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Abstract

Holm oak (*Quercus ilex*), a widespread urban street tree in the Mediterranean region, is widely used as biomonitor of persistent atmospheric pollutants, especially particulate-bound metals. By using lab- and field-based experimental approaches, we compared the leaf-level capacity for particles' capture and retention between *Q. ilex* and other common Mediterranean urban trees: *Quercus cerris*, *Platanus ×hispanica*, *Tilia cordata* and *Olea europaea.* All applied methods were effective in quantifying particulate capture and retention, although not univocal in ranking species performances. Distinctive morphological features of leaves led to differences in species' ability to trap and retain particles of different size classes and to accumulate metals after exposure to traffic in an urban street. Overall, *P. ×hispanica* and *T. cordata* showed the largest capture potential per unit leaf area for most model particles (Na^+ and powder particles), and street-level Cu and Pb, while *Q. ilex* acted intermediatelly. After wash-off experiments, *P. ×hispanica* leaves had the greatest retention capacity among the tested species and *O. europaea* the lowest. We concluded that the *Platanus* planting could be considered in Mediterranean urban environments due to its efficiency in accumulating and retaining airborne particulates; however, with atmospheric pollution being typically higher in winter, the evergreen *Q. ilex* represents a better year-round choice to mitigate the impact of airborne particulate pollutants.

Keywords: airborne particles, metals, leaf capture, *Quercus cerris*, *Quercus ilex*, *Platanus ×hispanica*, *Tilia cordata*, *Olea europaea*

Highlights

- London plane and lime tree leaves captured most Na^+ aerosol and powder particles per unit leaf area.
- London plane leaves showed the largest metals' (Pb, Zn, Cu) capture potential, near an urban street.
- London plane leaves also showed greatest capacity for particle retention after wash off.
- In a year-round scenario, Holm oak likely has the highest potential for PM removal due to its evergreen nature.

1. INTRODUCTION

Urban population is increasing worldwide and a further rise in urbanisation is predicted (Buhaug and Urdal, 2013). One of the main implications of urbanization is air pollution which is associated with several health outcomes for urban residents, including respiratory and cardiovascular illness, neurological disorders and cancers (e.g. Pope and Dockery, 2006; HEI, 2010). In many urban environments the airborne particulate matter (PM) affects more people than any other atmospheric pollutant and no threshold PM concentration has been identified below which no damage to health is observed (WHO, 2014). It has been estimated that PM causes 3.7 million premature deaths annually worldwide and more than 450,000 in Europe (WHO, 2014). Particulate matter from natural (sea salt, soil dust, volcanic ash, forest fires, pollen) or anthropogenic sources (fuel combustion in thermal power generation, traffic, incineration and domestic heating for households) is directly emitted to the atmosphere (primary) or is formed in air as secondary inorganic or organic aerosols from precursor gases such as SO_2, NO_x, NH_3, and volatile organic compounds. Therefore, the urban PM is a complex mixture of different phases, with different chemical composition and size. Particles with an aerodynamic diameter < 10 µm (PM_{10}) can enter the human airways, particles <2.5 µm ($PM_{2.5}$) can reach pulmonary air sacs (Baeza-Squiban et al., 1999) and those <0.1 µm enter the blood circulation system (EEA, 2014).

In cities, the traffic and especially diesel-fuelled vehicles are an important source -close to the ground - of PM-bearing metals and particulate-bound polyaromatic hydrocarbons which have been linked with adverse health effects (e.g. HEI, 2010). Non-exhaust emissions (tyre, brake and road surface wear, corrosion and dust re-suspension) from road traffic are about 50 % of exhaust emissions of primary PM_{10} and about 22% of the exhaust emissions of primary $PM_{2.5}$ (Hak et al., 2009). Therefore, even with zero tailpipe emissions, the traffic will continue to be a very important source of PM in urban environments (Kumar et al., 2013).

Particles can be removed from the atmosphere by various deposition mechanisms (NEGTAP, 2001), with dry deposition being the main pathway, especially in areas with scarce atmospheric precipitation such as the Mediterranean region. Vegetation has a pivotal role in the removal of the atmospheric particulate in terrestrial ecosystems. Dry deposition processes and the particle interception by trees are affected by many factors such the canopy characteristics, wind speed, temperature, particle size, gas solubility as well as leaf pubescence, size and morphology (Beckett et al., 2000; Freer-Smith et al., 2005; Hofman et al., 2014; Weber et al., 2014). Most particles adsorbed on leaves and other plant surfaces are often re-suspended to the atmosphere, washed off by rain, or dropped to the ground with leaf and twig fall. Although it is well-known that the temporary retention of particles by urban trees can reduce atmospheric PM concentrations (e.g. Beckett et al., 2000, Fowler et al., 2004; Novak et al., 2006) the effectiveness of street trees or vertical gardens as a long-term alternative to other measures such as the wet cleaning of streets is still debated (Litschke and Kuttler, 2008). Some previous quantitative estimates of PM_{10} reduction by urban vegetation on the city-scale suggested a small effect (often < 1%; e.g. Novak et al., 2006; Escobedo and Nowak, 2009; Tallis et al., 2011). However, as discussed by Litschke and Kuttler (2008), these estimates assumed a particle deposition velocity (i.e. the quotient of the particles' flow rate towards the leaf surface and the atmospheric particle concentration) of about 1 cm s^{-1}, whereas *in-situ* measurements indicate considerably higher values and literature data for PM_{10} deposition velocities to vegetation vary from ~ 0.01 to ~10 cm s^{-1}. This variability is due to particle characteristics, meteorological conditions as well as to tree species differences in canopy architecture, leaf morphology and surface properties (Pugh et al., 2012; Maher et al., 2013).

Modelling, as well as a number of experimental field and laboratory approaches, have been used to evaluate the PM interception by leaves from a number of plant species (e.g. Beckett et al., 2000; Sæbø et al., 2012; Räsänen et al., 2013). It is known that leaf morphology and wettability play an important role in the interception of airborne particles and in their re-suspension to the atmosphere (e.g. McPherson et al., 1994). However, limited information is available about the wash-off by rain of adsorbed particles from leaves of different tree species (Neinhuis and Barthlott, 1998).

In order to contribute to the selection and maintenance of tree species with a higher deposition velocity for an efficient PM interception in Italian cities we compared the particle capture and retention capacity by leaves from a popular and prevalent tree species in Italian urban and roadside environments - *Quercus ilex* L., to that of possible alternatives: *Quercus cerris* L., *Platanus ×hispanica* Münch., *Tilia cordata* Mill., and *Olea europaea* L. In Mediterranean regions, the evergreen holm oak (*Q. ilex*) has a wide natural distribution and in Italy it has been used since the sixteenth century in the landscaping of urban and rural parks and gardens. Holm oak has a large canopy, as wells as Leaf Area Index (LAI) typically higher than that of other broad-leaf species (Sgrigna et al., 2015); its leaves have a hair cover and thick waxy cuticles. Because of these leaf properties, which enhance the scavenging and retention of airborne particles and the incorporation of lipophilic organic contaminants, holm oak leaves were widely used for biomonitoring persistent pollutants in many Italian urban areas (e.g. Monaci et al., 2000; Gratani et al., 2008; Fantozzi et al., 2013; Ugolini et al., 2014). Through a quantitative analysis of PM fractions on *Q. ilex* leaves collected (three times in a year) in an urban environment, Sgrigna et al. (2015) found a mean surface PM deposition of 20.6 µg cm^{-2}, a value in the same range of that reported for other urban tree species by Dzierżanowski et al. (2011). Having in mind the need to diversify planting in order to increase the resilience of urban trees and decrease susceptibility to pests and diseases (Laćan and McBride, 2008), in our study the leaf particle interception and retention by *Q. ilex* were compared with those of other urban tree species to identify possible alternative/complementary trees as PM mitigating tools in Mediterranean urban environments. To evaluate if a cheap and accessible method can produce reliable estimates of tree leaf potential for PM interception, NaCl aerosol and talcum powder were blown onto the leaves in a simple wind tunnel. The results of these laboratory experiments were compared with those from metal particle accumulation in leaves exposed to traffic in an urban street. We chose three metals (Pb, Cu and Zn) routinely associated with anthropogenic pollution sources (Espinosa et al., 2002, Wang 2006) as indicators of street-level pollution; their concentrations are reported in numerous studies (e.g. Davis et al., 2001, Lindgren, 1996), so this should enable baseline comparisons. The leaf particle *retention* capability in the five tree species was also evaluated by simulating a rainfall. Thus, this work attempted to evaluate the agreement among different laboratory experiments and to compare the behavior of leaves from five selected tree species in terms of particle capture and retention, in the laboratory and the field.

2. MATERIALS AND METHODS

2.1. Plant material

The main leaf characteristics of the five tree species common in Mediterranean urban areas are summarized in Table 1. *Platanus ×hispanica* (London plane) has relatively large, stiff leaves coated with fine, firm hairs (during springtime); those of *T. cordata* (lime tree) are also large but mostly hairless, except for small tufts of hair in the leaf vein axils (Hölscher, 2003). Both *Q. ilex* (holm oak) and *Q. cerris* (Turkey oak) leaves have a water-repellent surface mainly due to the thick epicuticular waxy layer. *Quercus ilex* is also characterized by stellate trichomes on the surface (Quero et al., 2006). *Olea europea* (olive) has small silvery-green leaves with glossy and veined upper surface (Marchi et al., 2008).

In all experiments, young fully-expanded leaves of the current year's growth were used. Wind-tunnel and laboratory experiments were carried out in Summer 2012 at the University of Reading (UK) (see section 2.2) and leaves were collected from the 3-year-old trees maintained in ventilated glasshouses (*O. europaea*, *P. ×hispanica* and *Q. ilex*), or from nearby field-grown mature trees (*Q. cerris* and *T. cordata*); leaves from 2-year-old sections of the branches were used in all experiments. During the Summer 2013, short branches (from 2-year-old wood), of all tree species were excised from mature trees from the Siena Botanical Garden and were exposed to traffic in an urban street (see section 2.3).

2.2 Laboratory wind tunnel experiments

2.2.1 Method development

The wind tunnel used in the experiments to distribute the particles to the leaves was an open-circuit type (Figure 1), 50 cm long and 15 cm in diameter. Particles were generated from a 0.1 M NaCl solution with a pressure sprayer (nozzle outlet diameter = 1 mm) or by a powder-dispenser sieve containing fine powder (Johnson's powder, Johnson & Johnson, New Jersey, USA). Particles were dispensed in front of a splash-proof DC fan (IP54 Ebm-papst, Bachmühle, Germany) at the entry point to the wind tunnel. Droplet diameters and powder particle size were in the range from 0.05 μm to 15 μm.

Preliminary experiments were performed with Petri plates, glass slides and artificial leaves held by a custom made rigid mesh support, to establish optimal experimental conditions (i.e. length of application time, amount of NaCl solution and powder, and the distance between leaves and the fan). Artificial leaves were constructed to mimic the average shape and size of the five different tree species, tracing on paper three real leaves with a shape/size representing the average for every species and then laminating them. Variations in weight of Petri plates, slides and artificial leaves (before and after particle application; 10 replicates for each treatment) were determined with a precision balance. Preliminary tests using 30 ml NaCl solution or 5 g of powder, at a distance of 20 cm, with an exposure time of 5 s, and wind/air speed in the tunnel of 6.75 m s^{-1} gave the most reproducible results.

For the experiments, fresh leaves of the five tree species were then inserted into a mesh support (Figure 2) before exposing them to various treatments.

2.2.2. Capture and retention of NaCl aerosol

The fresh weight and the leaf area (LA) of 40 leaves of each tree species were measured before mounting leaves in a support and placing them into the wind tunnel for the exposure to NaCl aerosol. Additional three leaves per species (sprayed only with distilled water) represented controls. After aerosol exposure, all leaves were carefully laid out to air dry under a laminar extractor fan and then 20 leaves were oven-dried for 24 h at 70 ^{o}C. Dry leaf samples were pooled in groups of 2-3 leaves to produce 6-9 replicates per species; leaves were manually ground and homogenized using a mortar and pestle. About 500 mg of each sample were digested with concentrated HNO_3 at 120 °C for 8 h in a microwave pressurized digestion system. The mineralized samples of exposed and control leaves were analyzed with an atomic absorption spectrophotometer (AAS) and Na concentrations (expressed in µg g^{-1} d.w. basis) were determined by the method of standard additions. Procedural blanks were below the Na detection limit; the accuracy of digestion and analytical procedures was checked by routine determination of Na concentrations in standard reference materials (SRM No 2711a and 1515) from the National Institute of Standards and Technology (Gaithersburg, USA). The analytical recoveries from the certified values ranged from 86 to 97%.

The other 20 treated leaves and control leaves were inserted again into the wind tunnel and exposed to distilled water aerosol for 5 s at a distance of 20 cm from the fan; leaves were positioned perpendicularly to the air flow. Wash-off solution was collected in a Petri plate and analyzed for Na^+ concentrations with the AAS.

The leaf NaCl aerosol capture capacity was estimated by analysing leaf Na^+ concentrations in two ways. One was by simply subtracting leaf Na^+ concentrations before and after the experiment ('N'). This was done to assess the *total* Na^+ captured by each leaf, not taking into the account differences in leaf size.

Other was by accounting for the leaf weight and leaf area so that a Na^+ capture potential (Cp) *'per unit'* of leaf weight and leaf area (LA) of different species can be compared. To do this the following equation was used:

$$Cp = N \text{ x } (\text{leaf weight}/LA)$$

where 'N' was the difference in leaf Na^+ concentration before and after the experiment (expressed in mg g^{-1})~~ and 'D' was the leaf 'density' (obtained as a ratio of leaf weight and leaf area)~~.

The Na^+ wash-off (R) was calculated using the equation:

$$R= N_r/LA$$

where 'N_r' was the Na^+ concentration in the runoff (mg l^{-1}) and LA was leaf area from which runoff was collected.

2.2.3. Capture and retention of powder particulate

A further 40 leaves of each species, whose leaf area and fresh weight were previously determined, were exposed to 5 g of powder at 20 cm from the fan. Leaves were then carefully laid down in order to avoid loss of powder; a piece of leaf tissue (1 cm^2) was cut from the centre of the lamina of 20 leaves and fixed onto a microscope slide. Powder particle retention

of the adaxial leaf surface (facing the fan) was determined by counting the number of particles using a digital image-analysis system connected to a light microscope. For each tree species, the number of particles on 20 treated leaves was counted in four random squares of 1 μm^2 area per leaf and results were reported as particle number mm^{-2}. Two untreated leaves acting as controls were also analyzed using the same procedure.
The other 20 treated leaves were inserted again into the wind tunnel, exposed to distilled water aerosol as detailed above and then analyzed by light microscope to assess the powder retention after wash-off.

2.3 Field experiment

Three branches with similar length and leaf age were excised from each of the *Q. ilex*, *Q. cerris*, *P. ×hispanica*, *T. cordata* and *O. europaea* trees in the Botanical Garden of Siena (a green park with no adjacent traffic or other sources of airborne metals), and carefully washed with distilled water. Three subsamples of leaves from each tree species were analyzed for Cu, Zn and Pb concentrations to assess the metal concentrations before the exposure (for details on samples preparation, chemical digestion and analytical determination see Fantozzi et al., 2013). On 2 July 2013 the branches, inserted in 10 ml plastic flasks with water, which was changed every two days, were randomly placed on the 10 m long stretch of a wall (2 m above the ground and 1m away from a street in Siena city centre, with 200-1500 vehicles h^{-1} (ARPAT, 2011) (Figure 3). During the 21 days exposure there was no atmospheric precipitation. All exposed leaves on each branch were pooled and the 3 composite samples for each tree species were analyzed for total Cu, Zn and Pb concentrations.

2.4 Data analysis

Data were analyzed using GenStat (11th Edition, Lawes Agricultural Trust, Rothamsted Experimental Station, UK). Analysis of variance (ANOVA) was used to assess the effects of the plant species on measured parameters; variance levels were checked for homogeneity and values were presented as means with associated least significant differences (LSD, $P = 0.05$) or standard error (SE).

3. RESULTS

3.1 Laboratory wind-tunnel experiments

3.1.1 Capture and retention of NaCl aerosol

Table 2. shows NaCl aerosol capture ~~potential (Cp)~~ by the different tree leaves. Larger leaves like *P. ×hispanica* and *T. cordata* captured more Na^+ than smaller leaves (e.g. 1.97 mg g^{-1} *vs* 0.36 mg g^{-1} for Tilia compared to *Olea*, respectively); *Tilia* was additionally most efficient in Na^+ capture per unit leaf area (0.015 mg cm^{-2}), followed by *Q. cerris* and *P. ×hispanica* (0.009 and 0.008 mg cm^{-2}). *P. ×hispanica* and *T. cordata* leaves also showed significantly less ($p < 0.01$) Na^+ wash-off (Table 2). The Na^+ wash-off was most pronounced in *O. europaea* and intermediate in *Q. ilex* and *Q. cerris* (1.87, 0.55 and 0.53, respectively, Table 2).

3.1.2 Capture and retention of powder particulate

Table 3 summarizes the results of the powdering experiment with talcum and the following wash-off treatment. *Tilia cordata* and *O. europaea* leaves captured the greatest number of powder particles; all species, except *Q. cerris*, captured mostly particles in the 5-10 µm range (Table 3). Leaves from the two oak species showed a lower capture efficiency for the smaller particles (< 5 µm) and a significantly higher ($p < 0.01$) capture efficiency for coarser particles (> 10 µm) than the other three species. The wash-off treatment removed less than 10% of the total number of particles adsorbed on *P. ×hispanica* leaves and about 31, 48, and 64 % of those adsorbed on *Q. ilex*, *T. cordata*, and *Q. cerris*, respectively. However, under the adopted experimental conditions, about 60% of the finest particles (< 5 µm) were retained by *Q. ilex* leaves and about 42 % in those of London plane, lime tree and Turkey oak. The olive leaves showed a minimal capacity to retain adsorbed particles (only <13%) (Table 3).

3.2 Field experiment

Average concentrations (µg/g) of Cu, Pb and Zn in the leaves exposed for 21 days to the street-level polution in Siena varied between the plant species, and between the metals (Table 4). Lead (Pb) concentrations were no higher than 0.40 µg g^{-1}, but Cu and Zn up to 13-16 µg g^{-1} after 3 weeks of exposure to streel-level traffic in dry summer weather. In terms of leaf-level capture, for Pb for example, concentration increase after exposure ranged from 5.8 % in *O. europea* to 27.9 % in *P. ×hispanica*. For other metals, this range of increase in metal concentration between different species was smaller: e.g. for Cu it was between 12.9% (*O. europea*) to 26.6% (*P. ×hispanica*) and even smaller for Zn (9.20% in *Q. ilex* to15.4% for *P. ×hispanica*). *P. ×hispanica* showed a greatest increase in concentrations of metals after the exposure compared to other species. *Tilia* and *Q. ilex* were comparable in terms of concentration increase for Zn: 9 – 10 % for leaf Cu concentration increase after street-level exposure (Table 4).

4. DISCUSSION

Previous studies indicate that PM interception by trees is often (although not exclusively, see Hofman et al., 2014) affected by canopy architecture; thus conifers, in spite of the low unit needle-leaf area, usually show the highest capture efficiency (e.g. Beckett et al., 2000; Freer-Smith et al., 2004; Hwang et al., 2011; Räsänen et al., 2013). All the species considered in this study had leaves capable to distinctively collect airborne particulate; however, the spatial structure of branches and twigs of different species and the lack of foliage during the winter in chosen deciduous species would decrease their capacity for PM trapping on a year-round basis. The administration of NaCl aerosol, with an approach previously used by Beckett et al. (2000) and Räsänen et al. (2013), suggested a much higher capture potential (Cp) and a much lower Na^+ wash-off in *Tilia* and *P. ×hispanica* leaves than in the other species (Table 2). The leaf wettability affects the capture of aerosols (Freer-Smith et al., 2004) and some features of *Q. ilex*, *Q. cerris* and *O. europaea* leaves such as their sclerophylly, superficial roughness, presence of trichomes, convex epidermal cells and wax crystals can reduce the contact area between water and the leaf surface (Kardel et al., 2012) and consequently, the adsorption the Na^+ aerosol.

The *Tilia* and *P. ×hispanica* leaves, together with those of *O. europaea*, captured the highest number of talcum particles, especially those <10 µm, whereas those with a diameter >10 µm were mainly accumulated by oak leaves. After the wash-off treatment *P. ×hispanica* retained almost 90% of total particles, while *O. europaea* retained just 13% (dropping to only 5% in the <5 µm particle size). Small circumference-to-area ratio in olive might be a reason for the low capacity for particles retention (Freer-Smith et al., 2005). Holm oak leaves retained about 68% of total adsorbed particles, including those <5 µm. In agreement with the results of earlier studies (Freer-Smith et al., 1997; Lindberg and Lovett, 1992) indicating that the median diameter of particles collected by oak tree species would be around 9 µm, in our experiment the leaves of *Q. cerris* and *Q. ilex* also retained fewer particles in <5 µm range than the other tree species. Carpenter et al. (2005) reported that *Platanus* and *Tilia* leaves can collect a very variable range of particle sizes, and in agreement with another study (Jouraeva *et al.*, 2002) our results indicate that *Tilia* leaves are particularly efficient in the capture of <10 µm particles.

The tree species we studied ranked in the order: *P. ×hispanica* > *T. cordata* > *Q. ilex* > *Q. cerris* > *O. europaea* for the powder retention and in the order *T. cordata* > *P. ×hispanica* = *Q. cerris* > *Q. ilex* = *O. europaea* for the NaCl aerosol capture potential. Differences in ranking are likely due to leaf size differences between species (which affect capture even when size differences are accounted for, ~~at a ‘unit’ level – at which we expressed our capture capacities -~~ due to a change in turbulences, Beckett et al., 2000) and features including a smooth or wrinkled surface, the presence of micro-roughness, hairs, veins or trichomes (e.g. Beckett et al. 2000; Liu et al., 2012; Speak et al., 2012) and how they would affect the interception of (Na^+) aerosols vs powder. Thoennessen (2002) for instance, investigated the distribution of pollutants on leaves along a street with high traffic volume and distinguished between plants with very rough surfaces and higher pollutant deposition and those (self-cleaners) with smooth surfaces which reduce the particles deposition and favour their removal by precipitation and wind. Among leaf types in this study the relatively smaller leaf size, the sclerophylly (i.e. the reduced wettability) and the smoother surface are probably the main factors affecting the much lower retention of Na^+ and powder particles on olive and oaks.

In all tree species exposed to high-medium traffic intensities over a 3-week period at the street level, there was a statistically significant increase of average leaf Cu, Zn and Pb concentrations (Table 4). While there was no atmospheric precipitation and consequently wash-off of adsorbed particles during the exposure period, the results corroborated the lower capability of *O. europaea* and *Q. cerris* leaves to adsorb airborne particles. In our laboratory experiments *Q. cerris* leaves accumulated the minimum number of total particles; in the field it generally showed lowest particles concentration increase. Both our talcum powder experiment and other studies (Freer-Smith et al., 1997; Tomašević et al. 2008) showed that some oak species mainly capture larger particles.

Differences in particulate trapping efficiency have been widely studied in a number of tree species (e.g. Beckett et al. 2000; Freer-Smith et al., 2005; Dzierzanowski et al., 2011). Additional species considered in this study, showed a higher Na^+, talcum powder, Pb and Cu capture efficiency in London plane and lime tree leaves. However, after the powdering the *Q. ilex* leaves showed a capacity to retain a larger proportion of fine adsorbed particles, compared with London plane and lime. Additionally, holm oak has a significant practical advantage in being an evergreen species with high Leaf Area Index (i.e. a foliar density which should enhance air turbulence around leaves and the PM deposition; Sgrigna et al. 2015). Also, holm oak is well adapted to growing and functioning under the conditions of water deficit, regularly experienced in the Mediterranean region (Bussotti et al., 2002), which seems to give advantage to this species over the others as an interceptor of airborne particles in urban areas.

CONCLUSIONS

Comparisons of the leaf-level capture of aerosol and particles among five species of broadleaf trees which are common in many Mediterranean urban environments showed that *P. ×hispanica* and *T. cordata* leaves intercepted and retained NaCl aerosol and talcum particles more efficiently than *O. europea*, *Q. cerris*, and *Q. ilex* leaves. In agreement with the results of previous surveys with other tree species, the leaf behaviour seems a species-specific process depending above all on leaf surface morphology and wettability. In general, *Q. cerris* and especially *O. europea* leaves showed the weakest performances, while after the wash-off, *Q. ilex* leaves retained high proportion of fine intercepted particles. Thus, although London plane and lime tree leaves generally fared the best and these species should be considered to decrease the impact of airborne particles in urban environments, due to its evergreen nature, foliage distribution and density which is maintained in all seasons, the holm oak probably, has a greater potential for a year-round air pollutant sequestration in Mediterranean urban environments.

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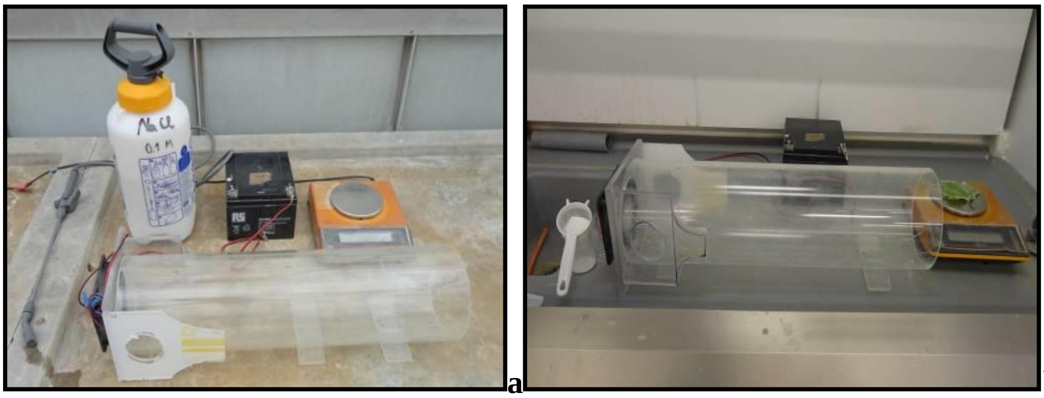
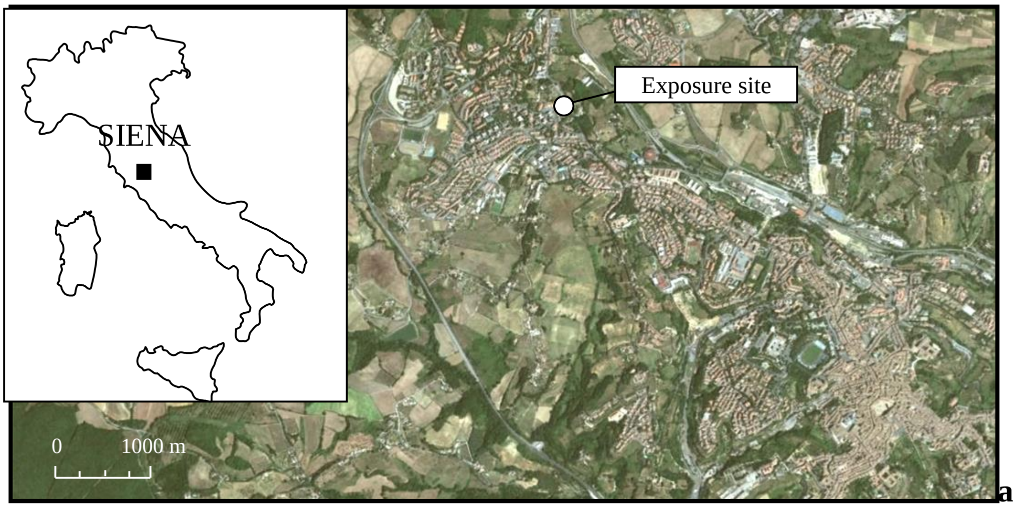


a b

Figure 1: Wind-water tunnel (open-circuit type) used in the experiments: **a.** with the spray dispenser, **b.** with the talcum powder.

c

Figure 2: Leaves of the five tree species inserted in an iron support, from left to right: *T. cordata*, *P. ×hispanica*, *Q. cerris*, *Q. ilex*, *O. europaea*.



a

Figure 3: Location of the urban street where three branches with five leaves each, for each studied species, were exposed on a wall (1m away from the street, 2 m above the ground).

Table 1: Tree species used in the experiment and their leaf properties.

Tree species	Leaf properties			
	fall/retention	Hairs	waxes	size (cm^2)*
Platanus ×hispanica (London plane)	Deciduous	Yes	Scarce	77.1±4.8
Tilia cordata (Lime tree)	Deciduous	Sparse	Scarce	42.5±4.3
Quercus ilex (Holm oak)	Evergreen	Sparse	Pronounced	14.3±0.9
Quercus cerris (Turkey oak)	Deciduous	No	Pronounced	24.1±2.1
Olea europaea (Olive tree)	Evergreen	Sparse	Pronounced	8.0±0.3

*Average leaf size of experimental leaves

Table 2. Leaf Na^+ concentrations after aerosol application (expressed in mg g^{-1} and mg cm^{-2}) and Na^+ concentrations in the runoff (mg l^{-1} cm^{-2}) after rinsing with r.o. water. Data are mean of 6-9 replicates per plant species, presented with associated LSD and d.f. Different letters next to the means in each column indicate that means are significantly different.

Tree species	Leaf Na concentration (mg g^{-1})	Leaf Na concentration 'Cp' (mg cm^{-2})	Runoff water Na concentration (mg l^{-1} cm^{-2})
P. ×hispanica	1.34±0.04 b	0.008±0.0003 b	0.30±0.02 b
T. cordata	1.97±0.14 a	0.015±0.0013 a	0.20±0.02 a
Q. cerris	0.55±0.03 c	0.009±0.0007 b	0.53±0.01 c
Q. ilex	0.53±0.07 c	0.003±0.0004 c	0.55±0.03 c
O. europaea	0.36±0.01 c	0.002±0.0002 c	1.87±0.09 d
LSD (d.f. = 35)	0.2511	0.0024	0.098

Table 3: Powder particle retention (mean number of particles mm^{-2} ± SEM as well as the associated LSD for each particle class size) and particle size (class: < 5, 5-10 and >10 µm) in tree leaves (n= 20) after powdering and wash-off steps for the five studied species. Different letters next to the means in each column indicate that means are significantly different.

	after powdering (num/mm^2)				**after wash-off (num/mm^2)**				
	<5 µm	5-10 µm	> 10 µm	tot	<5 µm	5-10 µm	> 10 µm	tot	**% change**
P.hispanica	2688 ± 65 b	3779 ± 80 b	1673 ± 49 c	8140 ± 65 b	1121 ± 46 c	3613 ± 66 a	2757 ± 96 a	7491 ± 69 a	93.6±2.0 a
T.cordata	3007 ± 556 a	5741 ± 600 a	787 ± 242 e	9535 ± 466 a	378 ± 189 b	3461 ± 648 b	1052 ± 245 c	4891 ± 361 b	52.2±2.3 c
Q.cerris	378 ± 172 d	2014 ± 217 c	2537 ± 273 a	4928 ± 220 d	159 ± 141 a	742 ± 153 d	856 ± 111 d	1757 ± 135 c	35.8±1.0 d
Q.ilex	795 ± 234 c	3891 ± 284 b	2243 ± 262 b	6929 ± 260 c	482 ± 198 b	2584 ± 223 c	1675 ± 176 b	4741 ± 199 b	68.6±1.1 b
O.europaea	3007 ± 471 a	5483 ± 595 a	1287 ± 309 d	9777 ± 458 a	151 ± 72 a	507 ± 103 e	575 ± 105 e	1233 ± 93 d	12.4±0.64 e
LSD (d.f. = 99)	245	275.8	170.2	373.1	118.4	229.3	165.4	319.6	4.33

Table 4: Average concentration (μg g^{-1}) with the associated least significant difference (LSD, d.f. = 14) of heavy metals in leaves (n=3) before (t_1) and after (t_2) the roadside exposure and percentage of increase with respect to t_1.

Species	Pb (μg g^{-1})			Cu (μg g^{-1})			Zn (μg g^{-1})		
	t_1	t_2	% increase	t_1	t_2	% increase	t_1	t_2	% increase
P. ×hispanica	0.13	0.18	27.9	7.00	9.54	26.6	12.46	14.7	15.40
T. cordata	0.14	0.18	22.3	10.96	13.78	20.4	12.83	14.28	10.15
Q. cerris	0.23	0.3	21.8	9.47	11.45	17.4	10.85	12.3	13.30
Q. ilex	0.33	0.37	12.2	10.5	12.59	16.5	15.29	16.84	9.20
O. europaea	0.38	0.39	5.8	7.42	8.55	12.9	13.02	14.75	11.73
LSD	0.049	0.058		0.655	0.656		0.627	0.87	