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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**

3
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15

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.
20 By using lab- and field-based experimental approaches, we compared the leaf-level capacity
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. ×hispanica* and *T.*
27 *cordata* showed the largest capture potential per unit leaf area for most model particles (Na⁺
28 and powder particles), and street-level Cu and Pb, while *Q. ilex* acted intermediately. After
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.

35

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

38

39

40

41 **Highlights**

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

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
65 such as SO₂, NO_x, NH₃, and volatile organic compounds. Therefore, the urban PM is a complex
66 mixture of different phases, with different chemical composition and size. Particles with an
67 aerodynamic diameter < 10 µm (PM₁₀) can enter the human airways, particles <2.5 µm (PM_{2.5})
68 can reach pulmonary air sacs (Baeza-Squiban et al., 1999) and those <0.1 µm enter the blood
69 circulation system (EEA, 2014).

70 In cities, the traffic and especially diesel-fuelled vehicles are an important source -close
71 to the ground - of PM-bearing metals and particulate-bound polycyclic aromatic hydrocarbons which
72 have been linked with adverse health effects (e.g. HEI, 2010). Non-exhaust emissions (tyre,
73 brake and road surface wear, corrosion and dust re-suspension) from road traffic are about 50
74 % of exhaust emissions of primary PM₁₀ and about 22% of the exhaust emissions of primary
75 PM_{2.5} (Hak et al., 2009). Therefore, even with zero tailpipe emissions, the traffic will continue
76 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 PM₁₀ 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
93 deposition velocity (i.e. the quotient of the particles' flow rate towards the leaf surface and the
94 atmospheric particle concentration) of about 1 cm s⁻¹, whereas *in-situ* measurements indicate
95 considerably higher values and literature data for PM₁₀ deposition velocities to vegetation vary
96 from ~ 0.01 to ~10 cm s⁻¹. This variability is due to particle characteristics, meteorological
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).

99 Modelling, as well as a number of experimental field and laboratory approaches, have
100 been used to evaluate the PM interception by leaves from a number of plant species (e.g.
101 Beckett et al., 2000; Sæbø et al., 2012; Räsänen et al., 2013). It is known that leaf morphology
102 and wettability play an important role in the interception of airborne particles and in their re-
103 suspension to the atmosphere (e.g. McPherson et al., 1994). However, limited information is
104 available about the wash-off by rain of adsorbed particles from leaves of different tree species
105 (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 well 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.
118 Monaci et al., 2000; Gratani et al., 2008; Fantozzi et al., 2013; Ugolini et al., 2014). Through

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 $\mu\text{g cm}^{-2}$,
121 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.

138

139 **2. MATERIALS AND METHODS**

140 **2.1.Plant material**

141 The main leaf characteristics of the five tree species common in Mediterranean urban
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).

149 In all experiments, young fully-expanded leaves of the current year's growth were used.
150 Wind-tunnel and laboratory experiments were carried out in Summer 2012 at the University of
151 Reading (UK) (see section 2.2) and leaves were collected from the 3-year-old trees maintained
152 in ventilated glasshouses (*O. europaea*, *P. ×hispanica* and *Q. ilex*), or from nearby field-grown

153 mature trees (*Q. cerris* and *T. cordata*); leaves from 2-year-old sections of the branches were
154 used in all experiments. During the Summer 2013, short branches (from 2-year-old wood), of
155 all tree species were excised from mature trees from the Siena Botanical Garden and were
156 exposed to traffic in an urban street (see section 2.3).

157

158 **2.2 Laboratory wind tunnel experiments**

159 *2.2.1 Method development*

160 The wind tunnel used in the experiments to distribute the particles to the leaves was an
161 open-circuit type (Figure 1), 50 cm long and 15 cm in diameter. Particles were generated from
162 a 0.1 M NaCl solution with a pressure sprayer (nozzle outlet diameter = 1 mm) or by a powder-
163 dispenser sieve containing fine powder (Johnson's powder, Johnson & Johnson, New Jersey,
164 USA). Particles were dispensed in front of a splash-proof DC fan (IP54 Ebm-papst,
165 Bachmühle, Germany) at the entry point to the wind tunnel. Droplet diameters and powder
166 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
176 speed in the tunnel of 6.75 m s^{-1} gave the most reproducible results.

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

179

180 *2.2.2. Capture and retention of NaCl aerosol*

181 The fresh weight and the leaf area (LA) of 40 leaves of each tree species were measured before
182 mounting leaves in a support and placing them into the wind tunnel for the exposure to NaCl
183 aerosol. Additional three leaves per species (sprayed only with distilled water) represented
184 controls. After aerosol exposure, all leaves were carefully laid out to air dry under a laminar
185 extractor fan and then 20 leaves were oven-dried for 24 h at 70 °C. Dry leaf samples were
186 pooled in groups of 2-3 leaves to produce 6-9 replicates per species; leaves were manually

187 ground and homogenized using a mortar and pestle. About 500 mg of each sample were
188 digested with concentrated HNO₃ at 120 °C for 8 h in a microwave pressurized digestion
189 system. The mineralized samples of exposed and control leaves were analyzed with an atomic
190 absorption spectrophotometer (AAS) and Na concentrations (expressed in μg g⁻¹ d.w. basis)
191 were determined by the method of standard additions. Procedural blanks were below the Na
192 detection limit; the accuracy of digestion and analytical procedures was checked by routine
193 determination of Na concentrations in standard reference materials (SRM No 2711a and 1515)
194 from the National Institute of Standards and Technology (Gaithersburg, USA). The analytical
195 recoveries from the certified values ranged from 86 to 97%.

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

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

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

$$207 \quad C_p = N \times (\text{leaf weight/LA})$$

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

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

$$212 \quad R = N_r / LA$$

213 where 'N_r' was the Na⁺ concentration in the runoff (mg l⁻¹) and LA was leaf area from which
214 runoff was collected.

215

216 2.2.3. Capture and retention of powder particulate

217 A further 40 leaves of each species, whose leaf area and fresh weight were previously
218 determined, were exposed to 5 g of powder at 20 cm from the fan. Leaves were then carefully
219 laid down in order to avoid loss of powder; a piece of leaf tissue (1 cm²) was cut from the
220 centre of the lamina of 20 leaves and fixed onto a microscope slide. Powder particle retention

221 of the adaxial leaf surface (facing the fan) was determined by counting the number of particles
222 using a digital image-analysis system connected to a light microscope. For each tree species,
223 the number of particles on 20 treated leaves was counted in four random squares of 1 μm^2 area
224 per leaf and results were reported as particle number mm^{-2} . Two untreated leaves acting as
225 controls were also analyzed using the same procedure.

226 The other 20 treated leaves were inserted again into the wind tunnel, exposed to distilled water
227 aerosol as detailed above and then analyzed by light microscope to assess the powder retention
228 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
239 1m away from a street in Siena city centre, with 200-1500 vehicles h^{-1} (ARPAT, 2011) (Figure
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**

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

250

251 **3. RESULTS**

252

253 **3.1 Laboratory wind-tunnel experiments**

254 *3.1.1 Capture and retention of NaCl aerosol*

255 Table 2. shows NaCl aerosol capture potential (C_p) by the different tree leaves. Larger leaves
256 like *P. ×hispanica* and *T. cordata* captured more Na^+ than smaller leaves (e.g. 1.97 mg g^{-1} vs
257 0.36 mg g^{-1} for *Tilia* compared to *Olea*, respectively); *Tilia* was additionally most efficient in
258 Na^+ capture per unit leaf area (0.015 mg cm^{-2}), followed by *Q. cerris* and *P. ×hispanica* (0.009
259 and 0.008 mg cm^{-2}). *P. ×hispanica* and *T. cordata* leaves also showed significantly less ($p <$
260 0.01) Na^+ wash-off (Table 2). The Na^+ wash-off was most pronounced in *O. europaea* and
261 intermediate in *Q. ilex* and *Q. cerris* (1.87 , 0.55 and 0.53 , respectively, Table 2).

262

263 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
266 number of powder particles; all species, except *Q. cerris*, captured mostly particles in the 5-10
267 μm range (Table 3). Leaves from the two oak species showed a lower capture efficiency for
268 the smaller particles ($< 5 \mu\text{m}$) and a significantly higher ($p < 0.01$) capture efficiency for coarser
269 particles ($> 10 \mu\text{m}$) 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 ($< 5 \mu\text{m}$) 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 ($\mu\text{g/g}$) of Cu, Pb and Zn in the leaves exposed for 21 days to the street-
278 level pollution in Siena varied between the plant species, and between the metals (Table 4).
279 Lead (Pb) concentrations were no higher than $0.40 \mu\text{g g}^{-1}$, but Cu and Zn up to $13\text{-}16 \mu\text{g g}^{-1}$
280 after 3 weeks of exposure to street-level traffic in dry summer weather. In terms of leaf-level
281 capture, for Pb for example, concentration increase after exposure ranged from 5.8 % in *O.*
282 *europaea* 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 *europaea*) 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

287 concentration increase for Zn: 9 – 10 % for leaf Cu concentration increase after street-level
288 exposure (Table 4).

289

290 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⁺ wash-off in *Tilia* and *P. ×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⁺ 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 µm, whereas those with a diameter >10 µm
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 µm. 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 µm 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. ×hispanica* > *T. cordata* > *Q. ilex* > *Q.*
321 *cerris* > *O. europaea* for the powder retention and in the order *T. cordata* > *P. ×hispanica* =
322 *Q. cerris* > *Q. ilex* = *O. 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⁺) aerosols vs powder. Thoennesen (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⁺ 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⁺, 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

353 experienced in the Mediterranean region (Bussotti et al., 2002), which seems to give advantage
354 to this species over the others as an interceptor of airborne particles in urban areas.

355

356 **CONCLUSIONS**

357

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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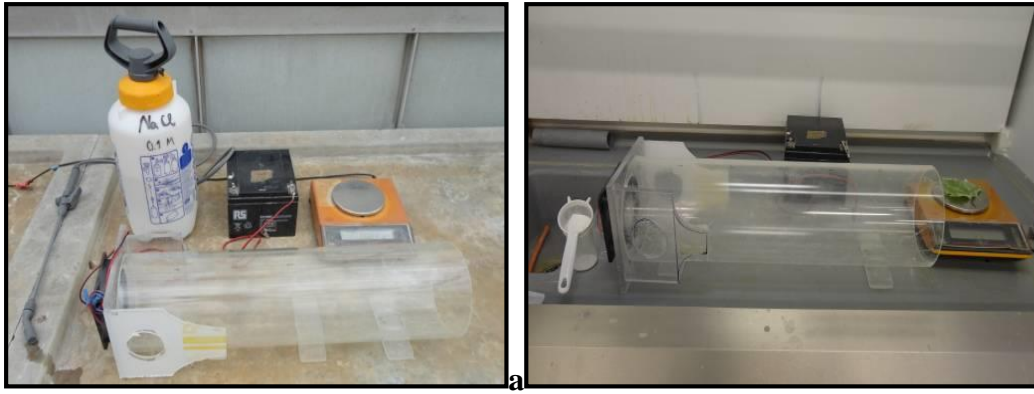
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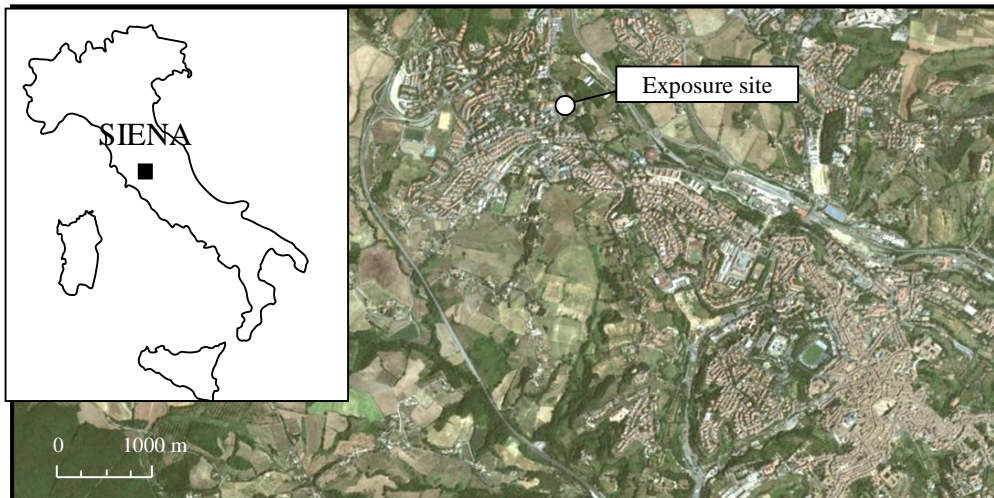
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522 Figure 1: Wind-water tunnel (open-circuit type) used in the experiments: **a.** with the spray dispenser,
 523 **b.** with the talcum powder.
 524



525

526 Figure 2: Leaves of the five tree species inserted in an iron support, from left to right: *T. cordata*, *P.*
 527 *×hispanica*, *Q. cerris*, *Q. ilex*, *O. europaea*.
 528



529

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 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).
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536 Table 1: Tree species used in the experiment and their leaf properties.
537

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

538 *Average leaf size of experimental leaves
539
540

541 Table 2. Leaf Na⁺ concentrations after aerosol application (expressed in mg g⁻¹ and mg cm⁻²)
542 and Na⁺ concentrations in the runoff (mg l⁻¹ cm⁻²) after rinsing with r.o. water. Data are mean
543 of 6-9 replicates per plant species, presented with associated LSD and d.f. Different letters
544 next to the means in each column indicate that means are significantly different.
545
546

Tree species	Leaf Na concentration (mg g ⁻¹)	Leaf Na concentration 'Cp' (mg cm ⁻²)	Runoff water Na concentration (mg l ⁻¹ cm ⁻²)
<i>P. ×hispanica</i>	1.34±0.04 b	0.008±0.0003 b	0.30±0.02 b
<i>T. cordata</i>	1.97±0.14 a	0.015±0.0013 a	0.20±0.02 a
<i>Q. cerris</i>	0.55±0.03 c	0.009±0.0007 b	0.53±0.01 c
<i>Q. ilex</i>	0.53±0.07 c	0.003±0.0004 c	0.55±0.03 c
<i>O. europaea</i>	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

547
548

549 Table 3: Powder particle retention (mean number of particles $\text{mm}^{-2} \pm \text{SEM}$ as well as the associated LSD for each particle class size) and particle
550 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
551 the means in each column indicate that means are significantly different.
552

	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
<i>P.hispanica</i>	2688 \pm 65 b	3779 \pm 80 b	1673 \pm 49 c	8140 \pm 65 b	1121 \pm 46 c	3613 \pm 66 a	2757 \pm 96 a	7491 \pm 69 a	93.6 \pm 2.0 a
<i>T.cordata</i>	3007 \pm 556 a	5741 \pm 600 a	787 \pm 242 e	9535 \pm 466 a	378 \pm 189 b	3461 \pm 648 b	1052 \pm 245 c	4891 \pm 361 b	52.2 \pm 2.3 c
<i>Q.cerris</i>	378 \pm 172 d	2014 \pm 217 c	2537 \pm 273 a	4928 \pm 220 d	159 \pm 141 a	742 \pm 153 d	856 \pm 111 d	1757 \pm 135 c	35.8 \pm 1.0 d
<i>Q.ilex</i>	795 \pm 234 c	3891 \pm 284 b	2243 \pm 262 b	6929 \pm 260 c	482 \pm 198 b	2584 \pm 223 c	1675 \pm 176 b	4741 \pm 199 b	68.6 \pm 1.1 b
<i>O.europaea</i>	3007 \pm 471 a	5483 \pm 595 a	1287 \pm 309 d	9777 \pm 458 a	151 \pm 72 a	507 \pm 103 e	575 \pm 105 e	1233 \pm 93 d	12.4 \pm 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

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Table 4: Average concentration ($\mu\text{g g}^{-1}$) 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 ($\mu\text{g g}^{-1}$)			Cu ($\mu\text{g g}^{-1}$)			Zn ($\mu\text{g g}^{-1}$)		
	t_1	t_2	% increase	t_1	t_2	% increase	t_1	t_2	% increase
<i>P. ×hispanica</i>	0.13	0.18	27.9	7.00	9.54	26.6	12.46	14.7	15.40
<i>T. cordata</i>	0.14	0.18	22.3	10.96	13.78	20.4	12.83	14.28	10.15
<i>Q. cerris</i>	0.23	0.3	21.8	9.47	11.45	17.4	10.85	12.3	13.30
<i>Q. ilex</i>	0.33	0.37	12.2	10.5	12.59	16.5	15.29	16.84	9.20
<i>O. europaea</i>	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	