

# Leaf trapping and retention of particles by holm oak and other common tree species in Mediterranean urban environments

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1	Leaf trapping and retention of particles by holm oak and other common tree species
2	in Mediterranean urban environments
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#### 16 Abstract

17

18 Holm oak (*Quercus ilex*), a widespread urban street tree in the Mediterranean region, is widely 19 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 20 21 for particles' capture and retention between Q. ilex and other common Mediterranean urban 22 trees: Quercus cerris, Platanus × hispanica, Tilia cordata and Olea europaea. All applied 23 methods were effective in quantifying particulate capture and retention, although not univocal 24 in ranking species performances. Distinctive morphological features of leaves led to 25 differences in species' ability to trap and retain particles of different size classes and to 26 accumulate metals after exposure to traffic in an urban street. Overall, P.  $\times$  hispanica and T. 27 cordata showed the largest capture potential per unit leaf area for most model particles (Na<sup>+</sup> and powder particles), and street-level Cu and Pb, while Q. ilex acted intermediatelly. After 28 29 wash-off experiments, P. × hispanica leaves had the greatest retention capacity among the 30 tested species and O. europaea the lowest. We concluded that the Platanus planting could be 31 considered in Mediterranean urban environments due to its efficiency in accumulating and 32 retaining airborne particulates; however, with atmospheric pollution being typically higher in 33 winter, the evergreen Q. ilex represents a better year-round choice to mitigate the impact of 34 airborne particulate pollutants.

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

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# 41 Highlights

42 -London plane and lime tree leaves captured most Na<sup>+</sup> aerosol and powder particles per 43 unit leaf area. London plane leaves showed the largest metals' (Pb, Zn, Cu) capture potential, near an 44 \_ 45 urban street. London plane leaves also showed greatest capacity for particle retention after wash off. 46 \_ - In a year-round scenario, Holm oak likely has the highest potential for PM removal due 47 to its evergreen nature. 48 49 50

#### 51 **1. INTRODUCTION**

52

53 Urban population is increasing worldwide and a further rise in urbanisation is predicted 54 (Buhaug and Urdal, 2013). One of the main implications of urbanization is air pollution which 55 is associated with several health outcomes for urban residents, including respiratory and 56 cardiovascular illness, neurological disorders and cancers (e.g. Pope and Dockery, 2006; HEI, 57 2010). In many urban environments the airborne particulate matter (PM) affects more people 58 than any other atmospheric pollutant and no threshold PM concentration has been identified 59 below which no damage to health is observed (WHO, 2014). It has been estimated that PM 60 causes 3.7 million premature deaths annually worldwide and more than 450,000 in Europe 61 (WHO, 2014). Particulate matter from natural (sea salt, soil dust, volcanic ash, forest fires, 62 pollen) or anthropogenic sources (fuel combustion in thermal power generation, traffic, 63 incineration and domestic heating for households) is directly emitted to the atmosphere 64 (primary) or is formed in air as secondary inorganic or organic aerosols from precursor gases such as SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, and volatile organic compounds. Therefore, the urban PM is a complex 65 66 mixture of different phases, with different chemical composition and size. Particles with an 67 aerodynamic diameter  $< 10 \,\mu\text{m}$  (PM<sub>10</sub>) can enter the human airways, particles  $< 2.5 \,\mu\text{m}$  (PM<sub>2.5</sub>) 68 can reach pulmonary air sacs (Baeza-Squiban et al., 1999) and those <0.1 µm enter the blood 69 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).

77 Particles can be removed from the atmosphere by various deposition mechanisms (NEGTAP, 78 2001), with dry deposition being the main pathway, especially in areas with scarce atmospheric 79 precipitation such as the Mediterranean region. Vegetation has a pivotal role in the removal of 80 the atmospheric particulate in terrestrial ecosystems. Dry deposition processes and the particle 81 interception by trees are affected by many factors such the canopy characteristics, wind speed, 82 temperature, particle size, gas solubility as well as leaf pubescence, size and morphology 83 (Beckett et al., 2000; Freer-Smith et al., 2005; Hofman et al., 2014; Weber et al., 2014). Most 84 particles adsorbed on leaves and other plant surfaces are often re-suspended to the atmosphere,

85 washed off by rain, or dropped to the ground with leaf and twig fall. Although it is well-known 86 that the temporary retention of particles by urban trees can reduce atmospheric PM 87 concentrations (e.g. Beckett et al., 2000, Fowler et al., 2004; Novak et al., 2006) the 88 effectiveness of street trees or vertical gardens as a long-term alternative to other measures 89 such as the wet cleaning of streets is still debated (Litschke and Kuttler, 2008). Some previous 90 quantitative estimates of PM<sub>10</sub> reduction by urban vegetation on the city-scale suggested a small 91 effect (often < 1%; e.g. Novak et al., 2006; Escobedo and Nowak, 2009; Tallis et al., 2011). 92 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 93 atmospheric particle concentration) of about 1 cm s<sup>-1</sup>, whereas *in-situ* measurements indicate 94 considerably higher values and literature data for PM<sub>10</sub> deposition velocities to vegetation vary 95 from ~ 0.01 to ~10 cm s<sup>-1</sup>. This variability is due to particle characteristics, meteorological 96 97 conditions as well as to tree species differences in canopy architecture, leaf morphology and 98 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 resuspension 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).

106 In order to contribute to the selection and maintenance of tree species with a higher 107 deposition velocity for an efficient PM interception in Italian cities we compared the particle 108 capture and retention capacity by leaves from a popular and prevalent tree species in Italian 109 urban and roadside environments - Quercus ilex L., to that of possible alternatives: Quercus 110 cerris L., Platanus × hispanica Münch., Tilia cordata Mill., and Olea europaea L. In 111 Mediterranean regions, the evergreen holm oak (Q. ilex) has a wide natural distribution and in 112 Italy it has been used since the sixteenth century in the landscaping of urban and rural parks 113 and gardens. Holm oak has a large canopy, as wells as Leaf Area Index (LAI) typically higher 114 than that of other broad-leaf species (Sgrigna et al., 2015); its leaves have a hair cover and thick 115 waxy cuticles. Because of these leaf properties, which enhance the scavenging and retention of 116 airborne particles and the incorporation of lipophilic organic contaminants, holm oak leaves 117 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 118

119 a quantitative analysis of PM fractions on Q. ilex leaves collected (three times in a year) in an 120 urban environment, Sgrigna et al. (2015) found a mean surface PM deposition of 20.6 µg cm 121  $^{2}$ , a value in the same range of that reported for other urban tree species by Dzierżanowski et 122 al. (2011). Having in mind the need to diversify planting in order to increase the resilience of 123 urban trees and decrease susceptibility to pests and diseases (Laćan and McBride, 2008), in our 124 study the leaf particle interception and retention by Q. ilex were compared with those of other 125 urban tree species to identify possible alternative/complementary trees as PM mitigating tools 126 in Mediterranean urban environments. To evaluate if a cheap and accessible method can 127 produce reliable estimates of tree leaf potential for PM interception, NaCl aerosol and talcum 128 powder were blown onto the leaves in a simple wind tunnel. The results of these laboratory 129 experiments were compared with those from metal particle accumulation in leaves exposed to 130 traffic in an urban street. We chose three metals (Pb, Cu and Zn) routinely associated with 131 anthropogenic pollution sources (Espinosa et al., 2002, Wang 2006) as indicators of street-level 132 pollution; their concentrations are reported in numerous studies (e.g. Davis et al., 2001, 133 Lindgren, 1996), so this should enable baseline comparisons. The leaf particle retention 134 capability in the five tree species was also evaluated by simulating a rainfall. Thus, this work 135 attempted to evaluate the agreement among different laboratory experiments and to compare 136 the behavior of leaves from five selected tree species in terms of particle capture and retention, 137 in the laboratory and the field.

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## 2. MATERIALS AND METHODS

140 **2.1.Plant material** 

The main leaf characteristics of the five tree species common in Mediterranean urban 141 142 areas are summarized in Table 1. *Platanus × hispanica* (London plane) has relatively large, stiff 143 leaves coated with fine, firm hairs (during springtime); those of T. cordata (lime tree) are also 144 large but mostly hairless, except for small tufts of hair in the leaf vein axils (Hölscher, 2003). 145 Both Q. ilex (holm oak) and Q. cerris (Turkey oak) leaves have a water-repellent surface 146 mainly due to the thick epicuticular waxy layer. Quercus ilex is also characterized by stellate 147 trichomes on the surface (Quero et al., 2006). Olea europea (olive) has small silvery-green 148 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.*  $\times$ *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).
- 157

#### **2.2 Laboratory wind tunnel experiments**

#### 159 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.

167 Preliminary experiments were performed with Petri plates, glass slides and artificial 168 leaves held by a custom made rigid mesh support, to establish optimal experimental conditions 169 (i.e. length of application time, amount of NaCl solution and powder, and the distance between 170 leaves and the fan). Artificial leaves were constructed to mimic the average shape and size of 171 the five different tree species, tracing on paper three real leaves with a shape/size representing 172 the average for every species and then laminating them. Variations in weight of Petri plates, 173 slides and artificial leaves (before and after particle application; 10 replicates for each 174 treatment) were determined with a precision balance. Preliminary tests using 30 ml NaCl 175 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  $6.75 \text{ m s}^{-1}$  gave the most reproducible results. 176

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

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#### 180 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 °C. Dry leaf samples were pooled in groups of 2-3 leaves to produce 6-9 replicates per species; leaves were manually digested with concentrated HNO<sub>3</sub> 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  $\mu g g^{-1} d.w.$  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

ground and homogenized using a mortar and pestle. About 500 mg of each sample were

- 195 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
- 199 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<sup>+</sup> 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:
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Cp = N x (leaf weight/LA)

where 'N' was the difference in leaf Na<sup>+</sup> concentration before and after the experiment (expressed in mg g<sup>-1</sup>) and 'D' was the leaf 'density' (obtained as a ratio of leaf weight and leaf

 $R = N_r/LA$ 

- 210 <del>area)</del>.
- 211 The Na<sup>+</sup> wash-off (R) was calculated using the equation:
- 212
- 213 where ' $N_r$ ' was the Na<sup>+</sup> concentration in the runoff (mg l<sup>-1</sup>) and LA was leaf area from which
- runoff was collected.
- 215
- 216 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  $(1 \text{ cm}^2)$  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  $\mu$ m<sup>2</sup> area per leaf and results were reported as particle number mm<sup>-2</sup>. 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.
- 229

# 230 2.3 Field experiment

231 Three branches with similar length and leaf age were excised from each of the Q. ilex, Q. cerris, 232 *P.* ×*hispanica*, *T. cordata* and *O. europaea* trees in the Botanical Garden of Siena (a green park 233 with no adjacent traffic or other sources of airborne metals), and carefully washed with distilled 234 water. Three subsamples of leaves from each tree species were analyzed for Cu, Zn and Pb 235 concentrations to assess the metal concentrations before the exposure (for details on samples 236 preparation, chemical digestion and analytical determination see Fantozzi et al., 2013). On 2 237 July 2013 the branches, inserted in 10 ml plastic flasks with water, which was changed every 238 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<sup>-1</sup> (ARPAT, 2011) (Figure 239 240 3). During the 21 days exposure there was no atmospheric precipitation. All exposed leaves on 241 each branch were pooled and the 3 composite samples for each tree species were analyzed for 242 total Cu, Zn and Pb concentrations.

243

#### 244 **2.4 Data analysis**

Data were analyzed using GenStat (11<sup>th</sup> 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).

- 250
- 251 **3. RESULTS**
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# 253 **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<sup>+</sup> than smaller leaves (e.g. 1.97 mg g<sup>-1</sup> vs 0.36 mg g<sup>-1</sup> for Tilia compared to *Olea*, respectively); *Tilia* was additionally most efficient in Na<sup>+</sup> capture per unit leaf area (0.015 mg cm<sup>-2</sup>), followed by *Q. cerris* and *P.* ×*hispanica* (0.009 and 0.008 mg cm<sup>-2</sup>). *P.* ×*hispanica* and *T. cordata* leaves also showed significantly less (p < 0.01) Na<sup>+</sup> wash-off (Table 2). The Na<sup>+</sup> 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).

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# 3.1.2 Capture and retention of powder particulate

264 Table 3 summarizes the results of the powdering experiment with talcum and the 265 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 5-10 266 267 µm range (Table 3). Leaves from the two oak species showed a lower capture efficiency for 268 the smaller particles ( $< 5 \mu m$ ) and a significantly higher (p < 0.01) capture efficiency for coarser 269 particles (> 10  $\mu$ m) than the other three species. The wash-off treatment removed less than 10% 270 of the total number of particles adsorbed on P. × hispanica leaves and about 31, 48, and 64 % 271 of those adsorbed on *Q. ilex*, *T. cordata*, and *Q. cerris*, respectively. However, under the 272 adopted experimental conditions, about 60% of the finest particles ( $< 5 \mu m$ ) were retained by 273 Q. ilex leaves and about 42 % in those of London plane, lime tree and Turkey oak. The olive 274 leaves showed a minimal capacity to retain adsorbed particles (only <13%) (Table 3).

275

#### 276 **3.2 Field experiment**

277 Average concentrations (µg/g) of Cu, Pb and Zn in the leaves exposed for 21 days to the street-278 level polution in Siena varied between the plant species, and between the metals (Table 4). Lead (Pb) concentrations were no higher than 0.40  $\mu$ g g<sup>-1</sup>, but Cu and Zn up to 13-16  $\mu$ g g<sup>-1</sup> 279 after 3 weeks of exposure to streel-level traffic in dry summer weather. In terms of leaf-level 280 281 capture, for Pb for example, concentration increase after exposure ranged from 5.8 % in O. 282 europea to 27.9 % in P. ×hispanica. For other metals, this range of increase in metal 283 concentration between different species was smaller: e.g. for Cu it was between 12.9% (O. 284 europea) to 26.6% (P. ×hispanica) and even smaller for Zn (9.20% in Q. ilex to 15.4% for P. 285 ×hispanica). P. ×hispanica showed a greatest increase in concentrations of metals after the 286 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).

289

#### **4. DISCUSSION**

291

292 Previous studies indicate that PM interception by trees is often (although not exclusively, see 293 Hofman et al., 2014) affected by canopy architecture; thus conifers, in spite of the low unit 294 needle-leaf area, usually show the highest capture efficiency (e.g. Beckett et al., 2000; Freer-295 Smith et al., 2004; Hwang et al., 2011; Räsänen et al., 2013). All the species considered in this 296 study had leaves capable to distinctively collect airborne particulate; however, the spatial 297 structure of branches and twigs of different species and the lack of foliage during the winter in 298 chosen deciduous species would decrease their capacity for PM trapping on a year-round basis. 299 The administration of NaCl aerosol, with an approach previously used by Beckett et al. (2000) 300 and Räsänen et al. (2013), suggested a much higher capture potential (Cp) and a much lower 301 Na<sup>+</sup> wash-off in *Tilia* and *P*.  $\times$  *hispanica* leaves than in the other species (Table 2). The leaf 302 wettability affects the capture of aerosols (Freer-Smith et al., 2004) and some features of Q. 303 ilex, Q. cerris and O. europaea leaves such as their sclerophylly, superficial roughness, 304 presence of trichomes, convex epidermal cells and wax crystals can reduce the contact area 305 between water and the leaf surface (Kardel et al., 2012) and consequently, the adsorption the 306 Na<sup>+</sup> aerosol.

307 The *Tilia* and *P.* ×*hispanica* leaves, together with those of *O. europaea*, captured the highest 308 number of talcum particles, especially those  $<10 \mu m$ , whereas those with a diameter  $>10 \mu m$ 309 were mainly accumulated by oak leaves. After the wash-off treatment P. ×hispanica retained 310 almost 90% of total particles, while O. europaea retained just 13% (dropping to only 5% in the 311 <5 µm particle size). Small circumference-to-area ratio in olive might be a reason for the low 312 capacity for particles retention (Freer-Smith et al., 2005). Holm oak leaves retained about 68% 313 of total adsorbed particles, including those  $<5 \mu m$ . In agreement with the results of earlier 314 studies (Freer-Smith et al., 1997; Lindberg and Lovett, 1992) indicating that the median 315 diameter of particles collected by oak tree species would be around 9 µm, in our experiment 316 the leaves of Q. cerris and Q. ilex also retained fewer particles in  $<5 \mu m$  range than the other 317 tree species. Carpenter et al. (2005) reported that *Platanus* and *Tilia* leaves can collect a very 318 variable range of particle sizes, and in agreement with another study (Jouraeva et al., 2002) our 319 results indicate that *Tilia* leaves are particularly efficient in the capture of <10 µm particles.

320 The tree species we studied ranked in the order: P.  $\times$  hispanica > T. cordata > Q. ilex > Q. cerris > O. europaea for the powder retention and in the order T. cordata > P.  $\times$  hispanica = 321 322 Q. cerris > Q. ilex = Q. europaea for the NaCl aerosol capture potential. Differences in ranking 323 are likely due to leaf size differences between species (which affect capture even when size 324 differences are accounted for, at a 'unit' level - at which we expressed our capture capacities -325 due to a change in turbulences, Beckett et al., 2000) and features including a smooth or 326 wrinkled surface, the presence of micro-roughness, hairs, veins or trichomes (e.g. Beckett et 327 al. 2000; Liu et al., 2012; Speak et al., 2012) and how they would affect the interception of 328 (Na<sup>+</sup>) aerosols vs powder. Thoennessen (2002) for instance, investigated the distribution of 329 pollutants on leaves along a street with high traffic volume and distinguished between plants 330 with very rough surfaces and higher pollutant deposition and those (self-cleaners) with smooth 331 surfaces which reduce the particles deposition and favour their removal by precipitation and 332 wind. Among leaf types in this study the relatively smaller leaf size, the sclerophylly (i.e. the 333 reduced wettability) and the smoother surface are probably the main factors affecting the much 334 lower retention of Na<sup>+</sup> and powder particles on olive and oaks.

335 In all tree species exposed to high-medium traffic intensities over a 3-week period at the street 336 level, there was a statistically significant increase of average leaf Cu, Zn and Pb concentrations 337 (Table 4). While there was no atmospheric precipitation and consequently wash-off of adsorbed 338 particles during the exposure period, the results corroborated the lower capability of O. 339 europaea and Q. cerris leaves to adsorb airborne particles. In our laboratory experiments Q. 340 *cerris* leaves accumulated the minimum number of total particles; in the field it generally 341 showed lowest particles concentration increase. Both our talcum powder experiment and other 342 studies (Freer-Smith et al., 1997; Tomašević et al. 2008) showed that some oak species mainly 343 capture larger particles.

344 Differences in particulate trapping efficiency have been widely studied in a number of tree 345 species (e.g. Beckett et al. 2000; Freer-Smith et al., 2005; Dzierzanowski et al., 2011). 346 Additional species considered in this study, showed a higher Na<sup>+</sup>, talcum powder, Pb and Cu 347 capture efficiency in London plane and lime tree leaves. However, after the powdering the Q. 348 ilex leaves showed a capacity to retain a larger proportion of fine adsorbed particles, compared 349 with London plane and lime. Additionally, holm oak has a significant practical advantage in 350 being an evergreen species with high Leaf Area Index (i.e. a foliar density which should 351 enhance air turbulence around leaves and the PM deposition; Sgrigna et al. 2015). Also, holm 352 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.

355

# 356 CONCLUSIONS

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358 Comparisons of the leaf-level capture of aerosol and particles among five species of broadleaf 359 trees which are common in many Mediterranean urban environments showed that P. 360 ×hispanica and T. cordata leaves intercepted and retained NaCl aerosol and talcum particles 361 more efficiently than O. europea, Q. cerris, and Q. ilex leaves. In agreement with the results 362 of previous surveys with other tree species, the leaf behaviour seems a species-specific process 363 depending above all on leaf surface morphology and wettability. In general, Q. cerris and 364 especially O. europea leaves showed the weakest performances, while after the wash-off, Q. 365 *ilex* leaves retained high proportion of fine intercepted particles. Thus, although London plane 366 and lime tree leaves generally fared the best and these species should be considered to decrease 367 the impact of airborne particles in urban environments, due to its evergreen nature, foliage 368 distribution and density which is maintained in all seasons, the holm oak probably, has a greater 369 potential for a year-round air pollutant sequestration in Mediterranean urban environments.

370

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372

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378

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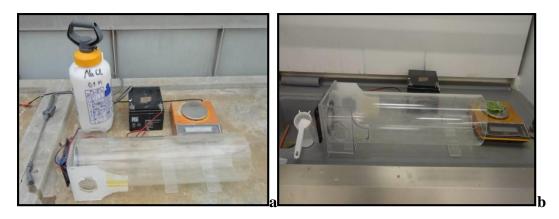
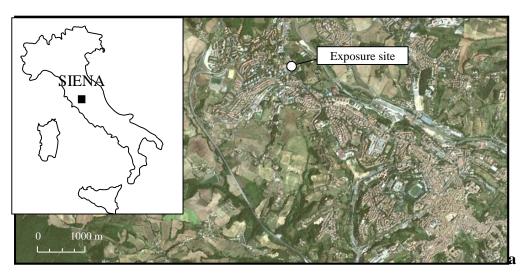


Figure 1: Wind-water tunnel (open-circuit type) used in the experiments: a. with the spray dispenser,
b. with the talcum powder.



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*.



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

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

537

Tree species	Leaf properties					
	fall/retention	Hairs	waxes	size (cm <sup>2</sup> )*		
Platanus ×hispanica		X7	C	77.1.4.0		
(London plane)	Deciduous	Yes	Scarce	77.1±4.8		
Tilia cordata	Derite	C	G	12 5 1 2		
(Lime tree)	Deciduous	Sparse	Scarce	42.5±4.3		
Quercus ilex	<b>F</b>	C	Duranal	14.2.0.0		
(Holm oak)	Evergreen	Sparse	Pronounced	14.3±0.9		
Quercus cerris	Desiduana	N	Duenerunged	24.1.2.1		
(Turkey oak)	Deciduous	No	Pronounced	24.1±2.1		
Olea europaea	<b>F</b>	C	Duenerunged	80.02		
(Olive tree)	Evergreen	Sparse	Pronounced	8.0±0.3		

8 \*Average leaf size of experimental leaves

538 539 540

Table 2. Leaf Na<sup>+</sup> concentrations after aerosol application (expressed in mg g<sup>-1</sup> and mg cm<sup>-2</sup>) and Na<sup>+</sup> concentrations in the runoff (mg l<sup>-1</sup> cm<sup>-2</sup>) 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.

545 546

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

Table 3: Powder particle retention (mean number of particles  $mm^{-2}\pm$  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.

552

	after powdering (num/mm <sup>2</sup> )				after wash-off (num/mm <sup>2</sup> )				
	<5 µm	5-10 µm	$> 10 \ \mu m$	tot	<5 µm	5-10 µm	$> 10 \ \mu m$	tot	% change
P.hispanica	$2688\pm65~b$	$3779\pm80\ b$	$1673\pm49~c$	$8140\ \pm 65\ b$	$1121 \pm 46$ c	3613 ± 66 a	$2757\pm96~a$	7491 ± 69 a	93.6±2.0 a
T.cordata	$3007\pm556~a$	$5741\pm600~a$	$787\pm242~e$	$9535\ \pm 466\ a$	$378 \pm 189 \ b$	$3461\pm 648\ b$	$1052\pm245~c$	$4891\ \pm 361\ b$	52.2±2.3 c
Q.cerris	$378 \pm 172 \; d$	$2014\pm217~c$	2537 ± 273 a	$4928\ \pm 220\ d$	$159\pm141$ a	$742\pm153~d$	$856\pm111~d$	$1757\ \pm 135\ c$	35.8±1.0 d
Q.ilex	$795\pm234~c$	$3891\pm284\ b$	$2243\pm262~b$	$6929\ \pm 260\ c$	$482\pm198\ b$	$2584\pm223~c$	$1675\pm176b$	$4741\ \pm 199\ b$	68.6±1.1 b
O.europaea	$3007\pm471~a$	$5483\pm595~a$	$1287\pm309~d$	$9777 \hspace{.1in} \pm 458 \hspace{.1in} a$	151 ± 72 a	$507 \pm 103 \text{ e}$	$575\pm105~\text{e}$	$1233\ \pm93\ d$	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

553

	Pb (µg g <sup>-1</sup> )			Cu (µg g <sup>-1</sup> )			Zn (µg g <sup>-1</sup> )		
Species	$t_1$	$t_2$	% 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	

Table 4: Average concentration ( $\mu g g^{-1}$ ) with the associated least significant difference (LSD, d.f. = 14) of heavy metals in leaves (n=3) before (t<sub>1</sub>) and after (t<sub>2</sub>) the roadside exposure and percentage of increase with respect to t<sub>1</sub>.