbon-degrading pathways. The influence of tree species, seasons and species-season interaction is reported. P_{FDR} is the p-value adjusted for multiple statistical tests.

	Host plant		Season		Host plant by season interaction	
Enzymatic reaction	*F*	P_{FDR}	*F*	P_{FDR}	*F*	P_{FDR}
naphthalene-1,2-dioxygenase	19.583	0.002	0.663	0.898	12.415	0.035
aryl-alcohol dehydrogenase	7.897	0.033	19.050	0.008	2.676	0.355
catechol-1,2-dioxygenase	39.407	< 0.001	1.436	0.898	6.179	0.120
catechol-2,3-dioxygenase	15.908	< 0.001	0.688	0.898	0.104	1.000

Focusing on the specific metabolic functions related to pollutant degradation, we considered the abundance of genes coding for key enzymes of PAH catabolic pathways. Metagenomics and qPCR data revealed that the microbial communities on the leaves of both cedar and magnolia harboured genes involved in the degradation of poly-aromatic hydrocarbons. Both the key enzymes involved in upper reactions of the catabolic pathways (e.g. naphthalene dioxygenases), responsible for the ring dihydroxylation, and the key enzymes of the lower pathways (catechol dioxygenases), responsible for aromatic ring cleavage, were annotated, thus suggesting that the microbial communities possess the complete metabolic pathways for the mineralization of PAHs (Mihelcic and Luthy, 1988). This fact confirms the results of traditional cultivation methods (Waight et al., 2007; Sazonova et al., 2017), amplicon (Gandolfi et al., 2017) and shotgun metagenomics sequencing (Imperato et al., 2019).

Magnolia hosted both higher proportions and higher absolute abundances of PAH-degrading microorganisms than cedar. However, the taxonomic identification of these hydrocarbon-degrading populations is not straightforward. Indeed, the most abundant bacterial genera, namely *Hymenobacter, Sphingomonas, Methylobacterium* and *Massilia,* represented most of the core microbiome of both plant species. Thus, they do not appear to be major candidates as main hydrocarbon-degraders, at least at genus level. On magnolia leaves, another genus was particularly abundant in winter and in spring, when also copy number of naphthalene dioxygenase was higher: *Amnibacterium* (7.6 ± 2.1% and 5.6 ± 1.9%, respectively). Moreover, this genus was less abundant on cedar needles in the same seasons, accounting for 0.9 ± 0.5% and 0.9 ± 0.4%, respectively. However, members of this genus have never been reported as hydrocarbon degraders.

The abundance of PAH-degrading microorganisms was not related to the amount of absorbed PAHs on the different plant species, which was higher on cedar. When considering differences among plant species, we can hypothesize that a selection process of epiphytic hydrocarbon-degrading microorganisms occurred, and that it was driven more by the leaf characteristics than by the selective pressure operated

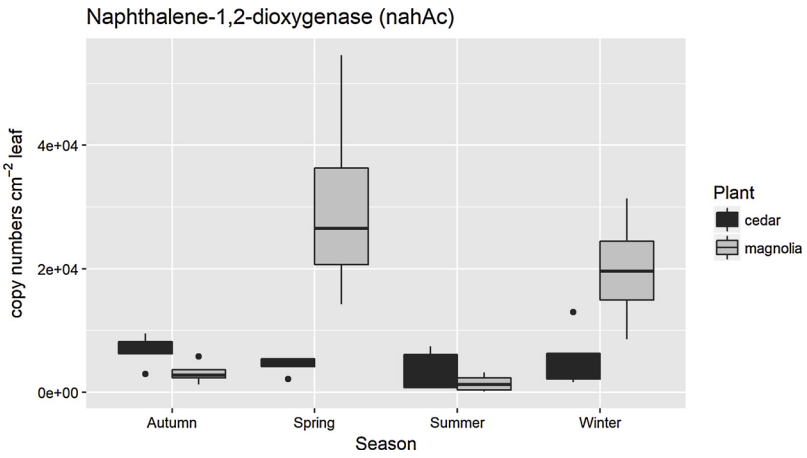

Fig. 5. Box-and-whisker plots reporting copy number of naphthalene 1,2 dioxygenase gene (*nah*Ac) estimated by qPCR. The lower and upper hinges correspond to the first and third quartiles (the 25th and 75th percentiles). The upper and the lower whiskers extend from the hinge to the largest/smallest values no further than ± 1.5 x IQR (inter-quartile range) from the hinge. Data beyond the end of the whiskers are plotted individually.

by the presence of pollutants as a possible carbon source. This result is consistent with previous works reporting high variability of composition and abundance of PAH-degrading bacterial communities across different plant hosts. For instance, Sazonova and colleagues (Sazonova et al., 2017) reported that the composition of PAH-degrading consortia enriched from leaves markedly differed among three woody tree plant species. Interestingly, Yutthammo et al. (Yutthammo et al., 2010) reported that the number of phenanthrene-degrading bacteria ranged from 10^1 to 10^4 per gram of leaf in ten ornamental plants and was positively correlated with the wax content but negatively correlated with leaf area. This is consistent with our observation of the higher abundance of PAH degrading bacteria on magnolia leaves, where the content of wax is much higher than in cedar needles (Güth et al., 1992; Maffei et al., 2004).

Magnolia and cedar also showed different behaviour in the temporal trend of PAH-degrading populations. Indeed, both metagenomics and qPCR results consistently indicated that this functional group of microorganisms was significantly more abundant in cold seasons than in warm ones only in the case of magnolia, whereas its abundance on cedar was constant along the year. The *nah*Ac gene, coding for a naphthalene 1,2-dioxygenase, which was used as a marker for naphthalene degrading microorganisms, was found at significantly higher abundances on magnolia leaves in winter and spring than in summer and autumn. This difference is particularly relevant since naphthalene is one of the most abundant compounds among PAHs on the leaves, and also the one that showed the most marked increase from warmer to colder seasons. The finding that this gene abundance increased on magnolia leaves when atmospheric pollutants reached their peak may support the main hypothesis that the absorption of hydrocarbons on leaves is one of the drivers that can confer a selective advantage to hydrocarbon-degrading microorganisms over other populations by providing an alternative source of energy and carbon. Admittedly, the results collected in this work are limited to magnolia and naphthalene, and we hope that further studies can improve the knowledge about this important mechanism. Moreover, the observed shifts in the abundance of hydrocarbon-degrading populations may be due not only to pollutant concentrations but also to seasonal variations of other parameters, which were not considered in this work. However, if confirmed, these findings would imply that the hydrocarbons are biodegraded by enriched microbial populations, which therefore would actively contribute to the removal of air pollution.

Since biodegradation processes lead to the removal of the contaminants, they have advantages over the adsorption processes, which only promote the transfer of pollutants from the atmosphere to other environmental compartments. So far, attempts to quantitatively evaluate the relevance of biodegradation in the removal of organic air pollution have been made only by lab-scale experimental approaches. Yutthammo and colleagues (Yutthammo et al., 2010) reported that unsterilized leaves of water jasmine (*Wrightia religiosa*) removed PAHs volatilized in 60 m L-vials with an efficiency ranging from 80.1% to 86.8%, while sterilized leaves removed 73.2–82.3 %. The PAH removal efficiency in a larger-scale (14 L) experiment was much lower due to the smaller amount of leaves in the chamber. More recently, it has been reported that pyrene removal rate on a jungle geranium (*Ixora coccinea*) leaf was 15.2 ± 1.0 μg day^{-1} (Siriratruengsuk et al., 2017). In our study, to evaluate the amount of naphthalene biodegraded during one season by epiphytic microorganisms on magnolia we considered the summer (1.54×10^3 copies cm^{-2}, lowest value) and the spring (3.05×10^4 copies cm^{-2}, highest value) average values of *nah*Ac copy number and their difference as an index of the growth of naphthalene-degrading microorganisms. Assuming that bacteria harbour on average two copies of the gene (Cébron et al., 2008), we can infer that 1.45×10^4 microbial cells per cm^2 grow using naphthalene as carbon source on the magnolia leaf surface. Considering the average weight of a single bacterial cell (10^{-12} g (Davis and Dulbecco, 1973)), and the growth yield on naphthalene (0.5 (Yu et al., 2006; Knightes and Peters, 2003)), we could estimate that the amount of naphthalene biodegraded in one season on magnolia leaves is 7.2 ng of naphthalene per cm^2 of leaf. Although this estimation is affected by high uncertainty due to the aforementioned assumptions, it resulted in the same order of magnitude of the amount of naphthalene accumulated on leaves (5.0 ng cm^{-2}, winter average value; see Table S3), thus suggesting that absorption onto magnolia leaves and biodegradation processes could be considered equally relevant in the removal of naphthalene from the urban air.

The magnitude of contaminant removal by biodegradation on urban tree leaves is therefore likely to have huge impacts at city scale. The estimation of total contaminant removal by tree leaves in a single city or urban region is still difficult to estimate. Considering a leaf area of 12 m^2 of magnolia leaves every m^2 of ground surface (see (Peper et al., 2001) for allometric equation of *Magnolia grandiflora* leaf area), we estimated that magnolia trees can remove approximately 0.864 g of naphthalene per m^2 of ground surface per year in our study area. These results thus confirm the importance of the interaction between plants and phyllosphere bacteria for the removal and degradation of pollutants from the air of urban areas. Indeed, plant-specific microbial communities occur on the leaf surface, and adsorption of pollutants onto leaves may further promote phyllospheric bacteria able to degrade these contaminants.

Data accessibility

Sequence data were submitted to European Nucleotide Archive (EBI-ENA), study accession number PRJEB28871 (http://www.ebi.ac.uk/ena/data/view/PRJEB28871).

Declaration of Competing Interest

The author Maddalena Papacchini is an employee of INAIL, which partially funded the work.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2019.121021.

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SUPPLEMENTARY MATERIAL

Plant-microorganisms interaction promotes removal of air pollutants in Milan (Italy) urban area

Andrea Franzetti[a], Isabella Gandolfi[a], Giuseppina Bestetti[a], Emilio Padoa Schioppa[a], Claudia Canedoli[a], Diego Brambilla[a], David Cappelletti[b], Bartolomeo Sebastiani[b], Ermanno Federici[b], Maddalena Papacchini[d], Roberto Ambrosini[c]

[a] Dept. of Earth and Environmental Sciences (DISAT) - University of Milano-Bicocca, Milano, ITALY

[b] Dipartimento di Chimica, Biologia e Biotecnologie, Università degli Studi di Perugia, Perugia, ITALY

[c] Dept. of Environmental Science and Policy, Università degli Studi di Milano, Milano, ITALY

[d] INAIL, Settore Ricerca, Certificazione e Verifica, Dipartimento di Innovazione Tecnologica (DIT), Laboratorio di Biotecnologie, Rome, Italy

METHODOLOGICAL DETAILS

Leaf surface area estimation

Surface area of magnolia leaves was measured with ImageJ software (NIH, USA) on leaf pictures including a reference ruler. For cedar needles we measured leaf length with a ruler and leaf width with a calliper and then estimated leaf area with the formula (Sellin, 2000):

Area = 2 × length × width.

Biomolecular analyses

Shotgun metagenomics sequencing and sequence processing

Shotgun metagenomics sequencing was performed on 16 samples: 8 samples of magnolia leaves (4 in winter and 4 in summer), and 8 samples of cedar needles (4 in winter and 4 in summer), with Illumina HiSeq 2000 using a 2 × 100 bp paired-end protocol. The paired-end reads were quality-trimmed (minimum length: 80 bp; minimum average quality score: 30) using Sickle (https://github.com/najoshi/sickle).

Filtered reads from all the samples were co-assembled using IDBA-UD (Peng et al., 2012). IDBA-UD iterated the value of k-mer from 40 to 99 (with a step of 5). Predicted genes were inferred from contigs with Prodigal (Hyatt et al., 2010) and annotated with Diamond (blastp) against Uniprot protein database (Buchfink et al., 2014; The UniProt Consortium, 2017). Annotation files were elaborated using MetaCyc database to identify metabolic pathways and enzymatic reactions in the metagenomes. Average per-base coverage of predicted genes was calculated using filtered reads with bowtie2 (Langmead and Salzberg, 2012), SAMtools (Li et al., 2009) and bedtools (Quinlan and Hall, 2010). To account for the different sequencing depth across the samples, sum of gene coverages was normalized to 1,000,000 for each sample.

2.3.4 Quantification of genes coding for naphthalene dioxygenase

Quantitative PCR (qPCR) was used to estimate the abundance of the gene coding for the naphthalene 1,2 dioxygenase of Gram-negative bacteria (*nah*Ac). The target 269-bp fragment was obtained from *Pseudomonas fluorescens* by PCR amplification with the primer pair P1&2 F and P1&2 R (Meynet et al., 2015). The resulting amplicon was cloned in pGEM®-T Easy Vector System (Promega Corporation, Madison, WI, USA) according to the manufacturer's instructions. Serial dilutions of the plasmid were used to build standard concentration curves for qPCR after measuring the concentration of plasmid DNA with a NanoPhotometer® NP80 (Implen GmbH, Munich, Germany). Each qPCR reaction was carried out in a total volume of 10 µL using the FluoCycle II™ Sybr® Master Mix (Euroclone, Pero, Italy) with 0.3 µM of each primer. The amplification was carried out with the Eco Real-Time PCR system (Illumina, San Diego, CA, USA) under the following conditions: initial denaturation at 94 °C for 4 min; 40 cycles at 94 °C for 15 s, 60 °C for 20 s and 72 °C for 12 s, with acquisition of the fluorescence on the FAM channel at the end of each 72 °C elongation step. Dilutions of the standards and of the samples were included in triplicate in each run.

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Table S1 - Sampling dates of PM_{10} and leaves

Date	Season	Collected samples
25/01/2016	Winter	PM_{10} and leaves
27/01/2016	Winter	PM_{10}
30/01/2016	Winter	PM_{10}
02/02/2016	Winter	PM_{10}
05/02/2016	Winter	PM_{10}
07/02/2016	Winter	PM_{10}
11/02/2016	Winter	PM_{10} and leaves
08/04/2016	Spring	PM_{10} and leaves
10/04/2016	Spring	PM_{10}
13/04/2016	Spring	PM_{10}
16/04/2016	Spring	PM_{10}
18/04/2016	Spring	PM_{10}
19/04/2016	Spring	PM_{10}
21/04/2016	Spring	PM_{10}
26/04/2016	Spring	PM_{10} and leaves
23/06/2016	Summer	Leaves
29/06/2016	Summer	PM_{10}
01/07/2016	Summer	PM_{10}
03/07/2016	Summer	PM_{10}
04/07/2016	Summer	PM_{10}
07/07/2016	Summer	PM_{10}
09/07/2016	Summer	PM_{10}
11/07/2016	Summer	Leaves
12/07/2016	Summer	PM_{10}
27/10/2016	Autumn	PM_{10} and leaves
29/10/2016	Autumn	PM_{10}
02/11/2016	Autumn	PM_{10}
04/11/2016	Autumn	PM_{10}
06/11/2016	Autumn	PM_{10}
08/11/2016	Autumn	PM_{10} and leaves

Table S2 – Concentrations of 39 PAH congeners in the analysed samples, expressed in ng m^{-3} of air for PM_{10} samples and in ng g^{-1} of leaf mass for leaf samples (Nap: naphthalene; Acy: acenaphthylene; Ace: acenaphthene; Fln: fluorene; Phe: phenanthrene; Ant: anthracene; Flt: fluoranthene; Pyr: pyrene; Bn21T: benzo[b]naphtho[2, 1-d]thiophene; BghiF: benzo[ghi]fluoranthene; BcP: benzo[c]phenanthrene; Bn12T: benzo[b]naphtho[1, 2-d]thiophene; Bn32T: benzo[b]naphtho[3, 2-d]thiophene; BaA: benz[a]anthracene; CcdP: cyclopenta[cd]pyrene; Tph: triphenylene; Chr: chrysene; BbF: benzo[b]fluoranthene; BkF: benzo[k]fluoranthene; BjF: benzo[j]fluoranthene; BaF: benzo[a]fluoranthene; BeP: benzo[e]pyrene; BaP: benzo[a]pyrene; Per: perylene; IcdF: indeno[1,2,3-cd] fluoranthene; DajA: dibenz[a,j]anthracene; DahA: dibenz[a,h]anthracene; IcdP: indeno[1,2,3-cd]pyrene; DacA: dibenz[a,c]anthracene; BbC: benzo[b]chrysene; Pic: picene; BghiPer: benzo[ghi]perylene; Att: anthanthrene; DalP: dibenzo[a,1]pyrene; DaeP: dibenzo[a,e]pyrene; Cor: coronene; BbPer: benzo[b]perylene; DaiP: dibenzo[a,i]pyrene; DahP: dibenzo[a,h]pyrene).

Sample	Date	Season	Nap	Acy	Ace	Fln	Phe	Ant	Flt	Pyr	Bn21T	BghiF
PM_{10}	25/01/2016	Winter	14.3710	0.5956	0.2127	3.5225	3.0186	0.4871	4.2443	5.3182	0.1368	2.8261
PM_{10}	27/01/2016	Winter	14.7237	0.2868	0.2247	3.3637	1.8923	0.1600	1.7286	1.9045	0.0517	0.9975
PM_{10}	30/01/2016	Winter	13.2000	0.1904	0.2445	3.5419	1.6991	0.1851	1.5185	1.5002	0.0311	0.9979
PM_{10}	02/02/2016	Winter	7.2745	0.1977	0.1820	3.6130	1.6989	0.0934	1.2854	1.2219	0.0290	0.5452
PM_{10}	05/02/2016	Winter	3.2214	0.1380	0.1823	3.4842	1.6126	0.1444	1.1747	1.3366	0.0396	0.5314
PM_{10}	07/02/2016	Winter	4.3909	0.0699	0.2797	5.5777	1.4003	0.0712	0.9292	0.8653	0.0320	0.4084
PM_{10}	11/02/2016	Winter	4.3300	0.2059	0.1784	3.5923	1.7660	0.1420	1.3460	1.2670	0.0194	0.5631
Average relative abundance			0.1892	0.0052	0.0046	0.0821	0.0403	0.0039	0.0376	0.0413	0.0010	0.0211
PM_{10}	08/04/2016	Spring	2.8219	0.1351	0.0998	5.4039	1.2127	0.0783	0.4418	0.1980	0.0057	0.0629
PM_{10}	10/04/2016	Spring	0.1968	0.0128	0.0222	0.1522	0.1127	0.0173	0.1065	0.1118	0.0013	0.0464
PM_{10}	13/04/2016	Spring	0.2121	0.0168	0.0127	0.0459	0.0834	0.0150	0.0866	0.0980	0.0012	0.0446
PM_{10}	16/04/2016	Spring	0.1394	0.0108	0.0118	0.1674	0.1056	0.0215	0.1009	0.1060	0.0011	0.0332
PM_{10}	18/04/2016	Spring	0.2429	0.0181	0.0238	0.2223	0.1180	0.0221	0.1185	0.1148	0.0013	0.0543
PM_{10}	19/04/2016	Spring	1.5536	0.1199	0.1664	5.1413	1.1312	0.0518	0.4243	0.2542	0.0090	0.0477
PM_{10}	21/04/2016	Spring	0.2359	0.0194	0.0183	0.1440	0.1388	0.0272	0.1747	0.1643	0.0032	0.0551
PM_{10}	26/04/2016	Spring	0.0602	0.0216	0.0395	0.2326	0.0901	0.0302	0.0948	0.1028	0.0010	0.0554
Average relative abundance			0.1561	0.0101	0.0113	0.3290	0.0855	0.0075	0.0443	0.0329	0.0007	0.0114
PM_{10}	29/06/2016	Summer	0.0231	0.0074	0.0023	0.0075	0.0413	0.0062	0.0404	0.0476	0.0007	0.0107
PM_{10}	01/07/2016	Summer	0.0334	0.0104	0.0423	0.0484	0.0818	0.0163	0.0761	0.0778	0.0009	0.0176
PM_{10}	03/07/2016	Summer	0.0195	0.0021	0.0052	0.0157	0.0394	0.0069	0.0408	0.0707	0.0000	0.0165
PM_{10}	04/07/2016	Summer	0.0282	0.0031	0.0099	0.0175	0.0664	0.0085	0.0711	0.1004	0.0000	0.0250
PM_{10}	07/07/2016	Summer	0.0361	0.0025	0.0081	0.0213	0.0766	0.0083	0.1051	0.0895	0.0034	0.0028
PM_{10}	09/07/2016	Summer	0.0444	0.0056	0.0109	0.0328	0.1273	0.0177	0.4985	0.4937	0.0040	0.1839
PM_{10}	12/07/2016	Summer	0.0349	0.0059	0.0117	0.0273	0.2142	0.0178	0.2506	0.2944	0.0034	0.0758
Average relative abundance			0.0182	0.0031	0.0075	0.0141	0.0536	0.0068	0.0897	0.0973	0.0010	0.0275
PM_{10}	27/10/2016	Autumn	0.0320	0.0314	0.0250	0.0763	0.2264	0.0381	0.2686	0.3426	0.0000	0.1180
PM_{10}	29/10/2016	Autumn	0.1886	0.0449	0.0185	0.0520	0.3220	0.0613	0.4330	0.5470	0.0000	0.2448
PM_{10}	02/11/2016	Autumn	0.2767	0.0968	0.0307	0.0935	0.5568	0.1289	0.7535	0.9702	0.0000	0.6161
PM_{10}	04/11/2016	Autumn	0.2657	0.0179	0.0188	0.0554	0.1932	0.0677	0.2869	0.3200	0.0026	0.1768
PM_{10}	06/11/2016	Autumn	0.2270	0.0392	0.0333	0.0625	0.3001	0.0686	0.4854	0.7022	0.0000	0.3223
PM_{10}	08/11/2016	Autumn	0.3686	0.1037	0.0335	0.1211	0.9113	0.2998	1.6400	2.2968	0.0014	2.0563
Average relative abundance			0.0105	0.0026	0.0012	0.0036	0.0194	0.0051	0.0300	0.0401	0.0000	0.0274

Table S2 – continued

Sample	Date	Season	Nap	Acy	Ace	Fln	Phe	Ant	Flt	Pyr	Bn21T	BghiF
Magnolia	25/01/2016	Winter	123.4654	4.5885	5.1421	67.9301	31.2470	2.0895	20.6460	18.3976	0.4116	4.2139
Magnolia	25/01/2016	Winter	221.1775	4.9787	6.0868	88.5437	36.6522	2.9281	26.9448	23.6095	0.3242	4.3802
Magnolia	25/01/2016	Winter	162.2320	6.0304	8.9515	76.8259	47.4510	6.4557	27.9905	24.6312	0.4870	4.5725
Magnolia	11/02/2016	Winter	90.8575	3.2691	4.9673	47.7028	30.2089	2.2406	17.5541	15.4399	0.3547	2.8274
Magnolia	11/02/2016	Winter	138.0572	3.9237	5.7997	71.2532	33.0526	3.5123	23.5042	19.8496	0.3353	0.8100
Magnolia	11/02/2016	Winter	178.0902	4.7938	7.1317	81.1453	38.2985	3.0724	19.9814	16.6690	0.3938	2.8388
Average relative abundance			0.4342	0.0131	0.0181	0.2059	0.1031	0.0096	0.0649	0.0563	0.0011	0.0093
Magnolia	08/04/2016	Spring	16.1598	0.3347	0.2737	1.4515	5.1207	0.5052	11.6866	10.9363	0.0207	1.7859
Magnolia	08/04/2016	Spring	9.4480	0.1615	0.4293	1.3875	2.2345	0.3270	6.2738	6.0153	0.0160	1.3946
Magnolia	08/04/2016	Spring	20.7114	1.3361	4.3882	10.8120	4.1749	0.3544	8.3437	8.5215	0.0000	1.4701
Magnolia	26/04/2016	Spring	102.2410	12.7146	22.0641	37.1711	3.3951	0.3033	7.4914	8.0052	0.0000	1.6310
Magnolia	26/04/2016	Spring	29.3952	3.8226	2.2200	6.9063	2.7255	0.2054	3.7802	5.6961	0.0175	0.8889
Magnolia	26/04/2016	Spring	1.6993	0.0712	0.1953	0.7784	2.0179	0.1810	2.5047	1.3200	0.0244	0.1511
Average relative abundance			0.3180	0.0326	0.0523	0.1036	0.0348	0.0033	0.0710	0.0717	0.0001	0.0130
Magnolia	23/06/2016	Summer	3.9345	0.3629	0.5322	2.7059	1.1738	0.1132	0.4024	0.2870	0.0101	0.0890
Magnolia	23/06/2016	Summer	2.3311	0.1491	0.2038	0.8858	1.6711	0.1267	0.7221	0.4679	0.0000	0.0756
Magnolia	23/06/2016	Summer	10.4342	0.7921	1.3301	4.3347	1.8198	0.1538	0.8508	0.6873	0.0199	0.1367
Magnolia	11/07/2016	Summer	1.5256	0.2199	0.2739	1.7479	1.2521	0.0807	0.4155	0.2614	0.0000	0.0885
Magnolia	11/07/2016	Summer	11.1388	1.0607	1.4969	4.5948	1.7718	0.1954	0.7142	0.5879	0.0132	0.1005
Magnolia	11/07/2016	Summer	5.5870	0.6282	0.8979	4.6013	1.7498	0.1359	0.6528	0.5484	0.0000	0.1155
Average relative abundance			0.3350	0.0308	0.0454	0.1809	0.0905	0.0077	0.0360	0.0272	0.0004	0.0058
Magnolia	27/10/2016	Autumn	1.2026	0.5590	0.8725	25.4349	13.2222	0.5823	8.0155	6.6342	0.2873	1.7684
Magnolia	27/10/2016	Autumn	18.0637	2.9888	3.7235	25.9772	16.5188	1.4152	13.4616	12.3812	0.2504	1.9941
Magnolia	27/10/2016	Autumn	1.0342	1.1300	2.7681	32.0518	18.3342	0.9925	14.3531	13.5931	0.9958	2.4678
Magnolia	08/11/2016	Autumn	14.1771	0.7878	0.9422	19.2517	14.3874	1.0422	11.9230	10.7035	0.2711	2.1715
Magnolia	08/11/2016	Autumn	17.0486	0.8258	1.2882	17.1756	17.2848	1.1676	19.9248	16.3188	0.6630	2.8238
Magnolia	08/11/2016	Autumn	4.0387	1.9796	2.3522	35.7455	20.4565	1.4082	15.2219	14.8599	0.5611	2.5108
Average relative abundance			0.0816	0.0121	0.0175	0.2285	0.1471	0.0097	0.1217	0.1094	0.0044	0.0202

Table S2 – continued

Sample	Date	Season	Nap	Acy	Ace	Fln	Phe	Ant	Flt	Pyr	Bn21T	BghiF
Cedar	25/01/2016	Winter	978.7795	32.1149	10.8970	121.1399	100.5751	7.3403	64.6495	63.4978	1.3373	16.4362
Cedar	25/01/2016	Winter	323.0645	7.8073	6.7501	42.7378	35.2089	3.1897	21.1542	20.7482	0.6287	6.5062
Cedar	25/01/2016	Winter	110.1725	4.6131	3.3827	34.5004	23.7570	1.1752	12.6073	11.8990	0.4019	3.7662
Cedar	11/02/2016	Winter	111.1155	9.8052	5.0666	43.6096	39.2193	2.7925	27.3277	25.1580	0.6252	7.4088
Cedar	11/02/2016	Winter	322.8828	10.7469	5.5863	63.7453	38.3709	2.4343	20.0796	18.5919	0.5616	4.4938
Cedar	11/02/2016	Winter	121.2313	9.3121	4.5112	39.7264	30.3682	2.0180	20.5392	19.9565	0.6715	7.5297
Cedar	11/02/2016	Winter	22.1775	2.1743	2.5422	2.3028	22.4774	0.9401	22.8810	24.5150	0.0246	5.3506
Average relative abundance			0.5463	0.0210	0.0106	0.0955	0.0796	0.0055	0.0520	0.0506	0.0012	0.0141
Cedar	08/04/2016	Spring	17.2921	9.6045	3.8065	42.5104	26.2878	1.0238	17.2347	23.2175	0.0595	1.2286
Cedar	08/04/2016	Spring	24.9984	8.1029	2.8412	24.5516	38.6977	1.4877	12.9912	16.7688	0.1123	1.7603
Cedar	08/04/2016	Spring	123.6241	2.8917	1.8928	25.5739	13.5500	0.4066	7.2339	8.5535	0.0271	0.9742
Cedar	26/04/2016	Spring	24.3572	9.5853	5.6719	42.8708	24.3516	1.2419	13.7508	18.7655	0.0420	0.9263
Cedar	26/04/2016	Spring	4.0688	2.2795	0.9734	7.6487	14.0408	0.3872	5.5469	7.9127	0.0281	0.6910
Cedar	26/04/2016	Spring	74.9021	6.2504	2.6591	27.5391	19.2223	1.8229	18.7672	16.7266	0.9962	5.4493
Average relative abundance			0.2565	0.0369	0.0170	0.1626	0.1297	0.0061	0.0720	0.0876	0.0012	0.0105
Cedar	23/06/2016	Summer	6.2811	0.8295	1.2109	7.3559	2.5791	0.5645	3.5456	4.5155	0.0254	0.5084
Cedar	23/06/2016	Summer	1.7435	0.3591	0.4463	4.1670	11.7420	0.9653	3.2310	3.6606	0.0336	0.3459
Cedar	23/06/2016	Summer	4.4693	1.3879	1.5755	8.2702	5.5390	0.2380	1.4534	1.5850	0.0302	0.0115
Cedar	23/06/2016	Summer	1.8731	0.5461	0.7292	3.3878	7.3718	1.5818	3.3199	3.7488	0.0000	0.3990
Cedar	11/07/2016	Summer	3.2741	0.4961	0.9717	6.6564	16.6944	0.5062	2.8816	3.1788	0.0266	0.2784
Cedar	11/07/2016	Summer	8.6912	0.6388	1.0090	8.0541	6.9509	0.7826	3.1015	3.2878	0.0195	0.1699
Cedar	11/07/2016	Summer	4.7962	0.5392	0.8331	5.1471	14.0908	0.2753	1.7892	1.9634	0.0107	0.2047
Cedar	11/07/2016	Summer	2.1656	0.3460	0.4462	2.9202	5.2806	0.5837	1.8367	2.0825	0.0082	0.2209
Average relative abundance			0.1172	0.0181	0.0254	0.1618	0.2473	0.0193	0.0745	0.0846	0.0005	0.0075
Cedar	27/10/2016	Autumn	10.8717	2.3601	3.7246	21.2195	21.9610	0.5605	6.3882	7.4486	0.0425	1.5501
Cedar	27/10/2016	Autumn	2.7971	0.5980	0.4183	5.0551	16.2443	0.8683	5.4505	6.3292	0.0221	1.1571
Cedar	27/10/2016	Autumn	3.0800	1.6781	1.5350	10.1413	7.8784	0.9160	6.3899	6.8199	0.0178	0.8919
Cedar	08/11/2016	Autumn	14.2975	3.2011	3.0547	18.0988	25.2790	0.9260	10.7276	11.7847	0.1002	2.1461
Cedar	08/11/2016	Autumn	13.0715	1.7855	3.2831	13.1201	22.8232	0.9021	9.5887	10.9873	0.1110	2.3127
Cedar	08/11/2016	Autumn	16.2825	3.8196	3.3880	17.1540	11.3504	0.4427	6.4960	6.9671	0.0613	1.1645
Average relative abundance			0.1176	0.0262	0.0300	0.1651	0.2055	0.0090	0.0877	0.0980	0.0007	0.0180

Table S2 – continued

Sample	Date	Season	BcP	Bn12T	Bn32T	BaA	CcdP	Tph	Chr	BbF	BkF	BjF
PM_{10}	25/01/2016	Winter	1.0599	0.0841	0.0388	5.2879	2.8413	1.3283	5.6339	5.2194	3.1308	3.4109
PM_{10}	27/01/2016	Winter	0.3606	0.0157	0.0073	1.6244	0.4775	0.6282	2.3375	3.0197	1.5231	1.7816
PM_{10}	30/01/2016	Winter	0.3308	0.0135	0.0057	1.6094	0.4866	0.5935	2.1343	2.3962	1.1476	1.3702
PM_{10}	02/02/2016	Winter	0.2158	0.0101	0.0056	0.9807	0.2522	0.2961	1.3682	1.6697	0.8627	0.9599
PM_{10}	05/02/2016	Winter	0.1839	0.0131	0.0073	0.9152	0.2783	0.3599	1.2042	1.2996	0.6365	0.7756
PM_{10}	07/02/2016	Winter	0.1396	0.0096	0.0069	0.6295	0.1824	0.2649	0.9051	1.3356	0.7280	0.7686
PM_{10}	11/02/2016	Winter	0.1853	0.0090	0.0050	0.8647	0.2842	0.3346	1.1121	1.2941	0.6745	0.7432
Average relative abundance			0.0076	0.0005	0.0002	0.0366	0.0148	0.0117	0.0452	0.0499	0.0268	0.0302
PM_{10}	08/04/2016	Spring	0.0193	0.0048	0.0048	0.0559	0.0235	0.0497	0.1284	0.1408	0.0603	0.0897
PM_{10}	10/04/2016	Spring	0.0120	0.0005	0.0014	0.0435	0.0048	0.0303	0.0794	0.1432	0.0594	0.0827
PM_{10}	13/04/2016	Spring	0.0091	0.0009	0.0006	0.0492	0.0042	0.0303	0.0799	0.0913	0.0429	0.0492
PM_{10}	16/04/2016	Spring	0.0074	0.0007	0.0010	0.0402	0.0091	0.0241	0.0624	0.1450	0.0683	0.0811
PM_{10}	18/04/2016	Spring	0.0141	0.0011	0.0019	0.0437	0.0043	0.0386	0.0883	0.3550	0.1411	0.1905
PM_{10}	19/04/2016	Spring	0.0139	0.0071	0.0056	0.0536	0.0209	0.0422	0.1015	0.1371	0.0628	0.0854
PM_{10}	21/04/2016	Spring	0.0163	0.0023	0.0000	0.0698	0.0071	0.0415	0.1082	0.2839	0.1210	0.1528
PM_{10}	26/04/2016	Spring	0.0118	0.0000	0.0000	0.0677	0.0061	0.0353	0.1168	0.2162	0.1039	0.1006
Average relative abundance			0.0030	0.0005	0.0004	0.0121	0.0023	0.0083	0.0219	0.0432	0.0189	0.0238
PM_{10}	29/06/2016	Summer	0.0032	0.0007	0.0000	0.0135	0.0016	0.0094	0.0262	0.0409	0.0217	0.0180
PM_{10}	01/07/2016	Summer	0.0047	0.0000	0.0000	0.0185	0.0027	0.0125	0.0490	0.0576	0.0229	0.0230
PM_{10}	03/07/2016	Summer	0.0038	0.0000	0.0000	0.0182	0.0025	0.0129	0.0309	0.0504	0.0130	0.0227
PM_{10}	04/07/2016	Summer	0.0047	0.0000	0.0000	0.0286	0.0039	0.0216	0.0562	0.0670	0.0223	0.0310
PM_{10}	07/07/2016	Summer	0.0024	0.0217	0.0030	0.0000	0.0000	0.0399	0.0009	0.0488	0.0168	0.0184
PM_{10}	09/07/2016	Summer	0.0524	0.0015	0.0014	0.1896	0.0189	0.1163	0.3376	0.6715	0.3341	0.2468
PM_{10}	12/07/2016	Summer	0.0126	0.0022	0.0018	0.0548	0.0034	0.0581	0.1295	0.1332	0.0642	0.0540
Average relative abundance			0.0069	0.0022	0.0005	0.0268	0.0027	0.0224	0.0522	0.0886	0.0410	0.0343
PM_{10}	27/10/2016	Autumn	0.0266	0.0037	0.0026	0.1883	0.0424	0.0707	0.2521	0.4723	0.2425	0.2202
PM_{10}	29/10/2016	Autumn	0.0581	0.0038	0.0024	0.5256	0.1078	0.1522	0.6176	1.3785	0.8342	0.6440
PM_{10}	02/11/2016	Autumn	0.1424	0.0064	0.0029	1.3951	0.3283	0.2822	1.3533	2.7638	1.9136	1.3140
PM_{10}	04/11/2016	Autumn	0.0409	0.0022	0.0009	0.3043	0.0255	0.0738	0.3911	0.6517	0.4142	0.3330
PM_{10}	06/11/2016	Autumn	0.0737	0.0031	0.0014	0.6022	0.1730	0.1205	0.6476	1.2310	0.7533	0.6179
PM_{10}	08/11/2016	Autumn	0.4544	0.0071	0.0027	4.9315	0.8029	0.6044	3.9091	5.0292	3.8869	2.3300
Average relative abundance			0.0062	0.0002	0.0001	0.0615	0.0115	0.0101	0.0555	0.0893	0.0623	0.0423

Table S2 – continued

Sample	Date	Season	BcP	Bn12T	Bn32T	BaA	CcdP	Tph	Chr	BbF	BkF	BjF
Magnolia	25/01/2016	Winter	2.6739	0.2592	0.0433	3.5008	0.1959	2.2997	6.9398	2.8105	1.2445	1.3802
Magnolia	25/01/2016	Winter	2.5261	0.2334	0.0477	3.0084	0.1880	2.0987	5.9911	2.3886	0.9935	1.1281
Magnolia	25/01/2016	Winter	2.6833	0.2727	0.0712	3.4809	0.3129	2.2994	6.6098	4.5093	2.4828	2.4460
Magnolia	11/02/2016	Winter	1.8560	0.2535	0.0345	2.2912	0.1325	1.3957	4.3646	2.2626	1.0829	1.1040
Magnolia	11/02/2016	Winter	2.0787	0.1604	0.0540	2.3877	0.1603	1.6616	4.7910	2.1542	0.9678	1.0430
Magnolia	11/02/2016	Winter	1.9184	0.2603	0.0463	2.2178	0.1441	1.7122	5.0448	2.2701	1.1077	1.1836
Average relative abundance			0.0065	0.0007	0.0001	0.0080	0.0005	0.0054	0.0160	0.0078	0.0037	0.0039
Magnolia	08/04/2016	Spring	1.8235	0.0000	0.0000	1.5902	0.0581	1.9269	5.6542	2.7287	1.1773	1.6490
Magnolia	08/04/2016	Spring	1.2438	0.0000	0.0000	1.2392	0.0282	1.1287	3.6444	1.0342	0.3574	0.6942
Magnolia	08/04/2016	Spring	1.4587	0.0000	0.0000	1.5396	0.0450	1.6006	4.8827	2.1719	0.9307	1.4648
Magnolia	26/04/2016	Spring	1.5064	0.0000	0.0000	1.4165	0.0266	1.6741	4.8779	1.2078	0.4440	0.6874
Magnolia	26/04/2016	Spring	0.7765	0.0145	0.0303	0.7867	0.0290	0.7549	2.3068	0.7254	0.3592	0.6084
Magnolia	26/04/2016	Spring	0.0885	0.0000	0.0000	0.1232	0.0000	0.1069	0.4212	0.1901	0.0874	0.1642
Average relative abundance			0.0122	0.0000	0.0001	0.0119	0.0003	0.0127	0.0386	0.0143	0.0059	0.0093
Magnolia	23/06/2016	Summer	0.0595	0.0000	0.0000	0.1039	0.0391	0.1040	0.3356	0.3444	0.1277	0.2179
Magnolia	23/06/2016	Summer	0.0438	0.0000	0.0000	0.0451	0.0052	0.1635	0.1628	0.1412	0.0375	0.0519
Magnolia	23/06/2016	Summer	0.0638	0.0121	0.0070	0.1209	0.0538	0.2635	0.4698	0.9926	0.3557	0.5463
Magnolia	11/07/2016	Summer	0.0320	0.0000	0.0000	0.0427	0.0175	0.1417	0.1733	0.1857	0.0532	0.0760
Magnolia	11/07/2016	Summer	0.0558	0.0067	0.0000	0.1105	0.0441	0.3106	0.4142	0.9582	0.3180	0.4671
Magnolia	11/07/2016	Summer	0.0792	0.0000	0.0000	0.1000	0.0153	0.3808	0.4107	0.3755	0.0795	0.1564
Average relative abundance			0.0032	0.0002	0.0001	0.0050	0.0017	0.0131	0.0188	0.0287	0.0093	0.0145
Magnolia	27/10/2016	Autumn	0.6896	0.1021	0.0527	1.2390	0.1955	1.1585	3.4791	0.8389	0.5164	0.4107
Magnolia	27/10/2016	Autumn	0.8757	0.0508	0.0412	1.3590	0.1932	1.3625	3.2029	0.8385	0.3071	0.3391
Magnolia	27/10/2016	Autumn	1.0928	0.5600	0.3736	2.2735	0.2755	1.7821	5.4632	1.5291	0.5015	0.9935
Magnolia	08/11/2016	Autumn	1.1942	0.0840	0.0630	1.8639	0.2386	1.7359	4.4384	1.0848	0.4525	0.7980
Magnolia	08/11/2016	Autumn	1.1394	0.2618	0.2052	2.0419	0.2454	1.8404	4.4400	1.2570	0.4409	0.6432
Magnolia	08/11/2016	Autumn	1.1248	0.1513	0.1119	2.2885	0.3639	1.6094	5.6768	1.4953	0.6596	0.9250
Average relative abundance			0.0090	0.0018	0.0012	0.0162	0.0022	0.0139	0.0392	0.0103	0.0042	0.0060

Table S2 – continued

Sample	Date	Season	BcP	Bn12T	Bn32T	BaA	CcdP	Tph	Chr	BbF	BkF	BjF
Cedar	25/01/2016	Winter	10.0980	0.5431	0.1335	18.2904	0.9800	11.0371	38.0854	12.8330	6.5932	6.8385
Cedar	25/01/2016	Winter	3.2060	0.2954	0.0753	6.2294	0.2856	3.8574	11.8555	4.8456	2.2215	2.4319
Cedar	25/01/2016	Winter	2.3331	0.1849	0.0462	3.6050	0.1531	3.1780	10.2160	4.0949	1.7619	2.0136
Cedar	11/02/2016	Winter	4.3731	0.2734	0.0780	7.9730	0.4082	5.0476	17.1971	6.5596	3.3111	3.4709
Cedar	11/02/2016	Winter	2.4860	0.1934	0.0561	4.7662	0.1961	2.6676	9.4793	3.9443	1.8961	1.9780
Cedar	11/02/2016	Winter	4.7801	0.3833	0.0685	7.2311	0.3684	6.2552	18.1333	7.0001	3.1694	3.6551
Cedar	11/02/2016	Winter	3.0079	0.0193	0.0185	5.0501	0.2586	7.8561	16.4973	3.5393	1.3068	2.1312
Average relative abundance			0.0083	0.0005	0.0001	0.0146	0.0007	0.0110	0.0334	0.0118	0.0056	0.0062
Cedar	08/04/2016	Spring	2.7910	0.0531	0.1014	2.1447	0.0708	3.3876	10.4396	2.3268	1.1404	2.0035
Cedar	08/04/2016	Spring	1.3611	0.0242	0.0307	2.5735	0.1147	2.3535	8.2985	2.8247	1.0038	1.7483
Cedar	08/04/2016	Spring	0.9817	0.0190	0.0414	1.5074	0.0874	2.2684	6.2624	1.6535	0.6326	1.2670
Cedar	26/04/2016	Spring	2.5674	0.0451	0.0387	2.4535	0.0384	4.3492	10.6676	1.9105	0.6309	1.7547
Cedar	26/04/2016	Spring	0.6300	0.0160	0.0000	1.0748	0.0426	1.3605	4.0226	1.4948	0.4568	1.0245
Cedar	26/04/2016	Spring	4.0004	0.5511	0.1440	6.3438	0.2666	5.2016	15.0789	5.6144	2.2809	2.7028
Average relative abundance			0.0117	0.0007	0.0003	0.0153	0.0006	0.0180	0.0522	0.0151	0.0059	0.0100
Cedar	23/06/2016	Summer	0.4397	0.0137	0.0000	0.5082	0.0298	1.8641	3.1422	0.5236	0.1410	0.3104
Cedar	23/06/2016	Summer	0.1807	0.0162	0.0142	0.3896	0.0246	1.3641	1.8279	0.8561	0.2889	0.5641
Cedar	23/06/2016	Summer	0.1420	0.1201	0.0000	0.1612	0.0126	1.0271	0.9836	0.3940	0.0534	0.1890
Cedar	23/06/2016	Summer	0.2579	0.0000	0.0000	0.2691	0.0616	1.7136	1.8151	0.6791	0.2015	0.4114
Cedar	11/07/2016	Summer	0.1692	0.0102	0.0000	0.3223	0.0287	1.8422	1.7563	1.1741	0.2970	0.4752
Cedar	11/07/2016	Summer	0.1302	0.0111	0.0179	0.1733	0.0209	0.6594	0.9886	0.3641	0.1128	0.1683
Cedar	11/07/2016	Summer	0.1335	0.0084	0.0000	0.2456	0.0188	1.0876	1.2665	0.6637	0.1412	0.3467
Cedar	11/07/2016	Summer	0.1159	0.0121	0.0000	0.1645	0.0307	0.9316	1.0488	0.3017	0.1077	0.1801
Average relative abundance			0.0055	0.0007	0.0001	0.0079	0.0008	0.0369	0.0452	0.0174	0.0047	0.0093
Cedar	27/10/2016	Autumn	0.6524	0.0182	0.0221	1.8812	0.1324	2.3820	4.8060	1.6061	0.5735	1.0496
Cedar	27/10/2016	Autumn	0.4843	0.0000	0.0000	1.1537	0.1362	2.3962	3.3397	1.1420	0.4632	0.6740
Cedar	27/10/2016	Autumn	0.4687	0.0118	0.0134	1.3303	0.2280	1.6873	3.2758	1.1287	0.2830	0.6736
Cedar	08/11/2016	Autumn	0.8648	0.0528	0.0447	2.6437	0.2057	3.6242	6.4519	2.8114	1.1594	1.4306
Cedar	08/11/2016	Autumn	0.9328	0.0462	0.0668	1.9683	0.1512	3.6304	6.2231	1.9045	0.7117	1.2372
Cedar	08/11/2016	Autumn	0.5679	0.0248	0.0125	1.0177	0.1421	2.5149	3.4611	1.2182	0.5629	0.6896
Average relative abundance			0.0077	0.0003	0.0003	0.0195	0.0019	0.0316	0.0537	0.0191	0.0073	0.0112

Table S2 – continued

Sample	Date	Season	BaF	BeP	BaP	Per	IcdF	DajA	DahA	IcdP	DacA	BbC
PM_{10}	25/01/2016	Winter	1.0584	4.1712	7.7794	1.3746	0.6586	0.2007	0.2644	5.9255	0.6257	0.7542
PM_{10}	27/01/2016	Winter	0.3056	2.3773	2.7737	0.4088	0.2997	0.0997	0.1448	3.1881	0.2811	0.1658
PM_{10}	30/01/2016	Winter	0.2820	1.7923	2.2332	0.3423	0.2817	0.0884	0.1343	2.3417	0.2343	0.2238
PM_{10}	02/02/2016	Winter	0.1887	1.2668	1.5438	0.2307	0.1914	0.0596	0.1380	1.8514	0.2320	0.1723
PM_{10}	05/02/2016	Winter	0.1623	1.0176	1.3317	0.2011	0.0953	0.0394	0.0648	1.1186	0.1062	0.0854
PM_{10}	07/02/2016	Winter	0.1550	1.0439	1.2006	0.1944	0.1267	0.0647	0.0760	1.3064	0.1353	0.1198
PM_{10}	11/02/2016	Winter	0.1908	1.0358	1.4774	0.2238	0.1269	0.0374	0.3028	1.2810	0.1443	0.1340
Average relative abundance			0.0072	0.0391	0.0564	0.0092	0.0055	0.0018	0.0035	0.0523	0.0054	0.0051
PM_{10}	08/04/2016	Spring	0.0090	0.1088	0.0813	0.0197	0.0140	0.0389	0.0204	0.1616	0.0309	0.0123
PM_{10}	10/04/2016	Spring	0.0055	0.1058	0.0738	0.0138	0.0126	0.0134	0.0117	0.1260	0.0148	0.0056
PM_{10}	13/04/2016	Spring	0.0063	0.0707	0.0582	0.0134	0.0096	0.0132	0.0123	0.0893	0.0127	0.0070
PM_{10}	16/04/2016	Spring	0.0069	0.0986	0.0870	0.0198	0.0124	0.0096	0.0114	0.1034	0.0138	0.0086
PM_{10}	18/04/2016	Spring	0.0056	0.2787	0.1048	0.0190	0.0164	0.0113	0.0092	0.1694	0.0086	0.0032
PM_{10}	19/04/2016	Spring	0.0129	0.1246	0.1096	0.0322	0.0184	0.0387	0.0217	0.1596	0.0270	0.0142
PM_{10}	21/04/2016	Spring	0.0147	0.2102	0.1423	0.0432	0.0296	0.0439	0.0426	0.1923	0.0438	0.0242
PM_{10}	26/04/2016	Spring	0.0074	0.1471	0.1300	0.0284	0.0123	0.0171	0.0157	0.1637	0.0165	0.0095
Average relative abundance			0.0020	0.0327	0.0225	0.0054	0.0036	0.0053	0.0041	0.0333	0.0048	0.0024
PM_{10}	29/06/2016	Summer	0.0020	0.0310	0.0239	0.0031	0.0039	0.0053	0.0080	0.0392	0.0054	0.0000
PM_{10}	01/07/2016	Summer	0.0029	0.0469	0.0308	0.0071	0.0069	0.0059	0.0067	0.0403	0.0065	0.0000
PM_{10}	03/07/2016	Summer	0.0017	0.0455	0.0307	0.0033	0.0061	0.0031	0.0029	0.0502	0.0026	0.0000
PM_{10}	04/07/2016	Summer	0.0023	0.0795	0.0626	0.0089	0.0088	0.0060	0.0037	0.0680	0.0037	0.0000
PM_{10}	07/07/2016	Summer	0.0010	0.0333	0.0203	0.0034	0.0063	0.0047	0.0067	0.0462	0.0072	0.0000
PM_{10}	09/07/2016	Summer	0.0131	0.2990	0.2472	0.0264	0.0740	0.0537	0.0535	0.6555	0.0639	0.0229
PM_{10}	12/07/2016	Summer	0.0030	0.1236	0.0613	0.0107	0.0161	0.0112	0.0084	0.1082	0.0088	0.0043
Average relative abundance			0.0022	0.0546	0.0395	0.0052	0.0101	0.0074	0.0075	0.0835	0.0081	0.0023
PM_{10}	27/10/2016	Autumn	0.0207	0.3164	0.3099	0.0513	0.0532	0.0569	0.0419	0.5627	0.0473	0.0315
PM_{10}	29/10/2016	Autumn	0.0631	0.8520	1.1160	0.1624	0.1441	0.1184	0.1215	1.5357	0.1325	0.0613
PM_{10}	02/11/2016	Autumn	0.1726	1.5552	2.6744	0.4487	0.2224	0.2198	0.2372	3.1513	0.2787	0.1638
PM_{10}	04/11/2016	Autumn	0.0188	0.3809	0.4279	0.0759	0.0674	0.0621	0.0518	0.7065	0.0543	0.0260
PM_{10}	06/11/2016	Autumn	0.0723	0.6917	1.1255	0.2270	0.1307	0.1225	0.1170	1.6409	0.1466	0.0637
PM_{10}	08/11/2016	Autumn	0.3298	2.4862	5.3855	0.9478	0.4539	0.3903	0.4093	6.1946	0.4989	0.2524
Average relative abundance			0.0052	0.0487	0.0855	0.0148	0.0083	0.0075	0.0076	0.1068	0.0090	0.0046

Table S2 – continued

Sample	Date	Season	BaF	BeP	BaP	Per	IcdF	DajA	DahA	IcdP	DacA	BbC
Magnolia	25/01/2016	Winter	0.0503	1.7786	1.1374	0.1376	0.2185	0.3295	0.0478	1.3355	0.1029	0.1078
Magnolia	25/01/2016	Winter	0.0233	1.3305	0.5664	0.0487	0.1475	0.0934	0.0679	0.8700	0.0807	0.0461
Magnolia	25/01/2016	Winter	0.1037	3.2210	2.8645	0.3294	0.3179	0.2314	0.1041	2.5334	0.2249	0.1504
Magnolia	11/02/2016	Winter	0.0486	1.4524	0.8223	0.0862	0.1750	0.1253	0.0535	1.2501	0.1026	0.0738
Magnolia	11/02/2016	Winter	0.0447	1.3139	0.7827	0.0786	0.1635	0.2726	0.0479	0.9363	0.0718	0.0554
Magnolia	11/02/2016	Winter	0.0517	1.4664	0.9318	0.1006	0.1733	0.1066	0.0513	0.9900	0.0863	0.0762
Average relative abundance			0.0002	0.0050	0.0034	0.0004	0.0006	0.0006	0.0002	0.0038	0.0003	0.0002
Magnolia	08/04/2016	Spring	0.0625	2.0871	0.7241	0.1850	0.1852	0.2209	0.1251	1.7329	0.2269	0.5900
Magnolia	08/04/2016	Spring	0.0000	0.6179	0.2179	0.0558	0.0577	0.0416	0.0478	0.4655	0.0827	0.1949
Magnolia	08/04/2016	Spring	0.0818	1.6397	0.7863	0.2851	0.1432	0.1558	0.1168	1.3052	0.1737	0.3994
Magnolia	26/04/2016	Spring	0.0481	0.7212	0.2794	0.1077	0.0527	0.0379	0.0601	0.3674	0.0643	0.0408
Magnolia	26/04/2016	Spring	0.0000	0.5264	0.1283	0.0367	0.0250	0.0250	0.0301	0.2905	0.0353	0.0685
Magnolia	26/04/2016	Spring	0.0120	0.0722	0.0287	0.0000	0.0172	0.0250	0.0237	0.1105	0.0442	0.1604
Average relative abundance			0.0004	0.0100	0.0038	0.0012	0.0009	0.0009	0.0007	0.0076	0.0011	0.0026
Magnolia	23/06/2016	Summer	0.0051	0.1966	0.1132	0.0147	0.1098	0.0809	0.0838	0.2973	0.1546	0.0155
Magnolia	23/06/2016	Summer	0.0036	0.0801	0.0387	0.0110	0.0919	0.0000	0.0443	0.1105	0.0451	0.0000
Magnolia	23/06/2016	Summer	0.0151	0.6628	0.1766	0.0525	0.3079	0.2431	0.2529	1.1057	0.2995	0.1380
Magnolia	11/07/2016	Summer	0.0045	0.1044	0.0458	0.0186	0.0440	0.0366	0.1074	0.2068	0.0621	0.0190
Magnolia	11/07/2016	Summer	0.0118	0.5805	0.1263	0.0192	0.2874	0.0787	0.0797	0.9701	0.0922	0.0241
Magnolia	11/07/2016	Summer	0.0076	0.2520	0.1214	0.0248	0.1250	0.0375	0.0339	0.3425	0.0411	0.0152
Average relative abundance			0.0005	0.0180	0.0060	0.0014	0.0093	0.0046	0.0058	0.0291	0.0067	0.0020
Magnolia	27/10/2016	Autumn	0.0251	1.1335	0.5795	0.1319	0.3420	0.5604	0.4282	1.5981	0.7072	0.3191
Magnolia	27/10/2016	Autumn	0.0349	1.0220	0.5368	0.1535	0.1828	0.3035	0.3534	1.1150	0.4749	0.2629
Magnolia	27/10/2016	Autumn	0.0852	2.6034	1.3468	0.4070	0.3888	0.5958	0.5851	2.2135	0.7348	0.4326
Magnolia	08/11/2016	Autumn	0.0407	1.4971	0.6716	0.2214	0.6010	0.5930	0.5803	1.8195	0.9737	0.2420
Magnolia	08/11/2016	Autumn	0.0529	1.4489	0.7840	0.1578	0.2879	0.5030	0.4446	1.7682	0.5699	0.3590
Magnolia	08/11/2016	Autumn	0.0761	2.5933	1.6969	0.4812	0.5166	0.6907	0.6826	2.4896	1.2643	0.3313
Average relative abundance			0.0005	0.0151	0.0082	0.0023	0.0034	0.0048	0.0045	0.0162	0.0069	0.0029

Table S2 – continued

Sample	Date	Season	BaF	BeP	BaP	Per	IcdF	DajA	DahA	IcdP	DacA	BbC
Cedar	25/01/2016	Winter	0.2331	6.8667	3.8585	0.4084	0.7213	0.1709	1.8746	0.1800	0.3062	0.2172
Cedar	25/01/2016	Winter	0.0727	2.5955	1.3212	0.1343	0.1840	0.0854	0.0809	1.1113	0.1238	0.0962
Cedar	25/01/2016	Winter	0.0396	2.1391	0.8379	0.0793	0.2387	0.0578	0.0542	1.5034	0.0878	0.0972
Cedar	11/02/2016	Winter	0.1103	3.5353	1.9771	0.2220	0.5390	0.1496	0.1526	3.4564	0.2232	0.1125
Cedar	11/02/2016	Winter	0.0657	2.1559	1.2645	0.1258	0.2074	0.1865	0.0663	1.3106	0.1378	0.1192
Cedar	11/02/2016	Winter	0.1100	3.9757	2.1070	0.2049	0.6385	0.1137	0.1080	3.1233	0.2457	0.1845
Cedar	11/02/2016	Winter	0.0433	2.1039	0.7930	0.1043	0.4642	0.1423	0.0936	2.8128	0.0783	0.0000
Average relative abundance			0.0002	0.0064	0.0033	0.0004	0.0008	0.0002	0.0007	0.0037	0.0003	0.0002
Cedar	08/04/2016	Spring	0.0000	1.4926	0.4481	0.0961	0.2404	0.0000	0.0000	1.1880	0.1379	0.0000
Cedar	08/04/2016	Spring	0.0571	1.6003	0.5973	0.1121	0.1595	0.0892	0.0815	1.3019	0.1499	0.4069
Cedar	08/04/2016	Spring	0.0000	1.0590	0.4611	0.1136	0.1314	0.0727	0.0813	1.1565	0.1425	0.0000
Cedar	26/04/2016	Spring	0.0491	0.9590	0.3256	0.0814	0.2721	0.1483	0.0000	1.9065	0.1672	0.2520
Cedar	26/04/2016	Spring	0.0340	0.7053	0.2236	0.0551	0.0757	0.0301	0.0322	0.4967	0.0341	0.1615
Cedar	26/04/2016	Spring	0.0813	2.9553	1.2683	0.1576	0.6052	0.1402	0.0879	2.7464	0.2365	0.0617
Average relative abundance			0.0002	0.0084	0.0032	0.0006	0.0014	0.0005	0.0003	0.0084	0.0008	0.0008
Cedar	23/06/2016	Summer	0.0000	0.4029	0.1679	0.0321	0.1074	0.0648	0.0449	0.3612	0.0310	0.0000
Cedar	23/06/2016	Summer	0.0219	0.9454	0.5953	0.4355	0.6298	1.3192	0.8058	2.0946	1.2024	0.3086
Cedar	23/06/2016	Summer	0.1545	0.0576	0.0259	0.0678	0.1246	0.0523	0.0361	0.2257	0.0487	0.0000
Cedar	23/06/2016	Summer	0.0218	0.8782	0.4820	0.0751	0.1628	0.1538	0.1850	0.8430	0.1378	0.0000
Cedar	11/07/2016	Summer	0.0222	0.5856	0.1523	0.0466	0.1466	0.0729	0.0426	0.4778	0.0406	0.0000
Cedar	11/07/2016	Summer	0.0096	0.4610	0.1951	0.0340	0.4102	0.0000	0.0000	0.3827	0.0000	0.0964
Cedar	11/07/2016	Summer	0.4078	0.1520	0.0511	0.1115	0.2750	0.1380	0.1754	0.8335	0.1251	0.0000
Cedar	11/07/2016	Summer	0.0051	0.3453	0.1748	0.0298	0.0975	0.0311	0.0362	0.3484	0.0281	0.0000
Average relative abundance			0.0023	0.0135	0.0065	0.0029	0.0069	0.0064	0.0047	0.0196	0.0057	0.0014
Cedar	27/10/2016	Autumn	0.0645	1.1247	0.7184	0.1098	0.3230	0.1132	0.0725	1.0007	0.0511	0.0309
Cedar	27/10/2016	Autumn	0.0298	0.8767	0.4779	0.0798	0.1869	0.0679	0.1208	0.7971	0.1059	0.0000
Cedar	27/10/2016	Autumn	0.0151	0.9690	0.6028	0.0874	0.0981	0.1788	0.1292	0.9424	0.1415	0.0347
Cedar	08/11/2016	Autumn	0.0386	2.9429	2.1870	0.4571	0.3498	0.3231	0.3042	2.9219	0.3319	0.1643
Cedar	08/11/2016	Autumn	0.0287	1.7750	0.8312	0.2832	0.6154	0.6531	0.5944	1.9151	0.6804	0.2116
Cedar	08/11/2016	Autumn	0.0270	1.2432	0.6189	0.1042	0.2019	0.1224	0.1307	0.9886	0.1447	0.0220
Average relative abundance			0.0004	0.0174	0.0106	0.0022	0.0035	0.0028	0.0026	0.0167	0.0028	0.0009

Table S2 – continued

Sample	Date	Season	Pyc	BghiPer	Att	DalP	DaeP	Cor	BbPer	DaiP	DahP
PM_{10}	25/01/2016	Winter	0.9465	6.2576	1.3191	0.9620	1.2389	7.3684	0.5021	0.4466	0.2073
PM_{10}	27/01/2016	Winter	0.3520	3.0047	0.2645	0.2468	0.3907	4.1981	0.0682	0.0468	0.0427
PM_{10}	30/01/2016	Winter	0.3943	2.3624	0.3160	0.5692	0.5499	3.5683	0.1376	0.0949	0.0715
PM_{10}	02/02/2016	Winter	0.3152	2.1112	0.2079	0.3622	0.3465	2.9328	0.0678	0.0568	0.0306
PM_{10}	05/02/2016	Winter	0.1364	1.2707	0.1336	0.1501	0.1740	1.5519	0.0501	0.0351	0.0180
PM_{10}	07/02/2016	Winter	0.1822	1.2346	0.1098	0.3525	0.1958	1.4080	0.0748	0.0535	0.0217
PM_{10}	11/02/2016	Winter	0.1396	1.2995	0.2229	0.2784	0.2195	1.7374	0.0477	0.0532	0.0257
Average relative abundance			0.0076	0.0540	0.0079	0.0090	0.0096	0.0700	0.0029	0.0024	0.0013
PM_{10}	08/04/2016	Spring	0.0215	0.1838	0.0181	0.0072	0.0079	0.1179	0.0000	0.0000	0.0000
PM_{10}	10/04/2016	Spring	0.0068	0.1067	0.0000	0.0067	0.0072	0.0572	0.0000	0.0000	0.0000
PM_{10}	13/04/2016	Spring	0.0095	0.0960	0.0000	0.0101	0.0089	0.0617	0.0011	0.0000	0.0000
PM_{10}	16/04/2016	Spring	0.0118	0.0929	0.0243	0.0000	0.0000	0.0407	0.0000	0.0000	0.0000
PM_{10}	18/04/2016	Spring	0.0072	0.1433	0.0000	0.0104	0.0059	0.0820	0.0000	0.0000	0.0000
PM_{10}	19/04/2016	Spring	0.0248	0.1844	0.0289	0.0076	0.0079	0.0968	0.0000	0.0000	0.0000
PM_{10}	21/04/2016	Spring	0.0245	0.1624	0.0122	0.0171	0.0198	0.1177	0.0000	0.0000	0.0000
PM_{10}	26/04/2016	Spring	0.0140	0.1517	0.0205	0.0151	0.0066	0.0628	0.0000	0.0000	0.0000
Average relative abundance			0.0034	0.0320	0.0030	0.0021	0.0018	0.0182	0.0000	0.0000	0.0000
PM_{10}	29/06/2016	Summer	0.0043	0.0454	0.0060	0.0045	0.0034	0.0278	0.0009	0.0000	0.0000
PM_{10}	01/07/2016	Summer	0.0038	0.0648	0.0117	0.0133	0.0051	0.0455	0.0000	0.0000	0.0000
PM_{10}	03/07/2016	Summer	0.0020	0.0616	0.0031	0.0053	0.0035	0.0389	0.0000	0.0000	0.0000
PM_{10}	04/07/2016	Summer	0.0046	0.1107	0.0068	0.0135	0.0099	0.0788	0.0000	0.0000	0.0000
PM_{10}	07/07/2016	Summer	0.0054	0.0516	0.0024	0.0093	0.0040	0.0356	0.0000	0.0000	0.0000
PM_{10}	09/07/2016	Summer	0.0910	0.4784	0.0453	0.0912	0.0647	0.3500	0.0074	0.0521	0.0000
PM_{10}	12/07/2016	Summer	0.0072	0.1969	0.0053	0.0130	0.0052	0.1137	0.0014	0.0000	0.0000
Average relative abundance			0.0098	0.0837	0.0067	0.0124	0.0079	0.0572	0.0008	0.0043	0.0000
PM_{10}	27/10/2016	Autumn	0.0554	0.5590	0.0506	0.0383	0.0324	0.3383	0.0081	0.0000	0.0000
PM_{10}	29/10/2016	Autumn	0.1311	1.4677	0.1430	0.1560	0.0941	0.7078	0.0104	0.0332	0.0000
PM_{10}	02/11/2016	Autumn	0.3364	3.7080	0.6775	0.3458	0.2075	1.5757	0.0305	0.1256	0.0529
PM_{10}	04/11/2016	Autumn	0.0652	0.6232	0.0513	0.0364	0.0280	0.2828	0.0036	0.0110	0.0000
PM_{10}	06/11/2016	Autumn	0.1305	1.5961	0.1851	0.1305	0.0635	0.7648	0.0097	0.0299	0.0000
PM_{10}	08/11/2016	Autumn	0.4268	4.8385	1.0830	1.5478	0.6786	4.6893	0.1201	0.3675	0.1509
Average relative abundance			0.0089	0.0991	0.0170	0.0175	0.0086	0.0647	0.0014	0.0044	0.0016

Table S2 – continued

Sample	Date	Season	Pyc	BghiPer	Att	DalP	DaeP	Cor	BbPer	DaiP	DahP
Magnolia	25/01/2016	Winter	0.2744	2.0058	0.1011	0.1251	0.1891	3.6302	0.0549	0.0000	0.0000
Magnolia	25/01/2016	Winter	0.1501	1.1051	0.0437	0.1262	0.1451	2.4191	0.0000	0.0000	0.0000
Magnolia	25/01/2016	Winter	0.5522	3.4694	0.1548	0.3925	0.4956	4.4264	0.1610	0.0000	0.0000
Magnolia	11/02/2016	Winter	0.2613	1.5243	0.0454	0.1707	0.1874	3.3391	0.0000	0.0000	0.0000
Magnolia	11/02/2016	Winter	0.1983	1.2156	0.0459	0.1794	0.1728	3.0488	0.0000	0.0000	0.0000
Magnolia	11/02/2016	Winter	0.3103	1.5210	0.0746	0.1658	0.2083	2.9454	0.0000	0.0000	0.0000
Average relative abundance			0.0008	0.0052	0.0002	0.0006	0.0007	0.0094	0.0001	0.0000	0.0000
Magnolia	08/04/2016	Spring	1.1396	14.6125	1.3539	1.0918	1.2457	24.4988	0.0000	0.0000	0.0000
Magnolia	08/04/2016	Spring	0.2746	3.8813	0.4492	0.2712	0.1691	4.0512	0.0000	0.0000	0.0000
Magnolia	08/04/2016	Spring	0.6862	11.9647	2.0496	0.8784	0.2469	12.7827	0.0000	0.0000	0.0000
Magnolia	26/04/2016	Spring	0.0624	0.8252	0.1094	0.0000	0.0000	0.7027	0.0000	0.0000	0.0000
Magnolia	26/04/2016	Spring	0.0821	1.5782	0.0000	0.0000	0.0000	1.4591	0.0000	0.0000	0.0000
Magnolia	26/04/2016	Spring	0.1941	1.6238	0.3595	0.2074	0.7338	3.6956	0.0000	0.0000	0.0000
Average relative abundance			0.0043	0.0610	0.0076	0.0043	0.0042	0.0835	0.0000	0.0000	0.0000
Magnolia	23/06/2016	Summer	0.0387	0.2254	0.0675	0.0602	0.0521	0.1731	0.0110	0.0000	0.0000
Magnolia	23/06/2016	Summer	0.0000	0.1020	0.0452	0.0000	0.0000	0.0591	0.0000	0.0000	0.0000
Magnolia	23/06/2016	Summer	0.2041	1.0264	0.1498	0.1924	0.1942	0.8020	0.0272	0.0000	0.0000
Magnolia	11/07/2016	Summer	0.0252	0.1439	0.0348	0.0790	0.0621	0.3872	0.0242	0.0000	0.0000
Magnolia	11/07/2016	Summer	0.0657	0.8063	0.0615	0.0583	0.0190	0.5205	0.0068	0.0000	0.0000
Magnolia	11/07/2016	Summer	0.0000	0.3545	0.0337	0.0396	0.0202	0.3353	0.0113	0.0000	0.0000
Average relative abundance			0.0032	0.0255	0.0038	0.0041	0.0033	0.0218	0.0008	0.0000	0.0000
Magnolia	27/10/2016	Autumn	0.4046	1.9460	0.3856	0.5049	0.6938	2.3620	0.1866	0.6029	0.8386
Magnolia	27/10/2016	Autumn	0.4499	1.4589	0.2831	0.3788	0.7180	1.3743	0.1483	0.4482	0.3344
Magnolia	27/10/2016	Autumn	0.5032	3.5092	0.6956	0.7843	0.8501	4.6203	0.3359	0.6017	0.5150
Magnolia	08/11/2016	Autumn	0.2898	2.1838	0.3730	0.9277	1.0282	2.5653	0.2630	0.6202	0.6171
Magnolia	08/11/2016	Autumn	0.4559	2.0693	0.3149	0.6375	0.6893	1.8172	0.1478	0.4571	0.3815
Magnolia	08/11/2016	Autumn	0.6316	3.5767	0.7079	0.5253	0.8476	4.6496	0.1951	0.5296	0.2680
Average relative abundance			0.0040	0.0216	0.0041	0.0055	0.0071	0.0255	0.0019	0.0048	0.0043

Table S2 – continued

Sample	Date	Season	Pyc	BghiPer	Att	DalP	DaeP	Cor	BbPer	DaiP	DahP
Cedar	25/01/2016	Winter	0.8728	4.9656	0.2002	0.3896	0.3910	7.7343	0.0811	0.0000	0.0000
Cedar	25/01/2016	Winter	0.3058	1.5906	0.0468	0.1787	0.2923	3.1512	0.0000	0.0000	0.0000
Cedar	25/01/2016	Winter	0.3344	1.6589	0.0386	0.1867	0.2158	3.3064	0.0000	0.0000	0.0000
Cedar	11/02/2016	Winter	0.4834	2.6602	0.0810	0.2638	0.2632	4.5027	0.0706	0.0743	0.0000
Cedar	11/02/2016	Winter	0.3998	1.5668	0.0526	0.2008	0.3089	2.9905	0.1070	0.1001	0.0000
Cedar	11/02/2016	Winter	0.7261	3.2172	0.0896	0.3296	0.5130	6.4318	0.0861	0.0614	0.1079
Cedar	11/02/2016	Winter	0.0409	1.6177	0.0964	0.1639	0.0754	1.5376	0.0000	0.0000	0.0000
Average relative abundance			0.0009	0.0047	0.0002	0.0005	0.0006	0.0081	0.0001	0.0001	0.0000
Cedar	08/04/2016	Spring	0.0000	6.4052	0.0000	0.0000	0.0000	5.2070	0.0000	0.0000	0.0000
Cedar	08/04/2016	Spring	0.9255	9.0423	1.3925	0.9316	0.8132	9.4263	0.0000	0.0000	0.0000
Cedar	08/04/2016	Spring	0.3403	6.3256	0.7517	0.0000	0.0000	5.5335	0.0000	0.0000	0.0000
Cedar	26/04/2016	Spring	0.5742	5.2781	0.9304	0.0000	0.0000	5.9142	0.0000	0.0000	0.0000
Cedar	26/04/2016	Spring	0.2962	2.6578	0.2726	0.0000	0.0000	2.9417	0.0000	0.0000	0.0000
Cedar	26/04/2016	Spring	0.2677	1.2259	0.1953	0.1209	0.1116	0.8559	0.0583	0.0274	0.0402
Average relative abundance			0.0023	0.0295	0.0034	0.0010	0.0009	0.0285	0.0001	0.0000	0.0000
Cedar	23/06/2016	Summer	0.0000	0.4334	0.0000	0.0330	0.0677	0.4254	0.0132	0.0528	0.0000
Cedar	23/06/2016	Summer	0.3512	1.2494	0.3025	0.0972	1.0512	1.4960	0.1288	0.1532	0.0000
Cedar	23/06/2016	Summer	0.0000	0.3035	0.0000	0.0513	0.0306	0.2881	0.0000	0.0000	0.0000
Cedar	23/06/2016	Summer	0.0887	0.9544	0.1750	0.0796	0.0596	1.1925	0.0000	0.0000	0.0000
Cedar	11/07/2016	Summer	0.0000	0.4639	0.0000	0.0551	0.0295	0.4198	0.0000	0.0000	0.0000
Cedar	11/07/2016	Summer	0.0000	0.3201	0.0000	0.0000	0.0000	0.1922	0.0000	0.0000	0.0000
Cedar	11/07/2016	Summer	0.0000	0.5695	0.0000	0.1431	0.0337	0.6497	0.0000	0.0000	0.0000
Cedar	11/07/2016	Summer	0.0000	0.3764	0.0777	0.0437	0.0409	0.4254	0.0000	0.0000	0.0000
Average relative abundance			0.0015	0.0164	0.0020	0.0018	0.0046	0.0179	0.0005	0.0007	0.0000
Cedar	27/10/2016	Autumn	0.0000	0.8050	0.0932	0.0000	0.0556	0.4279	0.0000	0.0000	0.0000
Cedar	27/10/2016	Autumn	0.0000	0.6185	0.0000	0.0908	0.0488	0.4536	0.0000	0.0000	0.0000
Cedar	27/10/2016	Autumn	0.0000	0.8642	0.0785	0.1086	0.0526	0.5478	0.0104	0.0000	0.0000
Cedar	08/11/2016	Autumn	0.1821	2.5468	0.0662	0.8842	0.3422	1.1431	0.0440	0.2056	0.0000
Cedar	08/11/2016	Autumn	0.2955	1.3582	0.0622	0.3480	0.3212	1.0260	0.0406	0.0000	0.0000
Cedar	08/11/2016	Autumn	0.0444	1.0248	0.1352	0.2156	0.1151	0.6654	0.0335	0.0000	0.0000
Average relative abundance			0.0010	0.0141	0.0008	0.0032	0.0018	0.0083	0.0003	0.0004	0.0000

Table S3 – Naphthalene concentrations on leaf samples expressed in ng g^{-1} of leaf mass and in ng cm^{-2} of leaf area.

Sample	Date	Season	Nap (ng g^{-1})	Nap (ng cm^{-2})
Magnolia	25/01/2016	Winter	123.4654	3.1278
Magnolia	25/01/2016	Winter	221.1775	7.5778
Magnolia	25/01/2016	Winter	162.2320	5.4282
Magnolia	11/02/2016	Winter	90.8575	2.3623
Magnolia	11/02/2016	Winter	138.0572	5.1682
Magnolia	11/02/2016	Winter	178.0902	6.2055
Magnolia	08/04/2016	Spring	16.1598	0.4751
Magnolia	08/04/2016	Spring	9.4480	0.4045
Magnolia	08/04/2016	Spring	20.7114	0.6237
Magnolia	26/04/2016	Spring	102.2410	3.0643
Magnolia	26/04/2016	Spring	29.3952	0.9796
Magnolia	26/04/2016	Spring	1.6993	0.5277
Magnolia	23/06/2016	Summer	3.9345	0.1358
Magnolia	23/06/2016	Summer	2.3311	0.0798
Magnolia	23/06/2016	Summer	10.4342	0.3031
Magnolia	11/07/2016	Summer	1.5256	0.1465
Magnolia	11/07/2016	Summer	11.1388	0.4160
Magnolia	11/07/2016	Summer	5.5870	0.1465
Magnolia	27/10/2016	Autumn	1.2026	0.0263
Magnolia	27/10/2016	Autumn	18.0637	0.5390
Magnolia	27/10/2016	Autumn	1.0342	0.0236
Magnolia	08/11/2016	Autumn	14.1771	0.3307
Magnolia	08/11/2016	Autumn	17.0486	0.4575
Magnolia	08/11/2016	Autumn	4.0387	0.1036
Cedar	25/01/2016	Winter	978.7795	23.9611
Cedar	25/01/2016	Winter	323.0645	12.9245
Cedar	25/01/2016	Winter	110.1725	4.1509
Cedar	11/02/2016	Winter	111.1155	4.1181
Cedar	11/02/2016	Winter	322.8828	13.9406
Cedar	11/02/2016	Winter	121.2313	3.4446
Cedar	11/02/2016	Winter	22.1775	1.4785
Cedar	08/04/2016	Spring	17.2921	0.5392
Cedar	08/04/2016	Spring	24.9984	0.7935
Cedar	08/04/2016	Spring	123.6241	3.5495
Cedar	26/04/2016	Spring	24.3572	0.8957
Cedar	26/04/2016	Spring	4.0688	0.1445
Cedar	26/04/2016	Spring	74.9021	2.8300
Cedar	23/06/2016	Summer	6.2811	0.2520
Cedar	23/06/2016	Summer	1.7435	0.0705
Cedar	23/06/2016	Summer	4.4693	0.1919
Cedar	23/06/2016	Summer	1.8731	0.0970
Cedar	11/07/2016	Summer	3.2741	0.1002
Cedar	11/07/2016	Summer	8.6912	0.2710
Cedar	11/07/2016	Summer	4.7962	0.1620
Cedar	11/07/2016	Summer	2.1656	0.1071
Cedar	27/10/2016	Autumn	10.8717	0.4362
Cedar	27/10/2016	Autumn	2.7971	0.1265
Cedar	27/10/2016	Autumn	3.0800	0.1501
Cedar	08/11/2016	Autumn	14.2975	0.5110
Cedar	08/11/2016	Autumn	13.0715	0.5247
Cedar	08/11/2016	Autumn	16.2825	0.5579

Figure S1 – Sampling location, land use and landscape surrounding the sampled plants.

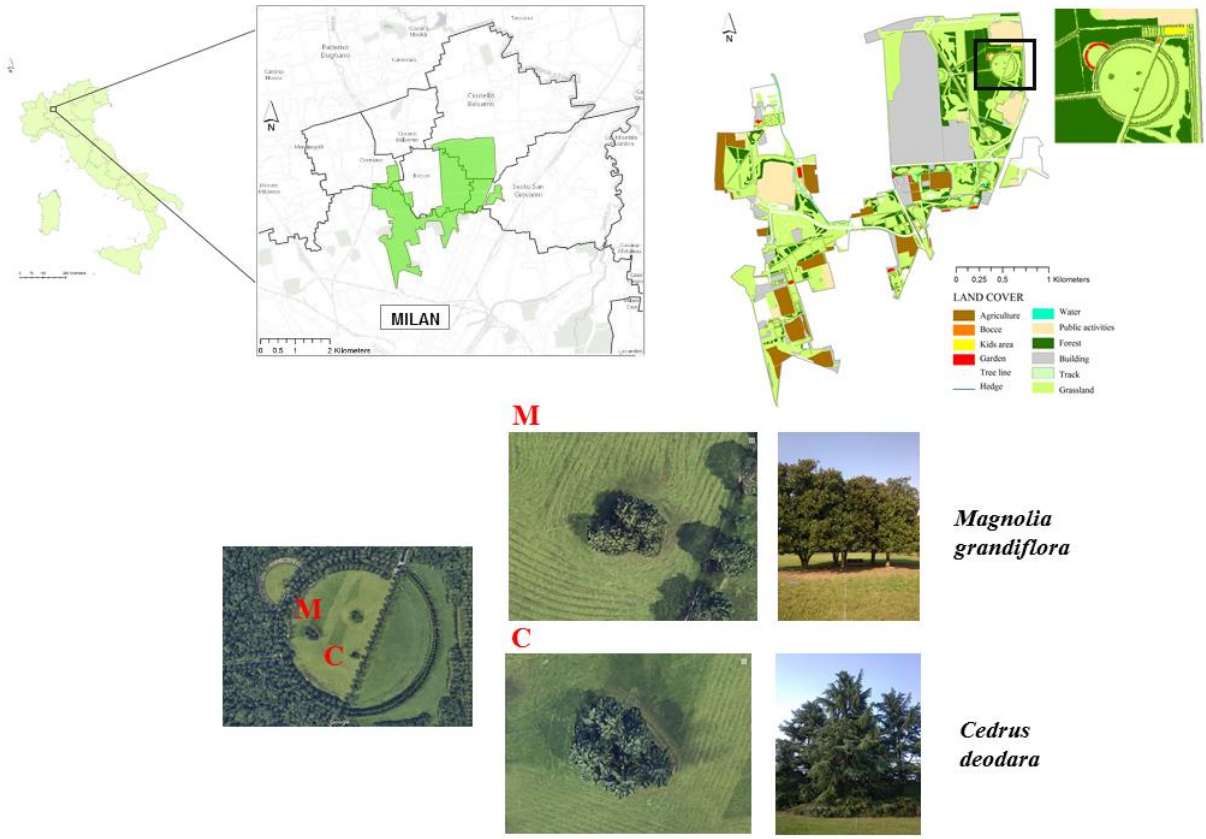

Figure S2 – PM_{10} and benzo(a)pyrene concentrations in the air, daily average temperature, rainfall, daily average radiation and relative humidity during 2016 recorded by the automatic station of the Regional Environmental Protection Agency (Arpa Lombardia) nearest to the locations where we collected leaves and PM_{10} (45°28'42.7"N, 9°13'54.0"E).

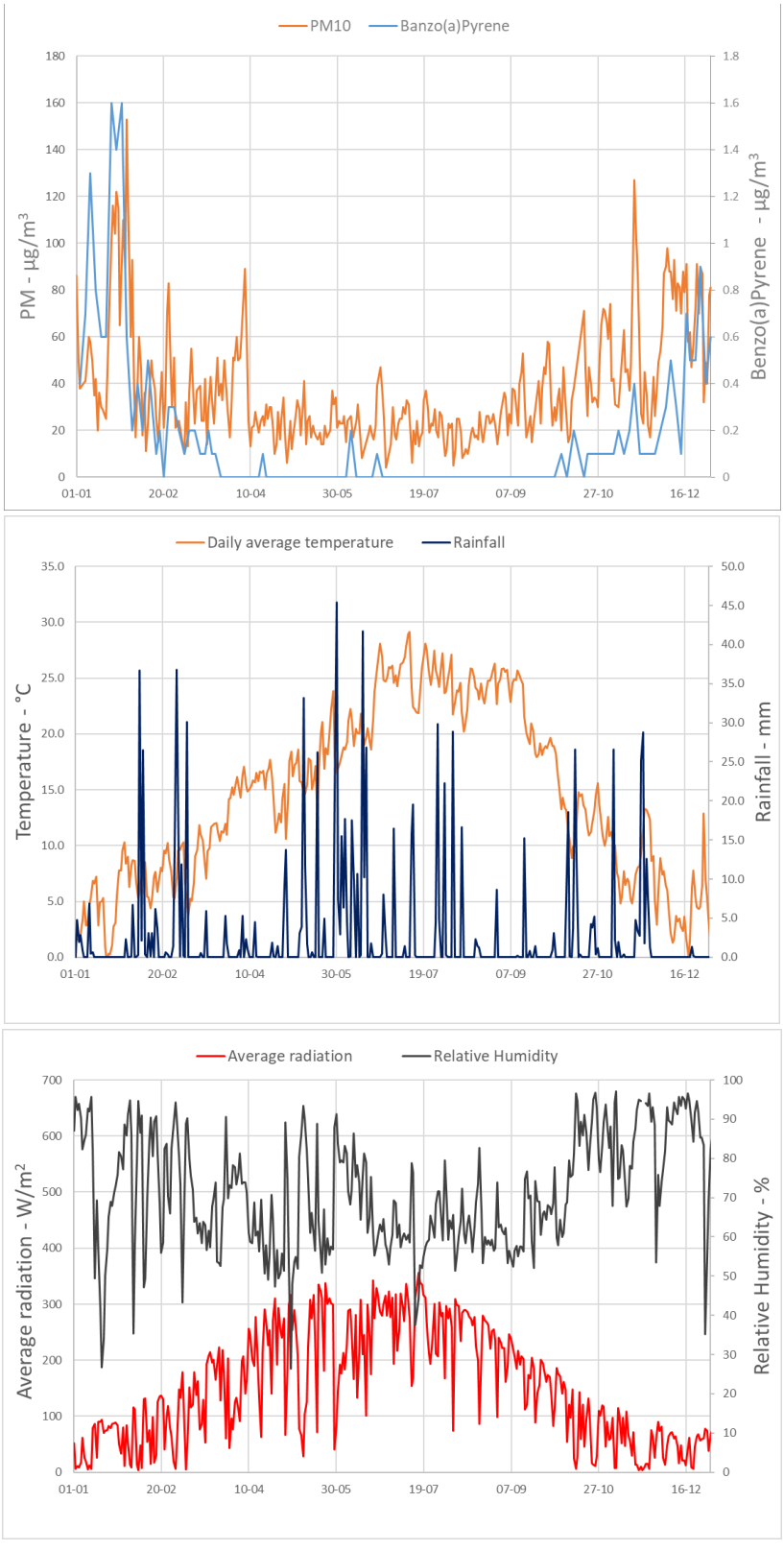

Figure S3 - Box-and-whisker plots reporting seasonal concentrations of light PAHs (2 and 3 rings) and heavy PAHs (more than 3 rings) in PM_{10} and in plant leaves. Data for PM_{10} are reported as mass/air volume concentrations (ng of PAHs / m^3 of sampled air) while in the case of plant leaves data are reported as mass/ mass units (ng of PAHs / g of leaf). The lower and upper hinges correspond to the first and third quartiles (the 25th and 75th percentiles). The upper and the lower whiskers extend from the hinge to the largest/smallest values no further than ±1.5 x IQR from the hinge.

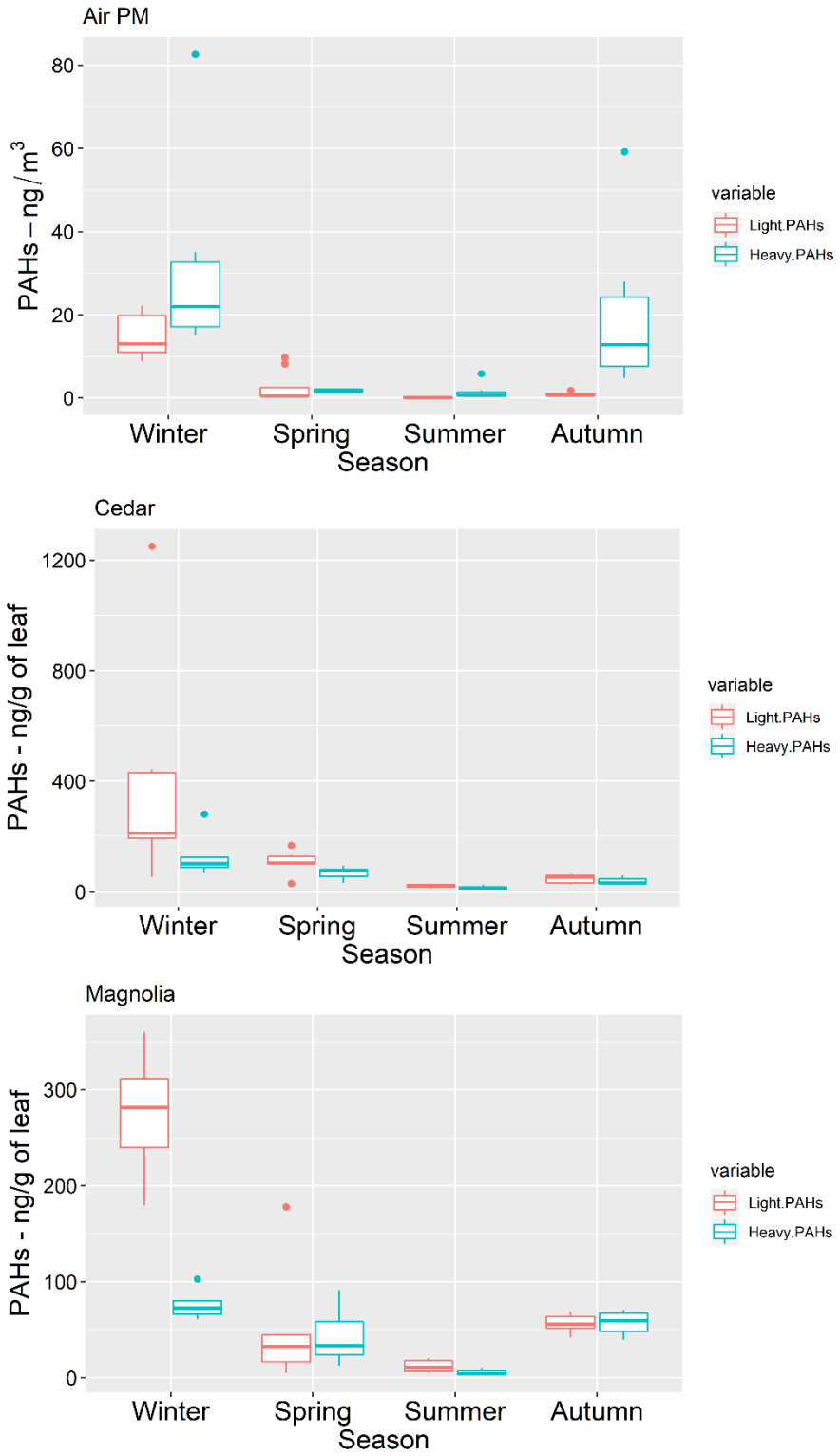

Figure S4 – Taxonomic classification of phyllosphere bacterial communities of magnolia and cedar at order level (A) and genus level (B). “Other Orders” contains orders that were less abundant than 1% in all groups of samples. “Other genera” contains genera that were less abundant than 2% in all groups of samples.